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Degradation and metabolism of tetraethyllead in soils

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SUMMARY

The objective of this study was to determine the disappearance of the leaded gasoline enhancer tetraethyllead (TEL), formation of degradation products, and mass balance in nonsterile and autoclaved Leon and Madison soils. Ethyl-1-^{14}C-labeled TEL was used so that mineralization rates of TEL and mass balance could be determined. ^{14}C-TEL in nonsterile and autoclaved surface and subsurface samples of the two soils disappeared rapidly, and ionic ethyllead products, water soluble nonlead organic products and bound residues were rapidly formed. A small fraction ($\leq$7.74%) of ^{14}C-TEL in nonsterile soil samples was mineralized to $^{14}CO_2$ in 28 days. Triethyllead (TREL) was the major ionic ethyllead product detected in both nonsterile and autoclaved soils; diethyllead (DEL) was occasionally detected. Recovery of ^{14}C from mass balance studies for all nonsterile and autoclaved soil samples after 28 days of incubation was poor, less than 50% of the ^{14}C applied. It appears that unknown volatile and/or gaseous organic products were the major degradation products of TEL in soils. Based on the observations of more rapid initial disappearance of ^{14}C-TEL, more rapid formation and more rapid disappearance of ^{14}C-DEL, and occurrence of $^{14}CO_2$ production in nonsterile soils, it was concluded that both biological and chemical degradation contributed to the degradation of TEL in soils, with chemical degradation being the major factor.

INTRODUCTION

For over 50 years, since its introduction in 1923 by General Motors, tetraethyllead (TEL) was the major antiknock agent used in leaded gasoline. After tetramethyllead (TML) was introduced in 1960, TEL and TML were added to gasoline as mixtures or singly to obtain desired octane numbers [6,16]. TEL was suitable for high speed engines, whereas TML was good for low speed engines [6]. Although leaded gasoline was completely phased out for use in automobiles in early 1980 in the United States, leaded gasoline containing TEL is still used in aircraft engines. Apparently there is no substitute for TEL in gasoline for high speed engines.

Both TEL and TML, and their ionic degradation products are toxic to all forms of life [6] and TEL is more toxic than its counterpart, TML. Even though triethyllead (TREL) and diethyllead (DEL) are ionic, the mode of action of TEL, TREL and DEL are similar [6]. Nonionic TEL and TML are generally more toxic to animals, whereas ionic forms of alkyl lead compounds are more toxic to plants and plankton [6,7,16]. Ionic trialkyllead compounds inhibit growth of bacteria and fungi [16,17]. Growth of algae is also inhibited by tetraalkyllead and trialkyllead compounds [17]. However, it is difficult to evaluate the toxicity from tetraalkyllead compounds in liquid media. These compounds are subject to rapid degradation to ionic trialkyllead compounds through chemical- and photo-degradation [8,14]. Depending upon the length of testing, toxicity to the test organisms may be from trialkyllead compounds, rather than from tetraalkyllead compounds.

Due to the extensive past use of leaded gasoline in automobiles, TEL, TML, and their degradation products, ionic alkyl lead compounds, were found to be ubiquitously present in the environment prior to late 1980 [14,16]. Ionic lead alkyl compounds were the major species detected in surface water, rain water, and sediments [3,4,14,15]. TEL in aqueous solutions appeared to undergo a series of sequential deethylations, first to triethyllead and then to diethyllead, eventually to Pb^{2+} [16]. Little information is available regarding the fate of TEL in soil. Triethyllead and diethyllead were the only organolead compounds detected in soil samples collected from an urban area [1]. Ou et al. [12] reported that TEL in nonsterile and autoclaved surface and subsurface soils were rapidly transformed to ionic ethyllead species, presumably to triethyllead and diethyllead, and both biological and chemical degradation contributed to the degradation of TEL. Furthermore, in addition to degradation of ionic ethyllead species and CO_2, unknown volatile products could also be formed. Microorganisms capable of degrading TEL or TML have not been reported. A strain of *Arthrobacter* sp., a strain of the wood decay fungus *Phaeolus schweinitzii*, and a number of hydrocarbon-utilizing bacteria have limited capacities to degrade ionic trimethyllead [9,10].

The objectives of this study are to determine: mineralization rates of ^{14}C-TEL in two soils, Leon sand and Madison loam; disappearance rates of ^{14}C-TEL and formation of degradation products in nonsterile and autoclaved soils; and the mass balance of ^{14}C in the two soils treated with ^{14}C-TEL.

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MATERIALS AND METHODS

Soils

Soil samples were collected at 15-cm increments to 45-cm depth from two forest sites located at the Tyndall Air Force Base (AFB), FL, USA and the Dobbins AFB, GA, USA; they were classified to be Leon sand and Madison loam, respectively. Soil samples were stored in the dark at 4 °C and used within 3 months after collection. Selected properties of the two soils are shown in Table 1.

Chemicals

Analytical grade TEL and TREL chloride salt were obtained from All-Chemie Inc. (Ft Lee, NJ, USA). Ethyl-1-^{14}C-TEL, which had a specific activity of 0.74 GBq mmol^{-1} and radiopurity of better than 99% was purchased from American Radiolabeled Chemicals (St Louis, MO, USA). We found that more than 99% of ^{14}C-TEL fractionated into hexane, indicating that more than 99% of ^{14}C was associated with a hydrophobic chemical and TEL is a hydrophobic chemical. ^{14}C-TREL was synthesized using ^{14}C-TEL as starting chemical, and ^{14}C-DEL and nonlabeled DEL were obtained using ^{14}C-TREL and nonlabeled TREL, respectively. The procedures for these conversions were described by Ou et al. [13]. All other chemicals were either analytical grade, scintillation grade, or the highest grade commercially available.

Mineralization studies

The experimental setup for mineralization of ^{14}C-TEL in soil was similar to the procedure described by Ou et al. [12], with slight modifications. In short, 50 g of nonsterile or autoclaved soil (oven-dry weight basis) were transferred to a 250-ml Erlenmeyer flask containing 3.7 kBq of ^{14}C-TEL and 1000 μg of TEL. An additional 50 g of new soil was then added immediately on top of the soil in the flask. After a brief but thorough mixing, the flask was closed tightly with a Teflon-lined screw cap under which a stainless steel vial containing 0.5 ml of 8 mol L^{-1} KOH was hung. All flasks were wrapped with aluminum foil and incubated in the dark at 28 °C. At predetermined intervals, all vials were removed and replaced with new vials containing fresh KOH. The removed vials were placed individually in small glass beakers and the KOH was diluted with deionized H_2O to 5 ml; 0.5 ml of the diluted KOH was used for ^{14}C determination by liquid scintillation counting (LSC). To reduce photocomposition, all operations were carried out under semi-dark conditions. At the end of incubation (28 days), 10 g of soil from each flask were transferred to a 50-ml glass centrifuge tube for solvent extraction as described below. The ^{14}C remaining (bound residues) in the solvent-extracted soil samples was combusted to $^{14}CO_2$ by a sample oxidizer and quantified by liquid scintillation counting (LSC) [11].

The experimental setup for determination of mineralization rates of ^{14}C-TREL in nonsterile Leon and Madison soils was similar to that for ^{14}C-TEL mineralization.

TABLE 1

Selected properties of Leon sand and Madison loam

Soil depth (cm)	pH	Soil-water content	Organic C	Sand	Silt	Clay
		(g kg^{-1})				
Leon						
0–15	5.1	40	15.5	900	30	70
15–30	5.0	21	1.1	960	30	30
30–45	5.1	87	11.0	820	90	90
Madison						
0–15	4.7	320	6.1	430	400	170
15–30	4.7	206	2.8	500	330	170
30–45	4.6	180	1.5	540	300	160

Disappearance of TEL and metabolite formation

A series of 50-ml glass centrifuge tubes wrapped with aluminum foil were set up in which each contained 5 g of nonsterile or autoclaved soil. After 100 μg of TEL and 2 kBq of ^{14}C-TEL were added to each tube, an additional 5 g of the same soil was added to the top of the soil in the tube. The tubes were closed tightly with Teflon-lined screw caps and were immediately shaken in a reciprocal shaker for 5 min. Two tubes from each treatment immediately received 10 ml of *n*-hexane and 10 ml of 0.053 mol L^{-1} EDTA solution (20 g of disodium EDTA in 1 L deionized H_2O), and were shaken for 30 min. The two tubes were then centrifuged at 500 × *g* for 10 min for separation of the two phases, hexane and EDTA. After 0.5 ml was removed from the hexane phase for ^{14}C determination by LSC, the rest of the solution was transferred to a 50-ml plastic centrifuge tube. The two tubes were centrifuged at 25 000 × *g* for 10 min. After removal of the hexane and the EDTA layers from the tube, 10 ml each of hexane and EDTA solution were added to soil in the tubes. The two tubes were subjected twice to the same process of shaking, centrifugation, and phase separation as described above. ^{14}C in the combined hexane extract and the combined EDTA fraction were quantified by LSC. The EDTA fraction then received 5 ml of *n*-hexane and 1 ml of 0.065 mol L^{-1} sodium diethyldithiocarbamate (NaDDTC, 20 g in 1 L deionized H_2O), and the tubes were then shaken for 15 min. After the hexane-NaDDTC phase was removed, the aqueous phase was subjected to the same hexane-NaDDTC extraction. The combined hexane-NaDDTC was then concentrated under a gentle stream of N_2 gas to 0.1 ml. The rest of the tubes were incubated in the dark at 28 °C. At predetermined intervals, two tubes from each treatment were removed for hexane and hexane-NaDDTC extractions. ^{14}C remaining in the extracted soil samples was combusted to $^{14}CO_2$ in a sample oxidizer and quantified by LSC. All operations were carried out under semi-dark conditions. All experiments were carried out in duplicate.

The concentrated hexane-NaDDTC extracts were spotted on silica gel G TLC plates and the plates were developed in acetone/*n*-hexane/propionic acid (30:70:2, v/v/v). After the plates were air-dry they were covered with x-ray films (SB-5, Eastman Kodak, Rochester, NY, USA). After 21 days of exposure, the films were developed, and silica gel in the radio-

active areas was scraped off and transferred to scintillation vials for ^{14}C determination by LSC.

Confirmation by GC-AAS

Degradation products of ^{14}C-TEL and ^{14}C-TEL in nonsterile and autoclaved soils were confirmed by established gas chromatography-atomic absorption spectrometry (GC-AAS) techniques [2,4]. A detailed description was given by Ou et al. [13] for confirmation of TREL, DEL and their degradation products in soil. Briefly, A GC (Hewlett Packard 5890, Palo Alto, CA, USA) equipped with an autosampler and an integrator was interfaced with an AAS (Perkin Elmer, Norwalk, CT, USA) equipped with a Perkin Elmer Pb detector with a deuterium background corrector through nickel tubing. Soil samples treated with analytical grade TEL were first extracted with *n*-hexane, and then with *n*-hexane-NaDDTC. The hexane extracts were injected directly into the GC-AAS for determination of TEL. Prior to injection into the instrument, ionic ethyllead compounds in the hexane-NaDDTC extracts were butylated with magnesium butylchloride. Retention times for TEL, TREL and DEL were 3.9, 6.7, and 8.5 min, respectively.

RESULTS AND DISCUSSION

Mineralization

Mineralization of ^{14}C-TEL in nonsterile surface and subsurface samples of Leon and Madison soils was initially rapid. Mineralization during the first 7 days accounted for more than half of the total mineralization during the entire 28 days of incubation (Fig. 1). After 7 days, mineralization of ^{14}C-TEL leveled off in all samples. In Leon soil more ^{14}C-TEL was mineralized in the surface samples than in the subsurface samples. In contrast to the Leon soil, in Madison soil more ^{14}C-TEL was mineralized in the subsurface samples than in the surface samples. After 28 days only 4.26–5.35% and 5.98–7.74% of the applied ^{14}C was mineralized in Leon and Madison soils, respectively. It should be noted that the mineralization results shown in Fig. 1 also included small amounts of volatile ^{14}C-organics, most likely ^{14}C-TEL, which could amount for a very small fraction of total ^{14}C trapped in KOH. Ou et al. [12] found that after 28 days of incubation less than 5% of total ^{14}C volatilized from ^{14}C-treated Arredondo fine sand and trapped in KOH was associated with volatile ^{14}C-organics, the rest being associated with $^{14}CO_2$. Ou et al. [12] also reported that mineralization of ^{14}C-TEL was a microbial process. No mineralization occurred in autoclaved Arredondo surface and subsurface soil samples. Thus, it is expected that mineralization of ^{14}C-TEL also would not occur in autoclaved Leon and Madison soils.

At the end of 28 days of incubation, total ^{14}C recoveries in the ^{14}C-TEL treated Leon and Madison soils were poor, ranging from 18.88 to 28.70% for the Leon samples, and from 24.41 to 33.55% for the Madison soil (Table 2). Total ^{14}C recoveries in the two surface samples were better than in their respective subsurface samples. Patterns of ^{14}C recoveries in the two soils were similar to Arredondo soil samples in which total ^{14}C recoveries were poor and more ^{14}C was recovered in surface samples than in subsurface samples [12].

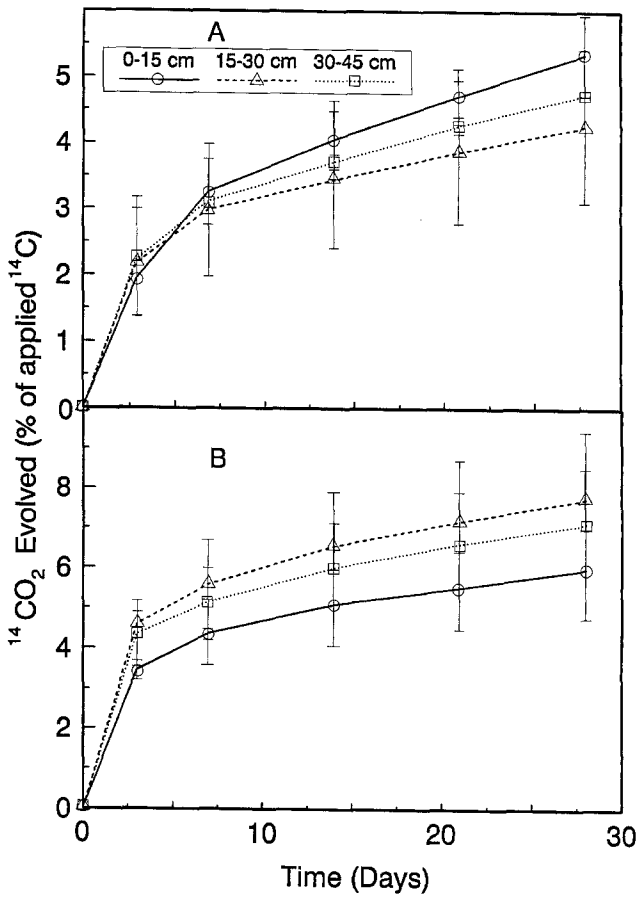

Fig. 1. Mineralization of ^{14}C-TEL in Leon (A) and Madison (B) soils during 28 days of incubation. The error bars indicate standard deviation values. Standard deviations below 0.1% are not shown.

Ionic TREL was the major metabolite of TEL in Leon and Madison soils, respectively (see below). Therefore, mineralization rates of ^{14}C-TREL in nonsterile Leon and Madison soils were investigated. Mineralization of ^{14}C-TREL in nonsterile surface and subsurface samples of the two soils was initially rapid. More than half of the applied ^{14}C mineralized was mineralized during the first 3 days, with the exception of the shallow subsurface sample (15–30 cm depth) of Leon soil (Fig. 2). In this sample, more than half of the applied ^{14}C mineralized was mineralized in the first 7 days. More ^{14}C-TREL was mineralized than ^{14}C-TEL during 28–36 days of incubation. Since TREL is water soluble and TEL is not, it is likely that TREL would be more easily utilized by microorganisms, resulting in a higher level of mineralization. Far better total ^{14}C recoveries (60.75–80.87%) were found in the ^{14}C-TREL-treated samples than in the ^{14}C-TEL-treated samples (Table 2). Higher ^{14}C recovery in the ^{14}C-TREL-treated soil samples was due to higher $^{14}CO_2$ production (nonsterile samples), higher levels of nonextractable ^{14}C, and higher levels of hexane-NaDDTC extractable ^{14}C (Leon soil samples).

TEL disappearance and formation of ionic metabolites

TEL has a very low water solubility, but is highly soluble in hydrophobic solvents such as hexane, benzene and gasoline [5,16]. Ou et al. [12] took advantage of these unique properties

TABLE 2

Distribution of ^{14}C in nonsterile Leon and Madison soils after 28 days of incubation with ^{14}C-TEL or ^{14}C-TREL

Soil depth (cm)	$^{14}CO_2$	^{14}C in hexane	^{14}C in EDTA	Nonextractable ^{14}C	Total ^{14}C
	(% of applied ^{14}C)				
			^{14}C-TEL		
Leon					
0–15	5.36	0.80	6.75	15.80	28.70
15–30	4.26	1.57	6.62	6.44	18.88
30–45	4.74	0.60	5.58	7.62	20.52
Madison					
0–15	5.98	0.23	1.55	25.80	33.55
15–30	7.74	0.42	1.19	16.40	25.77
30–45	7.10	0.45	0.75	17.60	24.41
			^{14}C-TREL		
Leon					
0–15	16.60	18.53	0.15	31.08	66.35
15–30	16.83	33.15	3.05	20.58	73.60
30–45	17.69	37.13	0.82	21.11	77.65
Madison					
0–15	11.31	0.53	1.93	67.11	80.87
15–30	16.52	0.37	1.35	54.85	72.63
30–45	18.63	0.68	3.15	38.30	60.75

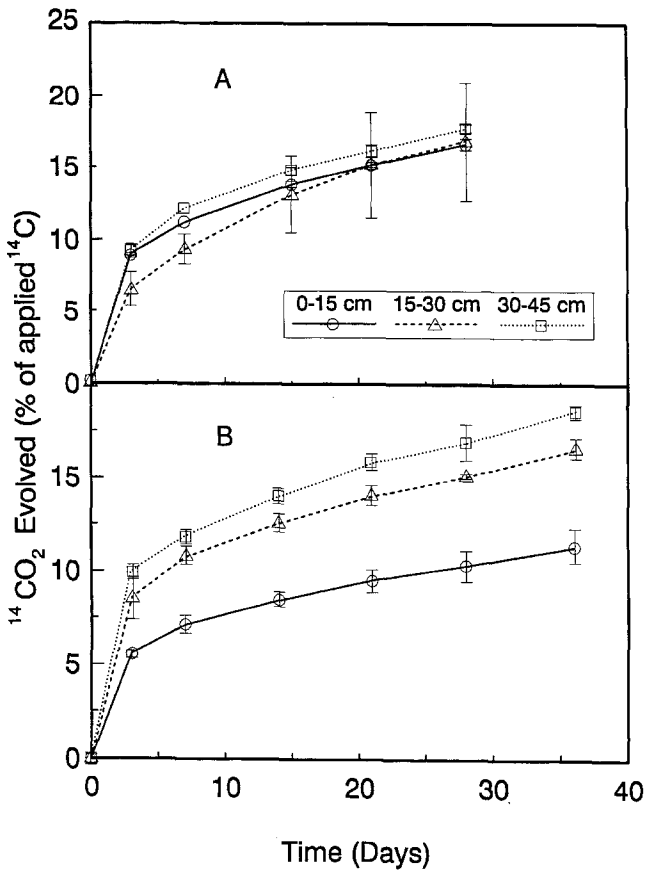

Fig. 2. Mineralization of ^{14}C-TREL in Leon (A) and Madison (B) soils during 28 and 36 days of incubation, respectively. The error bars indicate standard deviation values. Standard deviations below 0.1% are not shown.

for separation of TEL from ionic ethyllead compounds and unknown water-soluble nonlead compounds in soil. The present study went a step further in which ionic ethyllead species were separated from water-soluble nonlead organic compounds in soil. Nonionic ^{14}C-TEL in soil was first fractionated into hexane, and the remaining ionic ^{14}C-ethyllead compounds were then fractionated into hexane-NaDDTC. The remaining ^{14}C in aqueous solution was considered to be associated with water-soluble nonlead organic compounds. Since ^{14}C-TEL was used, TEL, ionic ethyllead compounds, and unknown water-soluble nonlead organic compounds could be easily quantified by LSC. The unknown water-soluble nonlead compounds were possibly more polar than ionic ethyllead compounds.

^{14}C-TEL in nonsterile and autoclaved surface (0–15 cm depth) and subsurface (30–45 cm depth) samples of Leon and Madison soils rapidly disappeared (Figs 3, 4). ^{14}C-TEL in nonsterile samples disappeared more rapidly during the first 3–7 days than in the corresponding autoclaved samples, and ^{14}C-TEL in surface samples disappeared more rapidly than in the corresponding subsurface samples. This indicates that both biological and chemical degradation were involved in the disappearance of TEL from soil, with chemical degradation being the major factor contributing to the disappearance of the chemical from soil. It was estimated that biological degradation was responsible for about 20% and 10% of the ^{14}C-TEL in the Leon and Madison soils, respectively, during the first day of incubation. Smaller disappearance rates in subsurface samples probably reflect the fact that these samples had low microbial activity (nonsterile samples) and low organic matter contents.

With the rapid disappearance of the hexane-extractable ^{14}C, which is associated with ^{14}C-TEL in nonsterile and autoclaved samples, ^{14}C in the EDTA fraction and the nonextractable ^{14}C (bound residues) were formed rapidly and the ^{14}C levels in both phases rapidly leveled off (Figs 3, 4). Total ^{14}C recoveries in nonsterile and autoclaved samples initially declined rapidly, and after 1 day, the declination had either leveled off or stabilized. After 28 days of incubation, total ^{14}C recoveries in all samples were very poor, ranging from 18.9 to 42.1% of the applied ^{14}C. Since $^{14}CO_2$ production in nonsterile samples was not determined, if the mineralization results given in Table 2, which ranged from 4.26 to 7.74% of the applied ^{14}C were used, total ^{14}C recoveries for the nonsterile samples were still very poor, below 50%. Rapid disappearance of ^{14}C in conjunction with rapid declination of total ^{14}C recovery in nonsterile and autoclaved samples indicated that during TEL degradation unknown volatile and/or gaseous ^{14}C-products, besides $^{14}CO_2$, were formed. The majority of the ^{14}C-products appeared to escape into the atmosphere. A small fraction of the products remained in soil and was subsequently mineralized to $^{14}CO_2$. Based on the chemical structure of TEL, the unknown volatile and/or gaseous ^{14}C-products could be ^{14}C-ethane and/or ^{14}C-ethanol.

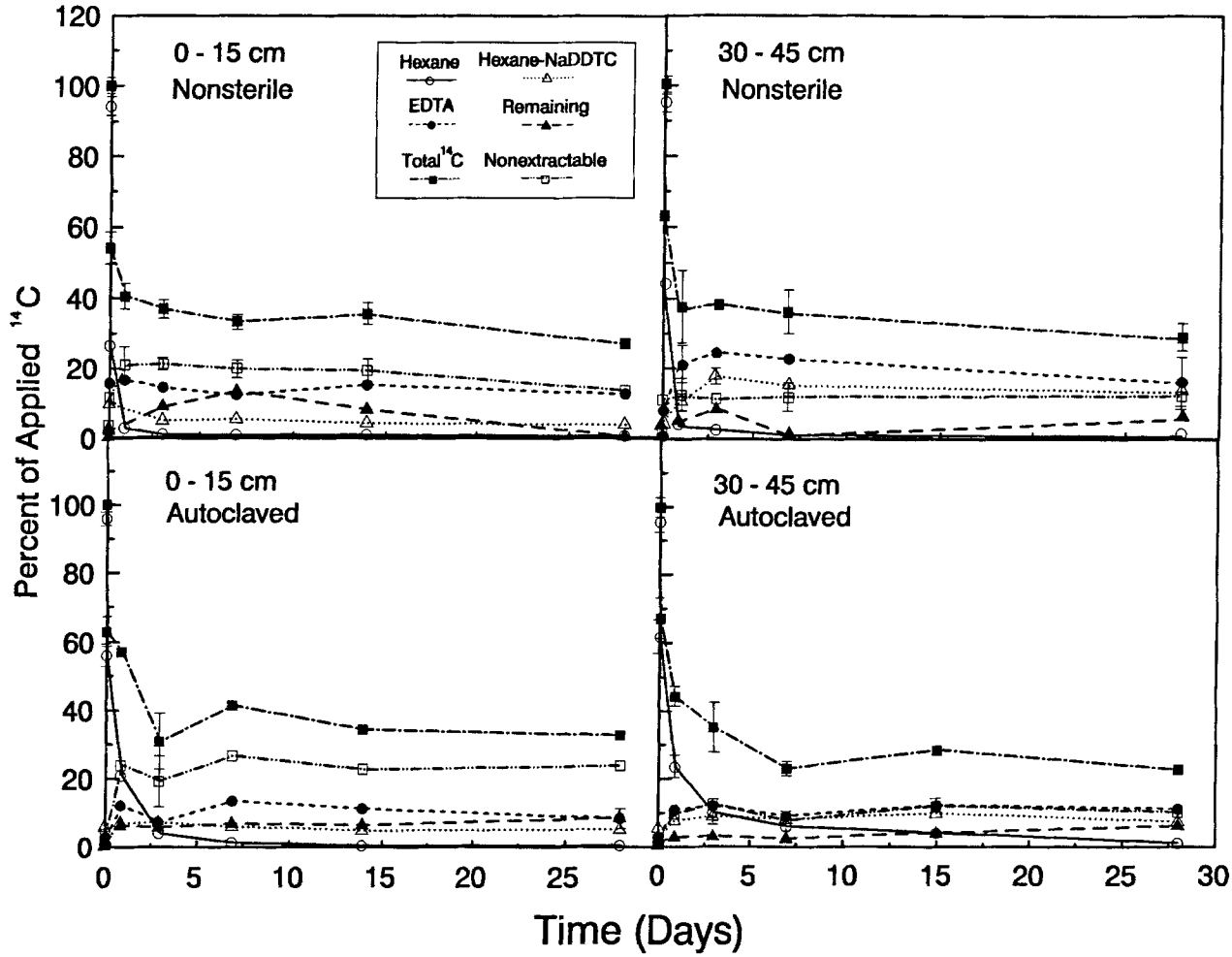

Fig. 3. Distribution of ^{14}C in the hexane phase, the EDTA phase, the hexane-NaDDTC phase, ^{14}C remaining, nonextractable, and total recovery from ^{14}C-TEL-treated nonsterile and autoclaved surface (0–15 cm depth) and subsurface (30–45 cm depth) samples of Leon soil. The error bars indicate standard deviation values. Standard deviations below 2% are not shown.

The ^{14}C remaining in the EDTA fraction represents water-soluble organic metabolites which include ionic ethyllead species and nonlead organic products. The ^{14}C associated with ethyllead compounds was fractionated into the hexane-NaDDTC, and the remaining ^{14}C in the aqueous phase was associated with water-soluble nonlead organic compounds. Ionic ^{14}C-ethyllead compounds were detected in ^{14}C-TEL-treated nonsterile and autoclaved soil samples during the entire 28 days of incubation (Figs 3, 4). Small amounts of ^{14}C remaining in the aqueous phase were also detected throughout the entire incubation period. Levels of ^{14}C remaining in the aqueous phase were generally more stable in autoclaved samples than in nonsterile samples.

Ionic ethyllead metabolites

It has been demonstrated that TEL in nonsterile and autoclaved soils is partially degraded to ionic ethyllead compounds [12]. However, no attempts were made by Ou et al. [12] to identify the nature of the ionic metabolites. In this study, ionic ethyllead species in the hexane-NaDDTC extracts were separated, detected and quantified by TLC-autoradiographic assays and LSC. In the ^{14}C-TEL-treated nonsterile surface samples of Leon and Madison soils, only ^{14}C-TREL was detected during the entire 28 days of incubation, with the exception of the Leon soil. In this sample, ^{14}C-DEL was detected once, one hour after incubation. ^{14}C-TREL was also the only or principal ionic ethyllead product in the autoclaved surface samples of Madison and Leon soils, respectively. ^{14}C-DEL appeared briefly (at 1 and 23 h after incubation) in the autoclaved Leon surface sample. ^{14}C-DEL in this sample accounted for 75 and 25% of total ionic ^{14}C-ethyllead species at 1 and 23 h of incubation, respectively. This indicates that TEL in soil is initially degraded to TREL, which is then degraded to DEL. Appearance of DEL in the nonsterile sample was shorter than in the corresponding autoclaved sample. This suggests that degradation involves both chemical and biological mechanisms, with chemical degradation being the major factor.

Similar to the nonsterile and autoclaved surface samples of the Madison soil, ^{14}C-TREL was the only ionic ethyllead product detected in the nonsterile and autoclaved subsurface samples (30–45 cm depth) of Madison soil during the entire 28 days of incubation. Whereas, in addition to ^{14}C-TREL, ^{14}C-DEL was also detected in nonsterile and autoclaved subsurface samples of Leon soil. The appearance of ^{14}C-DEL in the two

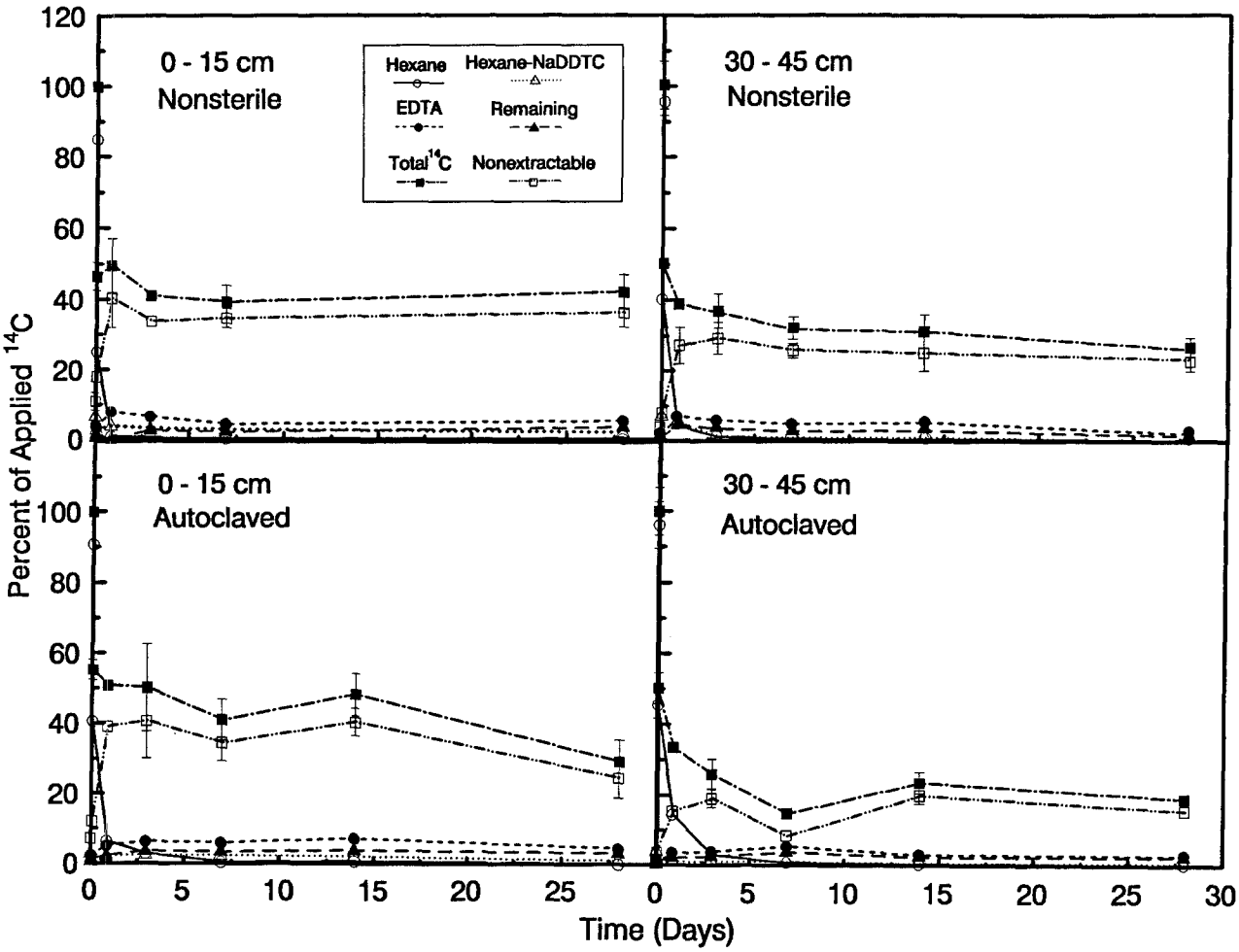

Fig. 4. Distribution of ^{14}C in the hexane phase, the EDTA phase, the hexane-NaDDTC phase, ^{14}C remaining, nonextractable, and total recovery from ^{14}C-TEL-treated nonsterile and autoclaved surface (0–15 cm depth) and subsurface (30–45 cm depth) samples of Madison soil. The error bars indicate standard deviation values. Standard deviations below 2% are not shown.

samples, though brief, lasted longer than in the corresponding surface samples. ^{14}C-DEL was detected in the nonsterile and autoclaved subsurface samples at 1 and 23 h, and at 23 h, 3 and 7 days, respectively. The levels of ^{14}C-DEL in the two samples declined progressively with time. The results of the formation of ^{14}C-DEL in the subsurface samples confirmed the results in the surface samples that both biological and chemical degradation contributed to the formation and subsequent degradation of TREL and DEL. Longer appearance of DEL in the nonsterile and autoclaved subsurface samples was probably due to low microbial activity and low organic matter content in the subsurface sample. It appears that DEL in soil was rapidly degraded to inorganic Pb^{2+}, without the formation of monoethyllead (MEL). MEL appears to be chemically unstable. This chemical was never detected in the environment [15,16]. GC-AAS confirmed the results of the TLC-autoradiographic analysis.

In conclusion, ^{14}C-TEL in nonsterile surface and subsurface soil samples of Leon and Madison soils was rapidly degraded to ^{14}C-TREL, ^{14}C-DEL, $^{14}CO_2$, water-soluble nonlead ^{14}C-organic products, unknown volatile and/or gaseous ^{14}C-products, and bound residues. Both biological and chemical degradation were responsible for the degradation (with the exception of $^{14}CO_2$ evolution), with chemical degradation being the major factor. $^{14}CO_2$ evolution from the ^{14}C-TEL-treated soils is a biological process. ^{14}C-TREL was the major ionic ethyllead metabolite in nonsterile and autoclaved soils.

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