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Reclamation of Hydrocarbon Contaminated Soils Using Soil Amendments and Native Plant Species

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Abstract: Petroleum hydrocarbons are among the top contaminants of the natural environment with serious concern worldwide due to their effects on soil, water, and surroundings. A two-year field experiment was implemented to evaluate reclamation of hydrocarbon contaminated (diesel fuel, crude oil) soils in central Alberta Canada using amendments (20% city waste compost, ammonium sulphate inorganic fertilizer) and seeding with a native grass mix. Soils amended with compost or compost-fertilizer had the greatest vegetation cover and biomass and lowest hydrocarbon concentrations at the end of the study. Fertilizer treatments had less vegetation cover and higher hydrocarbon concentrations, which were similar to the no amendment treatment. Seeding with native grasses had no effect on hydrocarbon degradation or total canopy cover, although vegetation composition showed some effect. Seeding increased cover of perennial native grasses in all amendment treatments, with greatest cover in compost and compost-fertilizer amended soils. Within two years after reclamation concentrations of F2 (carbon length > C10–C16) and F4 (>C34–C60) hydrocarbons in crude oil contaminated soils were below Canadian guidelines. Overall, compost was an effective amendment for reclamation of diesel fuel and crude oil contaminated soils and seeding was beneficial for reducing cover of non-native forbs. Fertilizer addition to compost may not enhance revegetation and remediation of hydrocarbon contaminated soils.

Keywords: forbs; grass; heavy metals; natural attenuation; petroleum hydrocarbons; remediation; soil amendments

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1. Introduction

The current global economy is heavily reliant on the petroleum industry and poses an environmental risk during processing and storage [1–3]. Despite increased environmental awareness, petroleum hydrocarbon consumption is predicted to increase 106.6 million barrels day^{-1} by the end of 2030 [4]. Canada has 1.6 to 1.7 trillion barrels of oil in bitumen deposits, representing the second largest bitumen deposit in the world and 13% of the global oil reserve [5]. These deposits are concentrated in northern Alberta over 142,000 km^2 or approximately 20% of the area of Alberta [6]. A significant percentage of these petroleum hydrocarbons are transported by pipelines within Canada and continental USA. There are approximately 440,000 km pipelines in Alberta Canada alone [5] and thus spills of petroleum hydrocarbons resulting from mechanical failures, leaks, or human error are occurring frequently and causes significant environmental issues [7–9]. Petroleum hydrocarbons include alkanes, cycloalkanes, polycyclic aromatic hydrocarbons, and many other organic pollutants [3], and some of these hydrocarbons are classified as major environmental pollutants due to their stability and durability in soils [1,3,10]. Dissolution, dispersion, photooxidation, and biodegradation of hydrocarbons can occur naturally but very slowly, with continuing adverse effects in contaminated exposed areas and their surroundings such as high potential risks and hazardous impacts on humans and other living organisms [11]. Therefore, contaminated sites require remediation and reclamation. Environmental regulations in many jurisdictions, including Alberta, require all industries to reclaim disturbed sites to pre-disturbance equivalent land capability [12]. However, contaminated soils are typically low in organic matter and nutrients, and lack desired seed sources, which can limit reclamation success [13,14].

Application of organic and/or inorganic amendments to contaminated soils is a process of biostimulation which can ameliorate poor soil conditions that may be limiting vegetation establishment [15–17] and can accelerate degradation of hydrocarbons by enhancing microbial activities [18]. Organic amendments are typically high in organic matter, have a neutral pH, and contain nutrients in a slow-release form due to slow carbon and nitrogen mineralization [9,19]. Among the most commonly used organic amendments are animal manure, compost, vermicompost, biosolids, sewage sludge, biochar, forest floor material, and peat soil; common inorganic amendments include gypsum, zeolite, pyrite, and fertilizers [20–22]. Soil organic amendments can immobilize contaminants to limit their bioavailability and improve soil quality by balancing pH, adding organic matter, increasing water holding capacity, re-establishing microbial communities, and alleviating compaction [22,23]. Among the organic amendments, compost is the most commonly and widely used in land reclamation, facilitation disturbed or contaminated sites to be remedied, revegetated, and revitalized [20–23]. Fertilizers are widely used inorganic amendments as they supply readily available nutrients over a short time which has a priming effect on microbial activity and plant growth [24,25]. The majority of remediation studies have assessed combined effects of organic and inorganic amendments on biodegradation [9,26,27]; however, there is a need to differentiate individual and combined effects of organic and inorganic amendments while using native plant species as an approach for remediation of petroleum hydrocarbon contaminated soils.

Vegetation can have a positive effect on hydrocarbon degradation or remediation in contaminated soils due to improved aeration, distribution of organic matter throughout the soil profile, and increased microbial activity in the rhizosphere [8,9,28]. To stimulate microbial activity and therefore contaminant degradation, plant species that develop fibrous root systems and establish rapidly are advantageous [20,29,30]. Vegetation on disturbed sites can stabilize the soil surface, reduce erosion, and contribute to nutrient cycling and soil aeration [21] which directly leads to successful reclamation. However, absence of adequate seed sources can limit establishment of desired plant communities on disturbed or contaminated sites. Seeding species that are tolerant of environmental conditions at the disturbed site is essential to improve revegetation success. The majority of remediation and revegetation research on disturbed sites is commonly conducted to test performance of agronomic rather than native species on contaminated soils [8,28,29,31]. Native species are adapted to environmental conditions (drought, nutrients, salt concentrations) of disturbed sites, and therefore may be a suitable option for reclamation and remediation of contaminated soils.

Bioremediation of petroleum contaminated soils has been investigated since the late 1940s, but interest in the field was not widespread. Bioremediation studies using compost, inorganic fertilizer, and/or plants were conducted under controlled greenhouse conditions, with optimized environmental factors [8,9,28]. However, in situ biodegradation under field conditions without combining amendments and native species has been rarely assessed. Thus, there is a need for field research, as ambient soil water content and temperature conditions play a crucial role in microbial activity and bioremediation success [13,14]. Inherent spatial variability of hydrocarbon spills limits experimental control and replication adequacy [32]. Therefore, a manipulated field experiment was conducted over two years using soil amendments and native seed mixes on petroleum hydrocarbon contaminated soils. The main objective of our study was to determine the impact of soil amendments and native plants species on hydrocarbon bioremediation and to test the hypothesis that amendments and native seed mixes can expedite hydrocarbon biodegradation in diesel fuel and crude oil contaminated soils. The outcome of this study can facilitate development of reclamation prescriptions for petroleum hydrocarbons contaminated soils in Alberta and elsewhere.

2. Materials and Methods

2.1. Experimental Design and Treatments

The field experiment was conducted at the City of Edmonton Waste Management Centre (53° 35′ 52″ N and 113° 20′ 0″ W), Alberta, Canada over two years. Mean annual temperature of the study area is 3.1 °C, ranging from −16.6 to 22.2 °C; mean annual precipitation is 466 mm [33]. Temperature conditions and precipitation over the study period were highly similar to the climate normal for the area. The soil was an industrially disturbed black chernozem with sandy loam texture and mean background soil hydrocarbon concentrations <100 mg kg^{-1} (Table A1).

Experimental units were 20 L (28.5 cm diameter, 40 cm depth) plastic pails with an internal drainage system of perforated PVC tube (1.9 cm diameter) and sterilized sand (5 cm depth) to remove leachate. A randomized complete block design was used with three factors (fuel types, amendments, seed mix) within each of four replicate blocks. Treatments were soil contaminated with two fuel types (diesel fuel, light crude oil) and an uncontaminated or control, three soil amendments (compost, fertilizer, compost + fertilizer) with a no amendment and seeded with native grass mix and unseeded. Each replicate was 5 × 5 m in size and contained 36 pails at 75 cm intervals, 25 cm away from the boundary. There was a 5 m buffer zone between replications. All pails were randomly assigned in each replication. Thus, the overall experimental design consisted of 4 replications × 3 contamination treatments (diesel fuel, light crude oil, uncontaminated) × 4 amendments (compost, fertilizer, compost + fertilizer, control or no amendment) × 2 seeding treatments (seeded, unseeded).

Soil was a mixture of approximately 40% topsoil and 60% subsoil (dry soil mass) to represent an admixed disturbed soil (Table A1). Soils were artificially contaminated with diesel fuel and light crude oil to ensure experimental control over the type and uniformity of contamination. Diesel fuel contains predominantly light hydrocarbon fractions, and light crude oil contains a mix of light and heavy fractions. Fuel was applied to the soil treatments at 1% (dry soil mass, equivalent to 10,000 mg kg^{-1} total extractable hydrocarbons) representing a moderate contamination suitable for remediation. The fertilizer treatment was ammonium sulphate (21-0-0-24) at 96 g $pail^{-1}$ (4100 mg kg^{-1}). The compost-fertilizer treatment was 76.8 g $pail^{-1}$ (3280 mg kg^{-1}) of ammonium sulphate fertilizer. Ammonium sulphate application was aimed at optimizing nutrient concentrations for microbial activity to ultimately stimulate biodegradation of hydrocarbons. A C:N ratio of 10:1 was considered ideal for microbial populations [14]; therefore, ammonium sulphate was incorporated into the soil at a rate to achieve a C:N ratio of 10:1. Petroleum hydrocarbon contaminated soils commonly have imbalanced nutrient ratios due to the large input of carbon from the fuel. Immobilization of available nutrients occurs when carbon to nutrient ratios increase, therefore inorganic fertilizers, such as ammonium sulphate, are applied to contaminated soils to supply readily available nutrients to the microbial populations [13,34,35]. Compost as an amendment was mixed with soil at 20% (dry soil mass, equivalent to 10% wet soil volume) in compost and compost-fertilizer treatments. This application rate was selected as it increased the total organic carbon level in the treatment soil to background levels for the area. The compost was collected from the City of Edmonton waste facilities and contained macro and micro nutrients, was neutral pH, and pathogen free (fecal coliforms, *Salmonella*). To maintain equal volumes of substrate (unamended and amended soil) in each pail, fertilizer and no amendment pails contained 23.2 kg of soil (dry mass) and compost and compost-fertilizer pails contained 18.6 kg of soil (dry mass). In this study, compost was used as an amendment to determine its applicability in fuel contaminated soils reclamation, therefore as an amendment its volume or dry mass was not considered during fuel contamination.

Fuel and amendments were incorporated into the soil using a cement mixer (one treatment replicate mixed at a time). Soils were settled into the pails by gently tapping the pail bottom on the ground. Pails were buried in the ground to approximately 35 cm depth to ensure treatments would be exposed to ambient environmental conditions. Pails were seeded with 300 pure live seeds m^{-2} with a native grass seed mix of *Elymus trachycaulus* (Link) Gould ex Shinners (slender wheatgrass), *Leymus innovatus* (Beal) Pilg. (hairy wildrye), *Deschampsia caespitosa* (L.) P. Beauv. (tufted hairgrass), and *Calamagrostis canadensis* (Michx.) P. Beauv. (bluejoint) after soil placement. The surface was lightly raked to a depth of 0.5 cm to enable incorporation and minimize seed loss. Germination tests were conducted prior to seeding to confirm seed viability. Species were selected based on seed availability, fibrous root systems, and relative tolerance to salt, hydrocarbons, and drought.

2.2. Soil Sampling and Analyses

Soil was sampled at the beginning for each treatment replicate to determine initial properties. At the end of the experiment soil was sampled from 0 to 20 cm depths of each pail. Each pail was removed from the ground, tipped then cut in half vertically to expose the center of the soil profile; half the pail was sampled for chemical analyses and the other half used for root biomass collection. Samples were analyzed for total extractable hydrocarbons between C11 and C60 (EPA 3550/8000-GC-FID) using gas chromatography—flame ionization detection, trace metals by inductively coupled plasma—mass spectrometry (EPA 6020), fecal coliforms using the multiple tube fermentation technique (APHA 9221), and Salmonella using cultural isolation and serotyping (APHA 9260B) [36]. Total organic carbon was determined by combustion [37], total kjeldahl nitrogen by titration [38], ammonium by potassium chloride extraction [39], nitrate by colorimetry (APHA 4500) [36], sulphate by the turbidimetric procedure [40], pH, electrical conductivity (EC) by saturation extract meters [41], and bulk density by ASTM D5057 method [42]. The sodium adsorption ratio was calculated from sodium, calcium, and magnesium concentrations [41]. Individual carbon lengths were summed into three groups to correspond with fraction 2 (F2) (>C10 to C16), fraction 3 (F3) (>C16 to C34), and fraction 4 (F4) (>C34) hydrocarbons [43]. Initial materials were analyzed for the above parameters, bulk density, and particle size distribution [44] (excluding compost samples). All samples were stored in coolers (5 °C) immediately after sampling and sent to a commercial laboratory in Edmonton, Alberta for analysis.

Following rainfall events in both years of the study, leachate was pumped from each treatment pail using a peristaltic or manual pump. Leachate volumes were recorded for each treatment pail; however, due to budget constraints no chemical analyses were conducted on the liquid. Removal of leachate from the pails ensured soils were at or below field capacity over the study period and were not exposed to anaerobic conditions.

2.3. Vegetation Assessments

Vegetation was assessed in September in year 1 and August in year 2. Total canopy cover (%) and species composition were determined each time. Individual species were classified as functional groups. Plant biomass was sampled at the end of the experiment in August. Above ground biomass (live vegetation, litter) was clipped from the entire pail area (0.064 m^{-2}) prior to soil sampling. Root biomass was removed from an intact half pail of soil by sieving through a 0.25 cm^2 wire screen and gently shaken, and then soil was sprayed with water to separate soil particles from the root material. To ensure all soil and organic material had been removed, the roots were gently massaged under water until only root material remained. All above and below ground biomass samples were oven dried at 80 °C for 48 h then weighed. Both above and below ground biomass data were expressed on a mass per area basis (g m^{-2}) where total rooting depth was 25 cm.

2.4. Data Analyses

All soil data, vegetation parameters, above and below ground biomass, and hydrocarbons were analyzed using two- and three-way analysis of variance (ANOVA) procedures. The effect of amendments and seeding on revegetation was determined separately for each fuel type using a two-way ANOVA with amendment and seeding as main factors. To determine whether treatment responses differed between uncontaminated and contaminated soils, a three-way ANOVA was conducted with fuel, amendment, and seeding as factors. Since a comparison between diesel and crude oil contaminated soils was not an objective of the study, separate ANOVAs were conducted for uncontaminated versus diesel and crude oil contaminated soils. Interactions between factors were tested and multiple comparisons conducted using the multicomp package v. 1.3-2 when significant differences were found. Paired t-tests were used to determine differences in vegetation cover between year 1 and 2. Prior to analyses, normality and homogeneity of variance were tested by examining residuals versus the fitted plots and normal q–q plots of the models, and no transformations were required. All statistical analyses were conducted using R statistics system version 3.2.5 [45] and a *p*-value of 0.05 was selected to determine significance.

3. Results and Discussion

3.1. Soil Property Responses

Soil properties were affected by amendments more than seeding. Fertilizer and compost-fertilizer treatments had significantly more ammonium than compost and no amendment treatments in contaminated and uncontaminated soils (Table 1). Under aerobic conditions, nitrification converts ammonium to nitrate and released hydrogen ions can lower pH, as found in the fertilizer treatment which was 2 units lower than other treatments. Most studies found vegetation cover and biomass increased following fertilizer application due to the stimulatory effect of inorganic nutrients [46]. The contradictory findings in our study can be linked to the acidic pH of the fertilizer treatment [47]. Fertilizer application with compost did not lower pH (Table 1), which suggests the compost buffered the effects of nitrification [26]. The higher initial concentration of ammonium in the compost-fertilizer treatment (Table 1) could explain the increased vegetation production relative to the compost treatment.

Compost addition to uncontaminated and contaminated soils significantly increased total organic carbon concentrations relative to fertilized and control treatments (Table 1). Total organic carbon in compost treatments were representative of background soils in the black zone relative to the unamended soil [48]. Compost addition also significantly increased total kjeldahl nitrogen and nitrate concentrations in compost and compost-fertilizer treatments relative to the other treatments (Table 1). Since plants uptake nitrogen in the form of nitrate, the positive vegetation growth observed in compost treatments may be partially linked to nutrient availability (Figures 1 and 2). Addition of organic matter to the soil in the form of compost resulted in lower bulk densities in the compost and compost-fertilizer treatments relative to the other treatments in uncontaminated and contaminated soils (Table 1). However, the sandy loam soil used in the study had a low bulk density, therefore the difference in bulk densities between the treatments has limited biological significance [49].

Table 1. Mean (±SE) soil properties at the end of study by soils treatment. Different letters indicate significant differences at $p = 0.05$ among amendments within a fuel type. Com + Fer = Compost + Fertilizer, No Amend = No amendment, TOC = total organic carbon, TKN = total kjeldahl nitrogen, C:N = carbon to nitrogen ratio, EC = electrical conductivity, SAR = sodium adsorption ratio.

Soil Properties	**Uncontaminated Soil**				**Diesel Fuel Soil**				**Crude Oil Soil**			
	No Amend	**Fertilizer**	**Compost**	**Com + Fer**	**No Amend**	**Fertilizer**	**Compost**	**Com + Fer**	**No Amend**	**Fertilizer**	**Compost**	**Com + Fer**
TOC (%)	2.2 (0.3) b	2.2 (0.2) b	6.7 (0.6) a	6.2 (0.4) a	3.3 (0.2) b	3.4 (0.2) b	6.7 (0.7) a	7.0 (0.3) a	2.8 (0.2) b	2.3 (0.1) b	6.0 (0.2) a	6.0 (0.3) a
TKN (%)	0.2 (0.03) b	0.2 (0.01) b	0.5 (0.04) a	0.5 (0.01) a	0.2 (0.01) b	0.3 (0.01) b	0.5 (0.02) a	0.5 (0.02) a	0.2 (0.02) b	0.2 (0.01) b	0.5 (0.01) a	0.4 (0.01) a
C:N	15.5 (1.5) a	11.9 (1) b	13.4 (0.2) b	13.2 (0.6) b	15.2 (0.7) a	12.9 (0.1) b	13.2 (0.6) b	13.5 (0.7) b	19.2 (1.1) a	14.0 (0.5) b	13.8 (0.6) b	14.9 (1.1) b
Ammonium (mg kg^{-1})	3.0 (0.3) b	72.3 (33.5) a	4.5 (0.4) b	6.1 (0.5) b	4.3 (0.7) b	131.6 (49) a	5.0 (0.4) b	6.6 (0.9) b	2.4 (0.2) a	68.3 (26.7) a	3.5 (0.3) a	5.2 (0.6) a
Nitrate (mg kg^{-1})	3.2 (0.5) b	81.5 (18.5) a	3.2 (0.3) b	128.6 (29.9) a	1.8 (0.2) b	55.7 (3.9) a	3.4 (0.4) b	49.6 (18.3) a	1.5 (0.2) b	79.7 (8.1) a	2.2 (0.3) b	4.6 (2.3) b
pH	7.15 (0.05) b	5.11 (0.04) c	7.71 (0.01) a	7.3 (0.04) b	7.01 (0.04) a	5.26 (0.13) b	7.34 (0.06) a	7.4 (0.05) a	7.19 (0.04) b	5.23 (0.03) c	7.58 (0.05) a	7.25 (0.02) b
EC (dS m^{-1})	0.54 (0.03) c	4.36 (0.4) b	4.39 (0.38) b	6.81 (1.01) a	0.48 (0.09) c	3.5 (0.49) b	5.29 (0.51) a	5.78 (0.23) a	0.54 (0.08) b	4.04 (0.12) a	4.15 (0.55) a	4.54 (0.18) a
SAR	0.4 (0.08) c	0.14 (0.04) c	6.75 (0.34) a	4.84 (0.51) b	0.68 (0.09) c	0.2 (0.04) c	6.46 (0.36) a	5.31 (0.48) b	0.69 (0.09) c	0.2 (0.04) c	5.98 (0.64) a	3.69 (0.38) b
Sodium (mg L^{-1})	14.9 (3.1) b	15.6 (4.9) b	565.0 (65.9) a	643.0 (129) a	22.5 (2.7) b	21.1 (6) b	677.0 (81.4) a	644.0 (66.6) a	25.0 (3.9) b	23.8 (3.8) b	548 (76.9) a	418 (43.1) a
Cadmium (mg kg^{-1})	0.5 (0.0) b	0.5 (0.0) b	1.25 (0.10) a	1.16 (0.03) a	0.5 (0.0) b	0.5 (0.0) b	1.29 (0.14) a	1.19 (0.06) a	0.5 (0) b	0.5 (0.0) b	0.89 (0.09) a	1.0 (0.09) a
Copper (mg kg^{-1})	13.6 (1) b	11.8 (0.9) b	86.3 (5.1) a	77.8 (1.9) a	15.4 (1.9) b	14.4 (0.5) b	72.5 (3.7) a	72.6 (4.7) a	11.8 (1.9) b	10.9 (0.7) b	60.9 (4.9) a	64.5 (6.2) a
Lead (mg kg^{-1})	12.4 (0.8) b	10.3 (0.8) b	64.8 (6.2) a	57.02.5) a	15.4 (3.6) b	12.5 (0.7) b	53.9 (2) a	53.8 (1.2) a	11.0 (2.4) b	9.9 (0.5) b	44.6 (4.0) a	49.8 (4.8) a
Zinc (mg kg^{-1})	54.5 (4.1) c	46.3 (2.1) c	270.8 (11.2) a	233.5 (6.2) b	55.8 (2.7) b	54.5 (1.9) b	233.1 (10) a	225.1 (11.3) a	45.8 (5.7) b	49.8 (2.4) b	185.8 (14.8) a	198.0 (18.5) a
Bulk density (mg m^{-3})	1.33 (0.03) a	1.3 (0.02) a	1.23 (0.05) a	1.07 (0.05) b	1.2 (0.04) a	1.15 (0.05) a	1.05 (0.03) b	1.03 (0.03) b	1.29 (0.03) a	1.27 (0.01) a	1.08 (0.03) b	1.09 (0.06) b

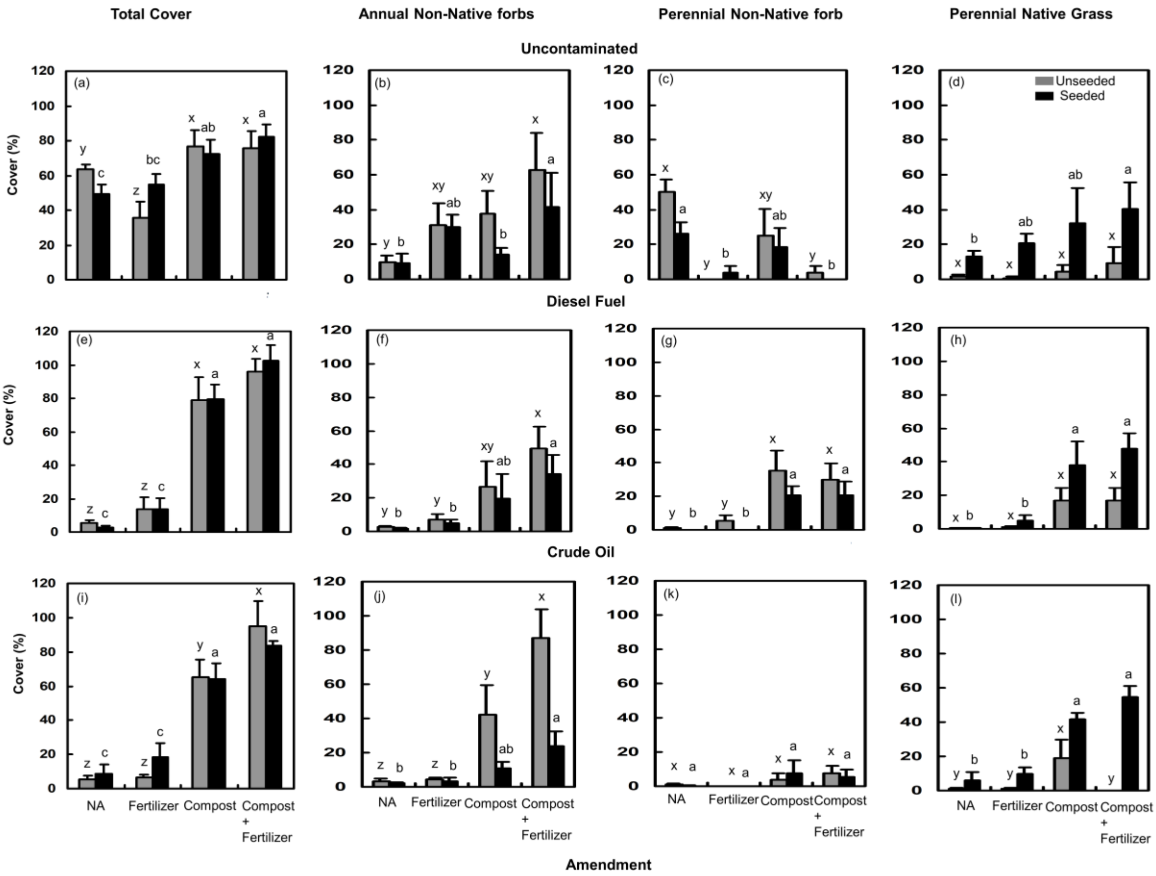

Figure 1. Mean (±SE) vegetation cover percent by group in (**a**–**d**) uncontaminated, (**e**–**h**) diesel fuel, and (**i**–**l**) crude oil contaminated soils. Different letters indicate significant differences at $p = 0.05$. NA = No Amendment.

Due to its source materials, the compost for our study had elevated electrical conductivity, sodium adsorption ratio, sodium, cadmium, copper, lead, and zinc (Table A1), resulting in compost and compost-fertilizer treatments having significantly higher values of these parameters than the no amendment treatment (Table 1), which exceeded CCME guidelines for soil quality [43]. The compost for this study was procured from the City of Edmonton which contained elevated concentrations of salts and certain heavy metals (Cd, Cu, Pb, Zn) due to feedstock materials. Feedstock materials play an important role in compost quality and its chemical constituents. Therefore, before using compost as an amendment, detailed chemical analyses or use of compost that contains known feedstock materials are highly recommended. Vegetation in both compost treatments did not respond negatively to salinity or metals. Metal concentrations before and after the experiment did not differ between amendment treatments, suggesting metals in compost were not bioavailable [9,19]. Kaschl et al. [50] found trace metals in municipal solid waste compost were present in very low percentages (<2.5%) in the bioavailable water extractable fraction. Despite the acidic pH of the fertilizer treatment, there were no significant metal losses (Table 2), which might reflect low concentrations of metals in the fertilizer treatment. Compost and compost-fertilizer treatments lost significantly more sodium than fertilizer or no amendment treatments (Table 2). This could be due to absorption by plants, downward water movement, or leachate. Greater plant cover and biomass in the compost treatment could be linked to greater plant absorption (Figures 1 and 2). Salts in soil systems are very soluble and therefore mobile [51]. As a result, the dominant mechanism for sodium loss in the compost treatments is likely downward water movement or leachate. Thus, consequences on downstream water and soil quality are a potential issue if compost is applied in ground water recharge areas or on lands with shallow water tables.

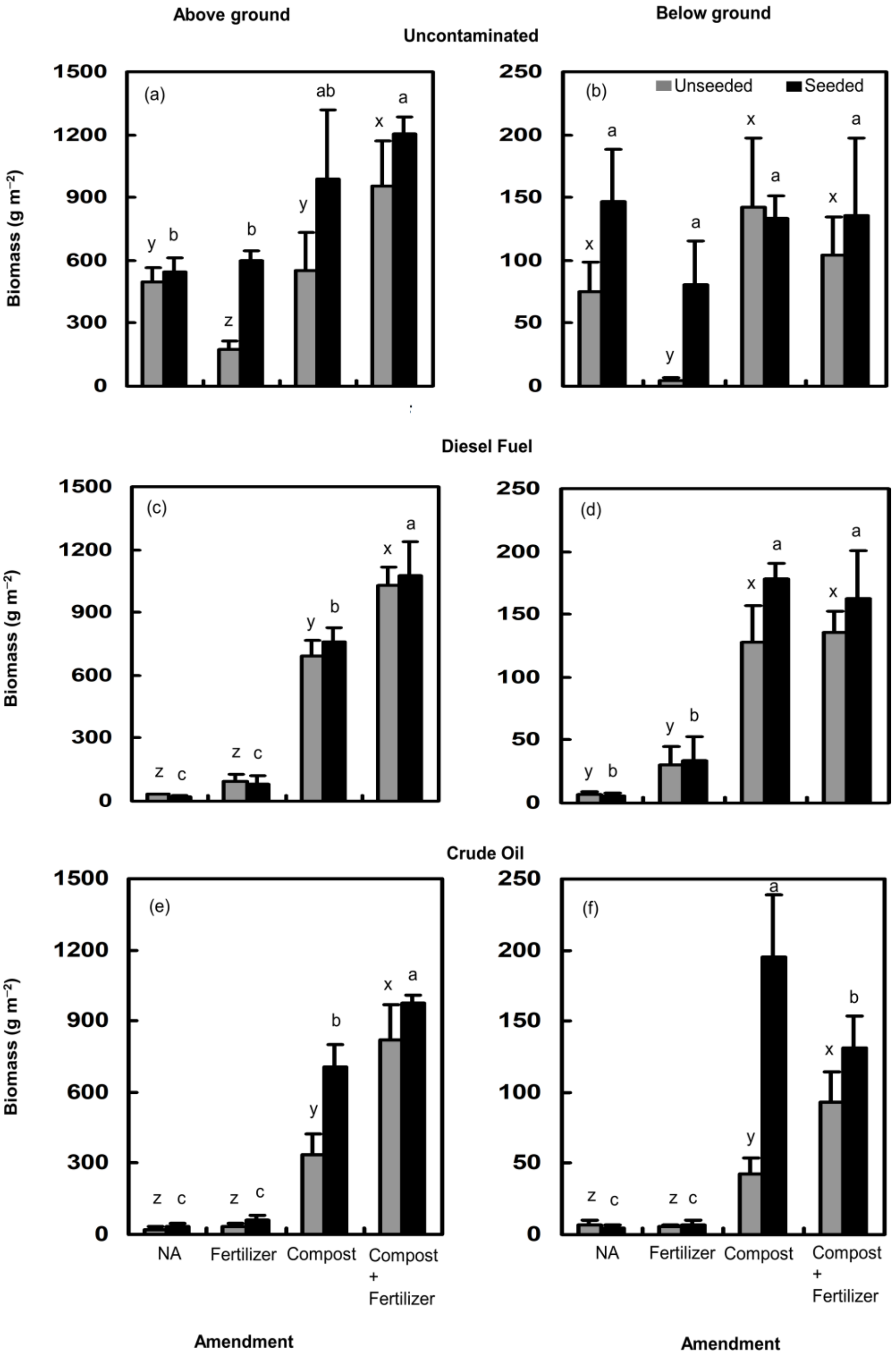

Figure 2. Mean (±SE) above and below ground biomass in (**a**,**b**) uncontaminated, (**c**,**d**) diesel fuel, and (**e**,**f**) crude oil contaminated soils. Different letters indicate significant differences at $p = 0.05$. NA = No Amendment.

Table 2. Mean (±SE) change of soil properties at beginning and end of the experiment by soil treatments. Different letters indicate significant differences at $p = 0.05$ among amendments within a fuel type. Com + Fer = Compost + Fertilizer, No Amend = No amendment, TOC = total organic carbon, TKN = total kjeldahl nitrogen, C:N = carbon to nitrogen ratio, EC = electrical conductivity, SAR = sodium adsorption ratio.

Soil Properties	**Uncontaminated Soil**				**Diesel Fuel Soil**				**Crude Oil Soil**			
	No Amend	**Fertilizer**	**Compost**	**Com + Fer**	**No Amend**	**Fertilizer**	**Compost**	**Com + Fer**	**No Amend**	**Fertilizer**	**Compost**	**Com + Fer**
TOC (%)	0.2 (0.2) a	0.7 (0.1) a	0.7 (0.7) a	−0.1 (0.2) a	−0.5 (0.2) ab	0.1 (0.4) bc	−1.6 (0.5) a	0.8 (0.4) c	−0.7 (0.4) a	−0.7 (0.4) a	−1.3 (0.7) a	−1.2 (0.4) a
TKN (%)	0.0 (0.02) a	00. (0.01) a	0.0 (0.07) a	−0.1 (0.04) a	0.0 (0.01) b	0.0 (0.03) b	−0.1 (0.03) a	−0.1 (0.06) ab	−0.1 (0.02) c	−0.1 (0.02) bc	−0.2 (0.02) ab	−0.2 (0.05) a
C:N	0.1 (0.7) a	4.4 (0.4) b	0.7 (0.4) a	1.4 (0.8) a	−1.0 (1.0) a	1.3 (0.4) ab	0.2 (0.7) ab	2.3 (1.0) b	1.4 (1.0) a	2.9 (0.6) a	1.2 (1.0) a	3.1 (1.2) a
Ammonium (mg kg^{-1})	−3.5 (0.3) c	−739.6 (33) a	−7.8 (3.2) c	−609.6 (1.9) b	0.8 (0.7) d	−684.5 (52) a	−3.2 (1.1) c	−604.6 (1.1) b	−1 (0.2) c	−740.7 (26) a	−2.8 (0.4) c	−601 (0.5) b
Nitrate (mg kg^{-1})	−5.1 (0.3) b	73.9 (18) b	−306.6 (50) a	−232.2 (29) a	−7.2 (0.5) bc	46.4 (3.8) c	−720.1 (29) a	−275.7 (36) b	−11.9 (7.5) b	76.1 (7.9) c	−336.6 (13) a	−282.7 (8.7) a
pH	0.5 (0) c	−1.51 (0.7) a	0.56 (0.06) c	0.07 (0.04) b	0.34 (0.09) b	−1.39 (0.12) a	0.09 (0.06) b	0.25 (0.02) b	0.26 (0.04) c	−1.7 (0.06) a	0.4 (0.04) d	−0.02 (0.08) b
EC (dS m^{-1})	0.09 (0.03) c	3.88 (0.37) d	−5.63 (0.37) a	−2.97 (1.17) b	0.03 (0.18) c	2.77 (0.63) d	−6.16 (0.87) a	−3.67 (0.21) b	0.03 (0.09) b	3.36 (0.24) c	−5.88 (0.7) a	−6.02 (0.52) a
SAR	0.13 (0.08) c	−0.16 (0.04) c	−2.8 (0.69) b	−4.84 (0.72) a	0.33 (0.1) b	−0.4 (0.25) b	−3.34 (0.74) a	−3.29 (0.49) a	0.41 (0.09) c	−0.13 (0.04) c	−2.68 (0.7) b	−5.56 (0.34) a
Sodium (mg L^{-1})	4.1 (3.1) b	4.9 (5) b	−807 (109) a	−735 (177) a	9.8 (4.1) b	−11.4 (21.2) b	−735 (162) a	−549 (91.1) a	14.3 (4.1) c	9.8 (3.8) c	−660 (83.4) b	−895 (59.5) a
Cadmium (mg kg^{-1})	0.0 (0.0) a	0.0 (0.0) a	0.2 (0.25) a	−0.01 (0.07) a	0.0 (0.0) a	0.0 (0.0) a	0.16 (0.13) a	0.21 (0.05) a	0.0 (0.0) a	0.0 (0.0) a	−0.06 (0.07) a	−0.03 (0.19) a
Copper (mg kg^{-1})	1.4 (2.2) a	0.0 (0.8) a	21.0 (14.0) a	9.0 (2.6) a	4.1 (1.8) a	1.6 (0.3) a	−1.0 (6.4) a	−21.6 (40.7) a	2.5 (1.8) a	1.1 (0.7) a	3.6 (3.6) a	5.3 (9.9) a
Lead (mg kg^{-1})	1.6 (1.9) a	0.0 (0.8) a	13.0 (10.3) a	5.0 (3.4) a	4.1 (3.7) a	0.8 (0.8) a	−2.6 (4.2) a	9.5 (1.7) a	3.3 (2.1) a	0.6 (0.9) a	1.9 (3.8) a	5.8 (6.9) a
Zinc (mg kg^{-1})	2.5 (6.9) b	−5.3 (1.8) b	54.0 (23.7) a	0.8 (9.5) b	5.8 (3.0) b	−0.8 (2.2) b	−0.9 (14.4) b	37.1 (12.6) a	6.3 (6.3) a	1.0 (2.9) a	−4.3 (11.7) a	0.5 (32.3) a
Bulkdensity (mg m^{-3})	0.5 (0) b	0.5 (0) b	1.25 (0.10) a	1.16 (0.03) a	0.5 (0) b	0.5 (0) b	1.29 (0.14) a	1.19 (0.06) a	0.5 (0.0) b	0.5 (0.0) b	0.89 (0.09) a	1.0 (0.09) a

Leachate quantities were lower for diesel and crude oil soils amended with compost or compost-fertilizer than fertilizer or unamended treatments (Figure A1). Reduced leachate from compost treatments can be linked to increased vegetation biomass and increased water retention. Compost increases organic matter content of soils, which improves water holding capacity and reduces impacts of petroleum induced water repellency [52]. Hydrocarbon compounds in contaminated soils bind to organic matter particles, reducing their bioavailability and phytotoxic effects [53]. Numerous studies have documented increased vegetation productivity following application of compost [3,16,27,54]. Therefore, combined effects of enhanced nutrient availability, reduced water repellency, and decreased phytotoxicity are likely responsible for the significant increase in vegetation growth in diesel and crude oil contaminated soils.

3.2. Vegetation Responses

Vegetation cover and biomass (above and below ground) were affected more by contaminants and amendments than seeding (Figures 1 and 2). Seeding with native grasses had no effect on total canopy cover, although vegetation groups showed some seeding impacts (Figure 1). In most cases total vegetation cover and biomass (above and below ground) were significantly greater in compost and compost-fertilizer treatments in uncontaminated (Figures 1a and 2a) and contaminated soils irrespective of seeding treatments (Figures 1e,i and 2c–f). With no amendment, total vegetation cover and biomass were significantly greater in uncontaminated than the contaminated soil treatment (Figures 1a,e,i and 2) except below ground biomass in the seeded fertilizer treatment (Figure 2b).

Wyszkowski and Kordala [9] found compost and mineral materials had a significant influence on heavy metals in aerial parts and biomass yield of maize. Wyszkowski and Ziółkowska [55] found compost had a positive influence on yield of *Lupinus luteus* L. but did not affect maize yields. Seeding increased cover of perennial native grasses in all amendment treatments irrespective of contamination, with a significant increase in compost and compost-fertilizer treatments relative to fertilizer and no amendment (Figure 1d,h,l). In most cases, seeding decreased cover of annual and perennial non-native forbs in contaminated (Figure 1f,g,j,k) and uncontaminated (Figure 1b,c) soils. Of the species seeded, *Agropyron trachycaulum* established most successfully and represented the majority of perennial native grass cover in most treatments. Perennial native grasses were not present in the seed bank or seed rain, therefore seeding contaminated sites can improve the cover of desired species. A number of annual and perennial non-native forbs established on uncontaminated and contaminated soils are listed as noxious or nuisance weeds (Table A2), and therefore must be controlled on sites through mechanical, chemical, or biological measures [56].

When vegetation response by year was considered, total vegetation cover in uncontaminated soil was significantly greater than in contaminated soil in both years. However, in year 2, vegetation cover decreased in uncontaminated soil (71.6 to 63.4%) and increased in diesel (0.1 to 5.4%) and crude oil (0.8 to 5.6%) contaminated soils (Figure 3a). Above and below ground biomass also showed similar responses, including cover in both years (Figure 3b). These findings are consistent with other studies [9,31,57]. Contamination of soils with petroleum hydrocarbons can affect vegetation due to an imbalance in nutrient ratios, water retention restrictions, and/or phytotoxic effects from compounds in the fuel [48,58].

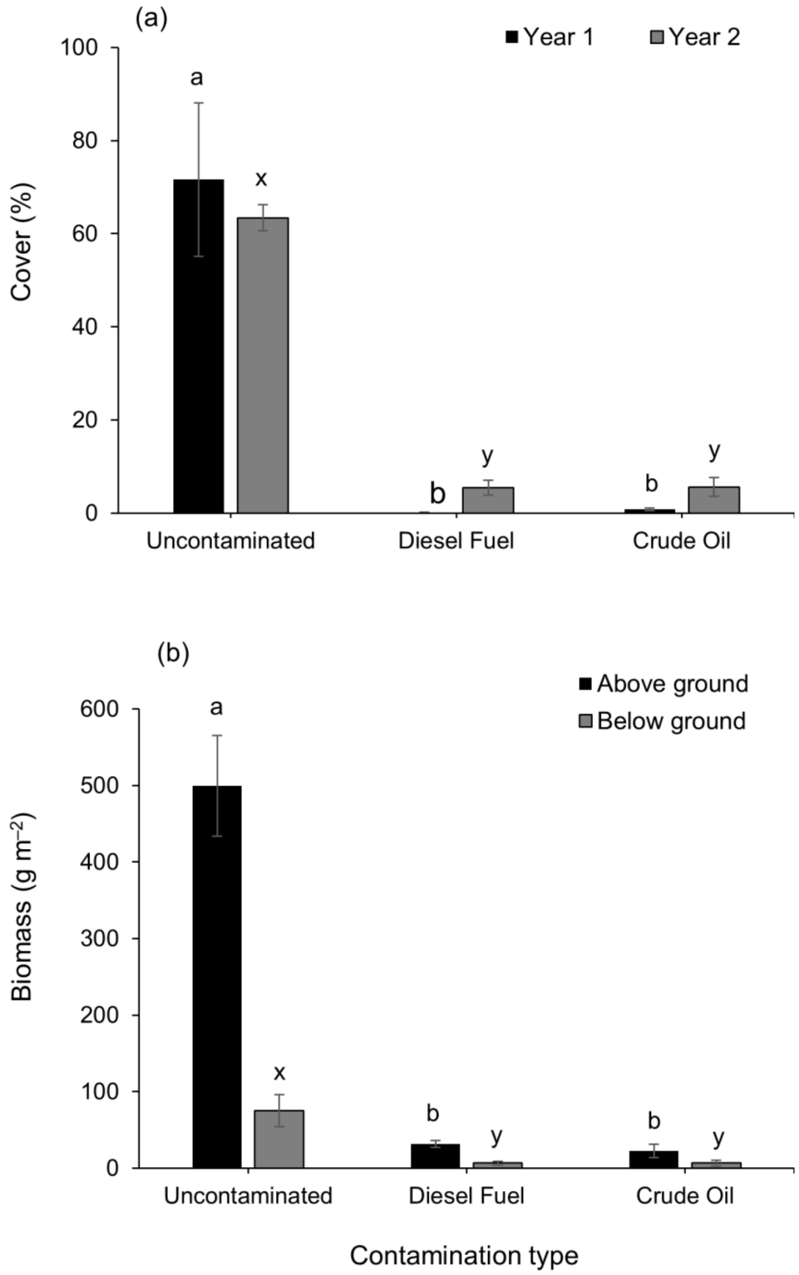

Figure 3. Mean (±SE) vegetation (**a**) cover percent and (**b**) biomass in uncontaminated and contaminated soils. Different letters indicate significant differences among contamination types by year and biomass at $p = 0.05$.

3.3. Hydrocarbon Remediation

Diesel or crude oil contaminated soils had greater concentrations of total extractable hydrocarbons before and after the experiment relative to uncontaminated soil, but by year 2, contamination decreased 42% in diesel and 59% in crude contaminated soil (Table 3). Diesel contaminated soil had significantly greater F2 and F3 hydrocarbons and crude oil soil had significantly greater F2, F3, and F4 hydrocarbons than uncontaminated soil. Diesel soil had more F2 hydrocarbons and less F4 hydrocarbons than crude oil soil due to different concentrations of hydrocarbon fractions in diesel and crude oil. Within two years after reclamation, concentration of F2 and F4 hydrocarbons in crude oil soils were below the CCME guideline for agriculture, commercial, and industrial land use [43], which was not the case for diesel fuel soils (Table 3). Hydrocarbons did not increase the C:N ratio to a level that would cause nutrient immobilization (Table 3). Water repellency is more common in contaminated soils and low soil water content plays an important role in repellency severity [59]. In our study, total leachate volumes were approximately three times greater in contaminated soils (diesel = 125 mm, crude oil = 125 mm) than uncontaminated (44 mm), which could be attributed to low vegetation biomass in contaminated soils (Table 3). Phytotoxicity from petroleum contamination depends upon type and composition of contamination, with lighter hydrocarbon fractions (F2) representing greater toxicity to plant establishment [60]. The low vegetation growth on contaminated soils is most affected by elevated concentrations of F2 hydrocarbons in diesel and crude oil soils (Table 3).

Table 3. Mean (±SE) overall soil hydrocarbon concentrations (mg kg^{-1}) in uncontaminated, diesel fuel, and crude oil contaminated soils before and after the experiment. Different letters indicate significant differences at $p = 0.05$.

Hydrocarbon Type	**Time**	**Uncontaminated**	**Diesel Fuel**	**Crude Oil**	**CCME Guideline (2001) (mg kg^{-1})**		
					Agricultural	**Commercial**	**Industrial**
Total extractable Hydrocarbon (mg kg^{-1})	Before	96.0 (33) b	12,500 (645) a	10,075 (950) a	-	-	-
	After	34.0 (13) b	5213 (220) a	5863 (597) a			
F2 Hydrocarbon (>C11–C16) (mg kg^{-1})	Before	5.0 (0) c	6888 (537) a	2730 (297) b	450	760	760
	After	5.0 (0) b	2064 (267) a	501 (51) b			
F3 Hydrocarbon (>C16–C34) (mg kg^{-1})	Before	63.0 (20) b	4698 (226) a	5278 (492) a	400	1700	1700
	After	33.0 (11) b	3125 (186) a	3593 (355) a			
F4 Hydrocarbon (>C34–C60) (mg kg^{-1})	Before	34.0 (12) b	106 (19) b	1845 (96) a	2800	3300	3300
	After	6.0 (1) b	44 (26) b	1770 (270) a			
Total Organic Carbon (mg kg^{-1})	Before	2.0 (0.2) b	3.7 (0.3) a	3.5 (0.4) a			
	After	2.2 (0.3) b	3.3 (0.2) a	2.8 (0.2) b			
Carbon to Nitrogen Ratio	Before	15.3 (0.9) b	16.2 (0.5) ab	17.8 (0.4) a			
	After	15.5 (1.5) b	15.2 (0.7) b	19.2 (1.1) a			
Leachate (mm)	After	44.0 (4) b	125 (18) a	125 (7) a			

Soil hydrocarbon remediation was influenced more by soil amendments than seeding except for uncontaminated soil where only F2 hydrocarbon changes in unseeded treatment showed some effect (Figure A2). This suggests seeding with native grasses alone may not remediate soil hydrocarbon, as seen in other studies [27,31]. Vouillamoz and Milke [27] found little difference in diesel fuel degradation between seeded (*Lolium perenne* L. (perennial ryegrass)) and unseeded treatments and concluded compost stimulated biodegradation more than vegetation. Similarly, Bailey and McGill [31] found no beneficial effect of annual cereal, pulse, and forage crops on biodegradation of oil and creosote contaminated soils. However, a contradictory finding with the grass *Festuca arundinacea* [28], perennial forages [61], *Lolium* spp. [32], or *Cynodon dactylon* (L.) pers. [8] reduced hydrocarbon contamination up to 58%.

No significant change in hydrocarbon concentrations was found with no amendment and fertilizer treatments; however, addition of compost increased the total extractable F3 and F4 hydrocarbons and led to changes over the study period (Figure 4a,d). Although no differences in F2 and F3 hydrocarbons in compost and compost-fertilizer treatments were found (Figure 4b,c), there were significantly greater F4 hydrocarbons (Figure 4d). The decline in hydrocarbon concentration in no amendment treatments suggested soil was capable of hydrocarbon degradation. Soils in our study were coarse texture, which likely assisted natural attenuation as degradation of polycyclic aromatic hydrocarbons has been greater in sandy soils relative to fine silt and clay texture soils [62]. The low biomass in no amendment fuel contaminated treatments suggests soil properties were limiting biological organisms and therefore likely hydrocarbon degradation.

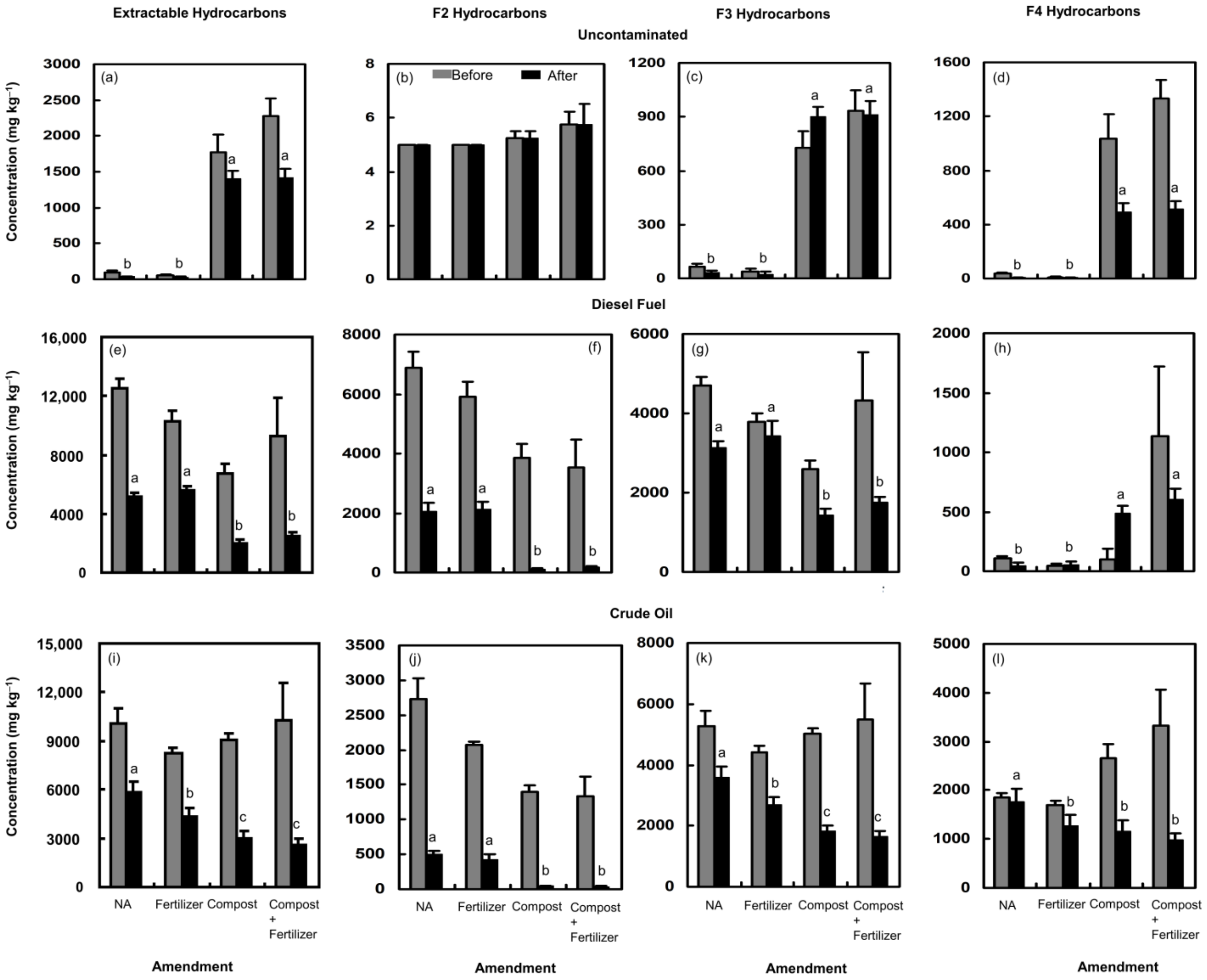

Figure 4. Mean (±SE) total extractable hydrocarbon concentrations in amended (**a**–**d**) uncontaminated, (**e**–**h**) diesel fuel and (**i**–**l**) crude oil contaminated soils before and after the study. Different letters indicate significant differences at $p = 0.05$. NA = No Amendment.

Compost and compost-fertilizer treatments had significantly lower total extractable, F2, F3, and F4 hydrocarbons than fertilizer and no amendment treatments, except F4 hydrocarbons in diesel soils (Figure 4e–l), indicating compost affected hydrocarbon remediation. Compost promoted degradation of polycyclic aromatic hydrocarbons [63,64], and improved vegetation establishment on contaminated soils [9,65]. Plant roots provide a living place for microorganisms [66], and microbial populations in the rhizosphere can be two to three orders of magnitude higher than in surrounding soils and have greater metabolic capabilities [30,48,67], thus vegetation is a crucial component in soil remediation. Chemical substances secreted by microorganisms improve bioavailability of petroleum pollutants, which is conducive to microbial metabolism and decomposition [68].

Binet et al. [69] found low concentrations of polycyclic aromatic hydrocarbons in roots, and root and shoot tissue of *Lolium* spp. (ryegrass), suggesting dissipation in the rhizosphere is due to biodegradation or biotransformation. Due to improvement in soil properties and vegetation establishment following compost application, increased hydrocarbon degradation has been observed in compost amended soils [64]. The results of our study support these observations for total extractable F2 and F3 hydrocarbons in contaminated soil. Hydrocarbon degradation in uncontaminated soils with compost consisted mainly of

F4 hydrocarbons (Figure 4d). Therefore, lower concentrations of total extractable F2 and F3 hydrocarbons with compost can be attributed to petroleum hydrocarbon removal rather than compost hydrocarbon removal. Kästner and Mahro [63] concluded organic matter in compost was likely the most important factor in degradation as microflora responsible for degradation were adapted to humic substances in organic matter.

Although most studies with inorganic fertilizers found enhanced hydrocarbon degradation [30,70], our results were contradictory. The acidic pH of the fertilizer treatment can likely explain the limited stimulatory effect on degradation (Table 1), as optimal pH for remediation is between 6 and 8 [14,71]. Microbial communities shift under acidic conditions and significant increases in degradation were found when acidic soils were neutralized [13]. Seklemova et al. [72] observed little mineralization of phenanthrene and diesel fuel following fertilizer application and concluded inorganic nutrients stimulated microbial metabolism of readily degradable carbon compounds rather than metabolism of the contaminant.

In our study, lower concentrations of hydrocarbons and higher vegetation biomass in compost treatments indicate bioremediation using compost can be a viable and effective response to soil contaminated by petroleum hydrocarbons as it improves soil quality, promotes vegetation establishment, facilitates hydrocarbon degradation. Use of compost in reclamation creates a market and provides a productive end use for the material, which ultimately reduces landfill accumulations. The negligible differences between the two compost treatments are evident since final hydrocarbons and magnitude of hydrocarbon change at the beginning and end of the experiment did not differ. Several studies suggest nutrient supplementation stimulates or enhances natural attenuation processes of petroleum hydrocarbon biodegradation by microorganisms [73–75], which was not the case in this study, as adding inorganic fertilizer showed little effects on hydrocarbon degradation. However, this study did not measure microbial activity which could limit the understanding of the microbial role in biodegrading. Although conservative long-term projections based on this short-term study can be made, it would be difficult to extrapolate results from the early stages of hydrocarbon remediation process to make a strong longer-term conclusion. Thus, further investigation into long term effects of compost and compost-fertilizer application including vegetation tissue analyses is necessary to conclusively state whether compost or compost-fertilizer is superior in enhancing biodegradation of hydrocarbon contaminated soil reclamation.

4. Conclusions

After two years of a field experiment, contamination with diesel fuel or light crude oil decreased total extractable hydrocarbon concentration in soil, resulting in increased vegetation cover and increased leachate production. Compost amended treatments had the lowest hydrocarbon concentrations and the greatest vegetation cover and biomass at the end of the study. Fertilizer addition to contaminated soils resulted in little to no stimulatory effect on hydrocarbon degradation or vegetation production relative to the no amended treatment. Seeding with native grasses had no effect on hydrocarbon degradation or total canopy cover, although composition of vegetation was different between seeded and unseeded treatments. Within two years after reclamation concentration of F2 (carbon length >C10–C16) and F4 (>C34–C60) hydrocarbons in crude oil soils were below the CCME guideline for agriculture, commercial, and industrial land use. Compost and compost-fertilizer amended treatments had the largest effect on hydrocarbon degradation and vegetation establishment, with little difference between these two amendments, suggesting organic fertilizer addition to compost may not be necessary for better revegetation or remediation of soil hydrocarbons.

Author Contributions: S.C.H. collected and analyzed data, wrote the thesis; A.D. analyzed data, prepared, reviewed, and edited the manuscript from the thesis; M.A.N. conceptualized the experiment, procured funding, developed the experimental design, supervised all the work, and reviewed and edited the manuscript. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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Appendix A

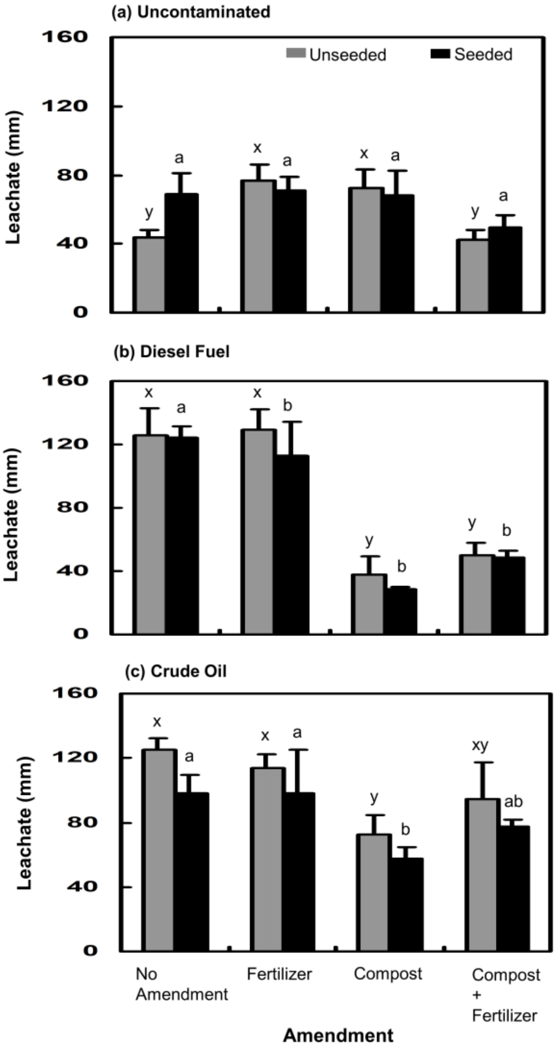

Figure A1. Mean (±SE) cumulative leachate quantities from (**a**) uncontaminated, (**b**) diesel fuel, and (**c**) crude oil contaminated soils. Letters indicate significant differences at $p = 0.05$ among amendment by seeding treatment.

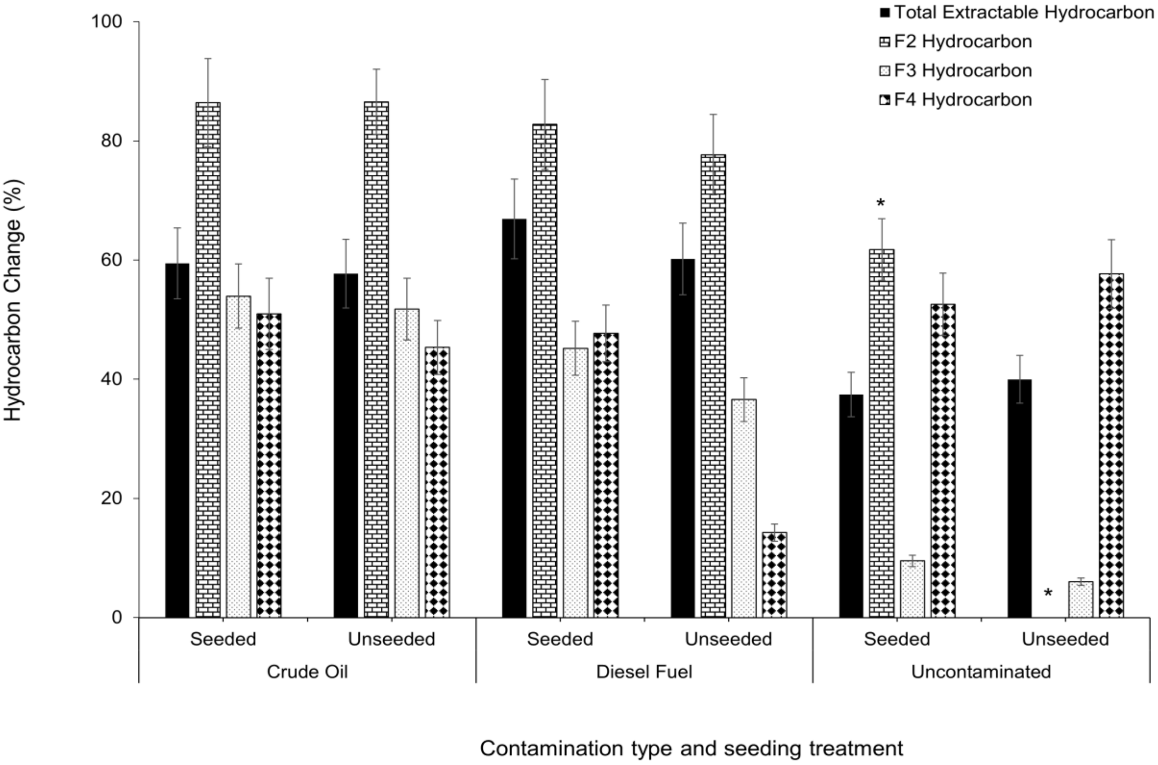

Figure A2. Mean (±SE) soil hydrocarbon percent concentrations change from beginning to end of the experiment in uncontaminated, diesel fuel, and crude oil contaminated soils by seed treatment. * Indicates significant difference between seeded and unseeded treatment in uncontaminated soil.

Table A1. Mean (±SE) compost and soil treatment characterization.

Soil Properties	Compost	Soil	Soil + Fertilizer	Soil + Compost	Soil + Compost + Fertilizer
Sand (%)	-	73 (0.5)			
Silt (%)	-	14 (0.9)			
Clay (%)	-	13 (0.5)			
Bulk density (mg m^{-3})	0.53 (0.01)	1.88 (0.02)	1.88 (0.02)	1.74 (0.01)	1.74 (0.01)
Total extractable hydrocarbons (mg kg^{-1})	9033 (491)	76 (15)	55 (13)	2053 (292)	2058 (163)
Total organic carbon (%)	19.9 (0.6)	1.8 (0.1)	1.6 (0.2)	6.4 (0.3)	6.3 (0.1)
Total kjeldahl nitrogen (%)	1.85 (0.01)	0.13 (0.01)	0.2 (0.01)	0.52 (0.02)	0.54 (0.01)
Carbon:Nitrogen	11.2 (0.4)	14.5 (0.5)	8.6 (0.7)	13 (0.2)	12.1 (0.3)
Ammonium (mg kg^{-1})	7.5 (0.2)	6.1 (0.3)	811.8 (0.2)	10.7 (1.5)	638.2 (22.9)
Nitrate (mg kg^{-1})	1867 (75)	8.5 (0.4)	8.3 (0.5)	344.4 (19.9)	317.5 (28.8)
pH	7.7 (0.03)	6.63 (0.02)	6.65 (0.04)	7.13 (0.04)	7.19 (0.02)
Electrical conductivity (dS m^{-1})	16.7 (0.47)	0.49 (0.02)	0.51 (0.02)	10.18 (0.28)	9.89 (0.11)
Sodium adsorption ratio	15.9 (0.4)	0.29 (0.01)	0.29 (0.01)	9.69 (0.25)	9.5 (0.12)
Sodium (mg L^{-1})	2387 (89)	11.3 (0.4)	11 (0.4)	1399.2 (40.1)	1355 (22.5)
Cadmium (mg kg^{-1})	3.0 (0.1)	0.5 (0)	0.5 (0)	1.01 (0.09)	1.04 (0.06)
Copper (mg kg^{-1})	200 (2.8)	12.8 (0.6)	11 (0.6)	62.6 (5.5)	63.1 (3)
Lead (mg kg^{-1})	152 (2.9)	11.1 (0.7)	9.7 (0.5)	47.3 (4.2)	46.5 (2.6)
Zinc (mg kg^{-1})	646 (9.9)	54.2 (2.6)	47.9 (1.9)	204.8 (17.5)	212.3 (12.9)
Fecal Coliforms (MPNU g^{-1})	<3 (0)	<3 (0)	<3 (0)	<3 (0)	<3 (0)
Salmonella	Not isolated	Not isolated	Not isolated	Not isolated	Not isolated

Table A2. Plant species grouped by functional type and weed status by the province of Alberta.

Functional Group	Common Name	Scientific Name	Weed Status
Annual non-native forb	Green foxtail	*Setaria viridis* (L.) Beauv.	Nuisance
	Redroot pigweed	*Amaranthus retroflexus* L.	Nuisance
	Wild buckwheat	*Fallopia convolvulus* (L.) Á. Löve	Nuisance
	Lamb's quarters	*Chenopodium album* L.	
	Kochia	*Bassia scoparia* (L.) A. J. Scott	
	Annual hawk's beard	*Crepis tectorum* L.	Nuisance
	Shepherd's purse	*Capsella bursa-pastoris* (L.) Medik.	Nuisance
	Flixweed	*Descurainia sophia* (L.) Webb ex Prantl	Nuisance
	Purslane	*Portulaca oleracea* L.	
	Prostrate knotweed	*Polygonum aviculare* L.	
Perennial non-native forb	Absinth	*Artemisia absinthium* L.	
	Perennial sow thistle	*Sonchus arvensis* L.	Noxious
	Dandelion	*Taraxacum officinale* Weber ex F. H. Wigg.	Nuisance
	Broad leaved plantain	*Plantago major* L.	
Annual native forb	Pygmy flower	*Androsace septentrionalis* L.	
	Pineapple weed	*Matricaria discoidea* DC.	
	Canada fleabane	*Erigeron canadensis* L.	
Perennial native forb	Rough cinquefoil	*Potentilla norvegica* L.	Nuisance
Annual non-native grass	Canola	*Brassica rapa* L.	
	Wheat	*Triticum aestivum* L.	
	Barnyard grass	*Echinochloa crus-galli* (L.) Beauv.	
Perennial native grass	Foxtail barley	*Hordeum jubatum* L.	
	Hairy wildrye	*Leymus innovates* (Beal) Pilg.	
	Slender wheatgrass	*Elymus trachycaulus* (Link) Gould ex Shinners	
	Tufted hairgrass	*Deschampsia cespitosa* (L.) P. Beauv.	
	Bluejoint	*Calamagrostis canadensis* (Michx. Beauv.)	

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