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Consumption of Freons CFC-11 and CFC-12 by Anaerobic Sediments and Soils

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■ A variety of anaerobic sediments and soils consumed CFC-11 ($CFCl_3$) and CFC-12 (CF_2Cl_2). An aerobic soil did not. Active microbial metabolism was required for CFC-12 uptake in all of the sediments examined. CFC-11 uptake was faster in the presence of microbial activity, but reduced components in the sediments also resulted in nonenzymatic CFC-11 consumption in most instances. CFC-12 uptake in a culture of *Clostridium pasteurianum* provided a model for the sediment uptake of CFC-11 and CFC-12 that required active microbial metabolism. Consumption of CFC-11 in the presence of reduced hematin demonstrated a potential mechanism for nonenzymatic CFC-11 consumption. These findings demonstrate that CFC-11 and CFC-12 are not biochemically inert under anaerobic conditions. This suggests that anaerobic degradation of CFC-11 and CFC-12 in anaerobic landfills might prevent some disposed CFC-11 and CFC-12 from entering the atmosphere. The results also suggest that CFC-11 and CFC-12 cannot be used as stable tracers in anaerobic environments. Furthermore, although the microbial sink for atmospheric CFC-11 and CFC-12 is much less than current anthropogenic release, this sink could have a significant long-term effect on the amount of CFC-11 and CFC-12 reaching the stratosphere.

Introduction

Freons CFC-11 ($CFCl_3$) and CFC-12 (CF_2Cl_2) have been implicated in depletion of stratospheric ozone (*1–4*). They are also important greenhouse gases (*5–7*). CFC-11 and CFC-12 have previously been "expected to be almost totally unreactive biologically" (*3*). The apparent stable nature of CFC-11 and CFC-12 and their known atmospheric history have suggested that they may be employed to age-date various surface waters and groundwaters (*8,9*).

However, recent studies have indicated that CFC-11 and CFC-12 are not stable in some environments. Concentrations of CFC-11 and CFC-12 were significantly lower inside termite mounds than in the ambient air, suggesting that the soils were removing CFC-11 and CFC-12 from the atmosphere (*10*). Furthermore, a recent report tentatively concluded that there may have been consumption of CFC-11 and CFC-12 from an enclosure placed over rice paddy soil (*11*). CFC-11 and CFC-12 were consumed in laboratory incubations of methanogenic sediments (*12*) and in bioreactors (*13*). The finding that, with titanium citrate as the reductant, corrinoids could catalyze the reductive dehalogenation of superatmospheric concentrations of CFC-11 and CFC-12 (*14*) further suggests that CFC-11 and CFC-12 might not be stable in anaerobic environments.

The environmental significance of this previously unrecognized sink for CFC-11 and CFC-12 has been difficult to evaluate because of a paucity of measurements and a lack of understanding of the mechanisms for CFC-11 and CFC-12 uptake. The purpose of this study was to determine the potential for anaerobic microorganisms to consume atmospheric CFC-11 and CFC-12.

Materials and Methods

Anaerobic Sediments. Freshwater sediments from the Potomac River, MD, were collected as previously described (*15*) and transferred (90 mL of wet sediment) under strict anaerobic conditions into glass serum bottles (160 mL), which were then sealed with thick butyl rubber stoppers. The atmosphere was N_2/CO_2 (93:7). In some instances,

sodium sulfate (10 mM) or poorly crystalline iron(III) oxide (100 mmol/L) was added from concentrated anaerobic stocks, as previously described (*16*), in order to convert the terminal electron-accepting process from methanogenesis to sulfate reduction or Fe(III) reduction. Microbial activity in some bottles was inhibited by heating them to 121 °C for 1 h on 3 consecutive days. An anaerobic standard (0.15 mL) that contained 1.3×10^{-6} atm CFC-12 and 1.4×10^{-6} atm CFC-11 in N_2 was added to the headspace. This provided initial concentrations of CFC-11 and CFC-12 that were ca. 3–5-fold higher than those in air. The bottles were shaken to equilibrate the CFC-11 and CFC-12 in the headspace with the porewater and then incubated upside down to prevent contact between the headspace and the stopper. Incubations were in the dark at 20 °C with the exception of the temperature-optimum experiment in which a range of incubation temperatures was employed. The headspace was sampled over time and analyzed with gas chromatographs equipped with an electron capture detector for CFC measurements and with a reduction gas analyzer for methane measurements. CFC-11 and CFC-12 were separated on a column ($^1/_8$-in. inner diameter, 4 ft) of Carbopack B (60/80 mesh, Supelco Inc.) at 70 °C with argon/methane (95:5) as the carrier at 60 mL/min.

In order to generate oxidized, heat-killed sediments, the sediments were stirred under air (*17*) to remove reduced components prior to transferring into anaerobic serum bottles and treating with heat as above. For the oxidized, heat-killed sediments and the studies with empty bottles, 0.06 mL of the CFC-11 and CFC-12 standard was added, and the incubation temperature was 30 °C.

Studies with sediments and soils from other sites were conducted the same as with the Potomac River sediments, with the exception that the aerobic soil was incubated under air. Sediment volumes were 90 mL, except for sediments from urban sites, which were 70 mL. Aerobic soil, urban cattail marsh, and urban pond sediments were collected in Virginia; salt marsh and rural cattail marsh sediments were collected in Maryland; swamp sediments were from South Carolina.

Defined Systems. *Clostridium pasteurianum* (ATCC 6013) and *Escherichia coli* (ATCC 4157) were obtained from the American Type Culture Collection, Rockville, MD. In order to investigate CFC-11 and CFC-12 uptake by anaerobic microorganisms under more defined conditions, a heavy suspension (1 mL) of *C. pasteurianum* or *E. coli* was inoculated into 26-mL anaerobic pressure tubes containing 10.5 mL of anaerobic medium (*18*) with glucose (50 mM) as the electron donor. In the results shown here, this provided an initial concentration of 0.6 mg of *C. pasteurianum* cell protein per milliliter. Where noted, cells were autoclaved (121 °C, 15 min) after addition to the medium and prior to addition of the CFC-11 and CFC-12.

Studies with hematin were conducted as previously described (*19*), with the exceptions that dissolved oxygen was removed from all solutions by bubbling with N_2 and the bottles were sealed with butyl rubber stoppers. Hematin (1 mg) or autoclaved (121 °C, 15 min) hematin was added to 100 mL of 0.1 M K_2HPO_4 buffer (pH 7.0) in 160-mL serum bottles with cysteine (0.1 M) included as the reductant, where noted.

Initial concentrations of CFC-12 were ca. 1×10^{-9} atm and 1.7×10^{-9} atm for studies with *C. pasteuranium* and hematin, respectively. Initial CFC-11 concentration for hematin studies was ca. 1×10^{-9} atm. The incubation temperature was 30 °C.

Results and Discussion

Methanogenic Potomac River Sediments. CFC-12 in the gas phase overlying the Potomac River sediments was consumed over time, but there was no CFC-12 uptake when the microorganisms in the sediment were killed with heat prior to the incubation (Figure 1). After 33 days of incubation, some of the heat-killed sediments were inoculated with microorganisms by injecting 3 mL of an anaerobic sediment slurry (Figure 1). With the addition of microorganisms, CFC-12 was consumed after a brief lag which corresponded with a lag in the initiation of methane production. These results indicated that CFC-12 uptake in the sediments required active microbial metabolism.

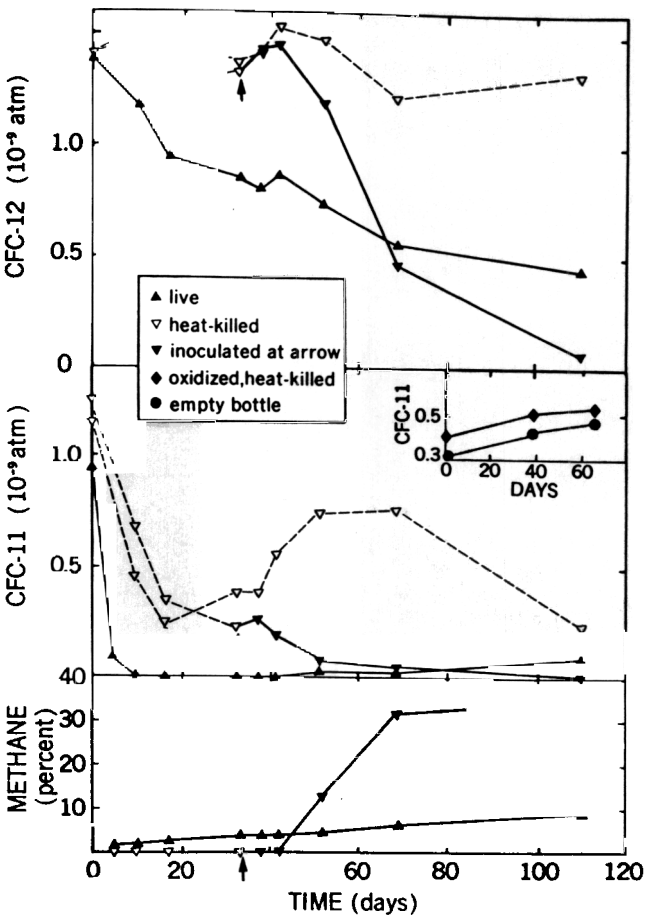

Figure 1. Concentrations of CFC-12, CFC-11, and methane over time in the gas phase overlying anaerobic freshwater sediments from the Potomac River. At 33 days of incubation (arrow) some of the heat-killed sediments were inoculated with microorganisms by injecting 3 mL of an anaerobic sediment slurry. The results of one representative bottle of triplicate incubations are shown.

Once methane production and CFC-12 uptake began in the heat-killed sediments that had been inoculated with microorganisms, the rates of methane production and CFC-12 uptake were faster than in the untreated sediments. The higher rates of methane production presumably were because the prior heat treatment had made some of the recalcitrant organic matter more available for microbial decomposition. The finding that CFC-12 was consumed at faster rates when the rate of organic matter decomposition was faster further suggests that CFC-12 uptake was a consequence of microbial metabolism.

The temperature optimum for CFC-12 uptake was also characteristic of a microbially catalyzed reaction (Figure 2). The rate of CFC-12 uptake increased with increasing incubation temperatures up to 30 °C. However, the CFC-12 consumption was progressively slower as temperatures were increased above 30 °C. Such a response is consistent with an enzymatically catalyzed reaction

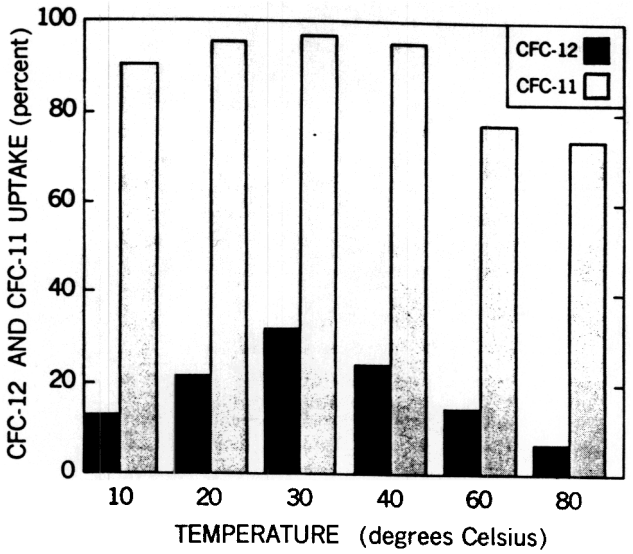

Figure 2. Extent of CFC-12 and CFC-11 uptake in Potomac River sediments for 14 days at the designated temperatures. Values are means of triplicate determinations. Initial concentrations of CFC-11 and CFC-12 were ca 1×10^{-9} and 2×10^{-9} atm, respectively.

whereas a nonenzymatic reaction would be expected to proceed faster as the temperature was raised above 30 °C.

The sediments also consumed CFC-11 (Figure 1). However, studies on CFC-11 uptake were complicated by the fact that the bottle stoppers released CFC-11 into empty bottles (Figure 1, inset) at a rate which is estimated to have added ca. 6×10^{-12} atm/day to the headspace overlying the sediments. CFC-11 uptake was faster in live sediments than in heat-killed sediments, and inoculation of heat-killed sediments with microorganisms stimulated CFC-11 uptake. However, in contrast to CFC-12, CFC-11 was also consumed in heat-killed sediments. Temperatures higher than 40 °C slightly inhibited CFC-11 uptake, but there was still considerable CFC-11 uptake at 80 °C (Figure 2). These results suggest that there was a nonenzymatic as well as an enzymatic component to CFC-11 uptake. The nonenzymatic CFC-11 uptake in the anaerobic, heat-killed sediments appeared to be the result of interaction(s) with reduced components in the sediments as there was no CFC-11 uptake in heat-killed sediments that had been oxidized by mixing them under air for several days prior to the anaerobic incubation (Figure 1, inset).

Other Sediments and Soils. As expected from previous studies which have indicated that aerobic soils do not consume CFC-11 or CFC-12 (*20*), an organic-rich, aerobic forest soil incubated under air did not consume CFC-11 or CFC-12 (Figure 3). However, a wide variety of anaerobic soils and sediments did (Figures 3 and 4). CFC-11 uptake was consistently faster than CFC-12 uptake. Potomac River sediments converted to sulfate reduction or Fe(III) reduction consumed CFC-11 and CFC-12 at rates as fast or faster than those observed in methanogenic sediments (Figure 3). Salt marsh sediments in which sulfate reduction was the terminal electron-accepting process also had active CFC-11 and CFC-12 uptake. These results indicate that methane production is not required for CFC-11 and CFC-12 consumption.

Rates of CFC-11 and CFC-12 uptake varied between sediments (Figure 3). Even sediments from two different cattail marshes differed more than 2-fold in the rate of CFC-12 uptake. There was no direct relationship between the rates of CFC-11 and CFC-12 consumption. For example, swamp sediments from South Carolina (Figure 4) consumed CFC-11 at rates comparable to those observed in the Potomac River (Figure 1), but the rates of F-12

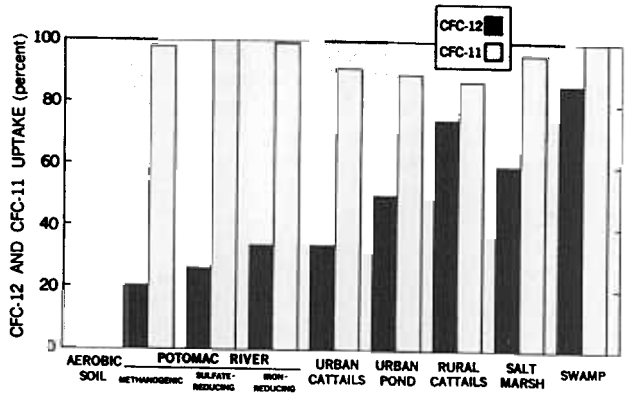

Figure 3. Relative extent of CFC-12 and CFC-11 uptake in various aquatic sediments and soils after 1 month of incubation. Values are means of triplicate determinations. Initial CFC-11 concentrations ranged from ca. 6×10^{-10} to 2×10^{-9} atm. Initial CFC-12 concentrations ranged from ca. 6×10^{-10} to 4×10^{-9} atm.

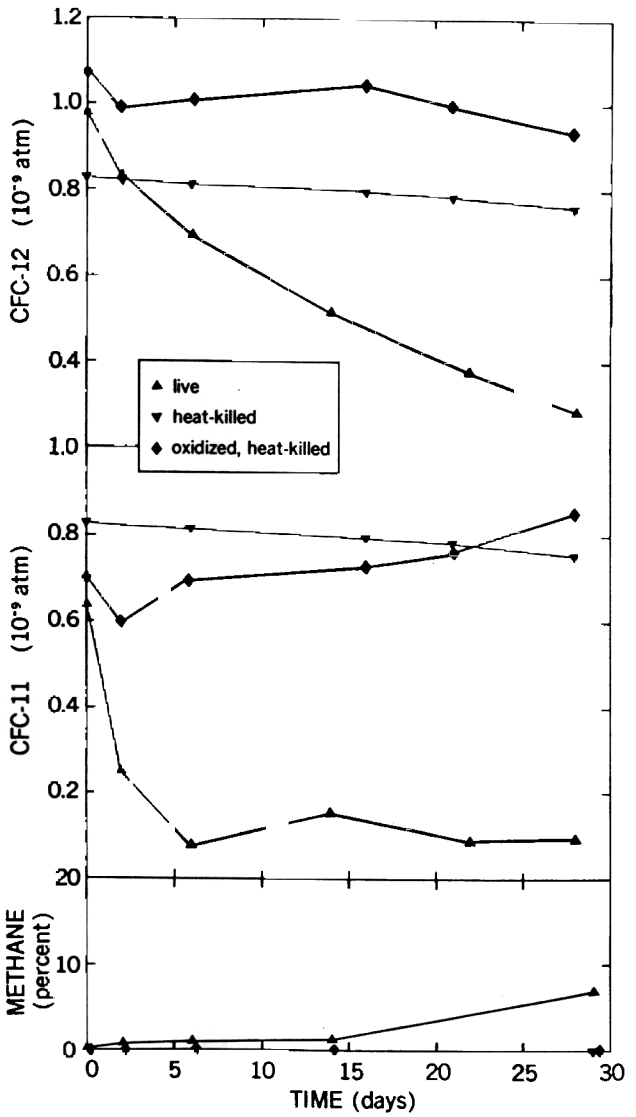

Figure 4. Concentrations of CFC-12, CFC-11, and methane over time in the gas phase overlying anaerobic freshwater sediments from a swamp located in South Carolina. The results of one representative bottle of triplicate incubations are shown.

uptake were ca. 3–5-fold faster in the swamp sediments than they were in the river sediments.

In contrast to the Potomac River sediments, there was no consumption of CFC-11 in heat-killed swamp sediments

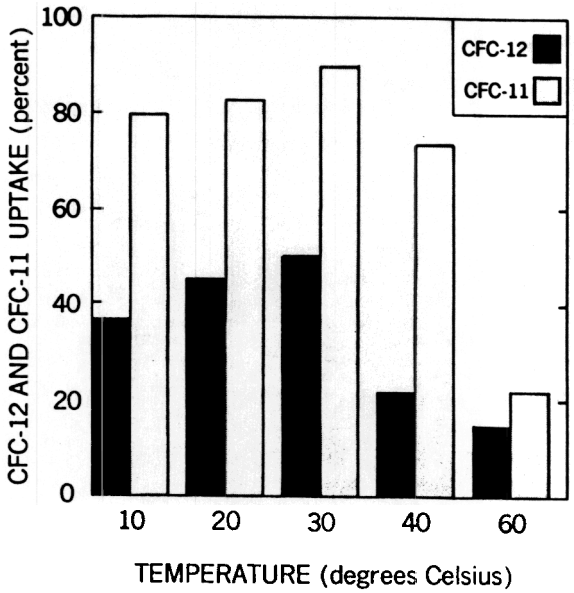

Figure 5. Extent of CFC-12 and CFC-11 uptake in swamp sediments incubated for 14 days at the designated temperatures. Values are means of duplicate determinations. Initial concentrations of CFC-11 and CFC-12 were ca. 8×10^{-10} and 1×10^{-9} atm, respectively.

(Figure 4). Temperature optimum studies also indicated that CFC-11 uptake in the swamp sediments was primarily dependent upon enzymatic activity (Figure 5). However, with the exception of the sediments from the urban pond, CFC-11 was consumed in heat-killed sediments from all of the other sources shown in Figure 3. As with the Potomac River sediments, the heat treatment inhibited CFC-12 uptake in all of the sediments examined, and in the instances examined (urban cattails and pond), reinoculation of the heat-killed sediments with live sediments resulted in CFC-12 uptake.

Defined Systems. In order to investigate whether the metabolism of anaerobic microorganisms could result in CFC-11 and CFC-12 uptake, *C. pasteurianum*, a typical anaerobic fermentative microorganism, was grown in the presence of CFC-11 and CFC-12 (Table I). CFC-12 was readily consumed in the presence of cells but not in sterile medium. CFC-12 uptake was inhibited when the cells were killed with heat prior to the incubation. CFC-11 uptake could not be quantified because an unknown component from the medium interfered with the chromatographic analysis. Similar studies with *E. coli* did not result in CFC-11 or CFC-12 consumption.

CCl_4 can be dechlorinated through nonenzymatic reactions in which heat-stable metalloporphyrins or other reduced components produced by microorganisms react with CCl_4 (*14, 19, 21–23*). When CFC-11 was incubated in the presence of the iron porphyrin, hematin, as well as a reductant to maintain the iron in a reduced state, CFC-11 was consumed (Table I). Even after being autoclaved, hematin in the reduced state consumed CFC-11. Hematin without a reducing agent did not consume CFC-11, nor did the reductant alone. There was no loss of CFC-12 in the presence of reduced or oxidized hematin. These results provide a model for the nonenzymatic uptake of CFC-11 in the heat-killed sediments.

Implications. The results suggest that, in anaerobic environments, CFC-11 and CFC-12 may be degraded in a manner similar to that previously described for the structurally similar CCl_4. CCl_4 is also relatively stable under aerobic conditions but, under anaerobic conditions, a wide variety of microorganisms can degrade it (*21–22, 24–29*). CCl_4 may be dechlorinated to various extents

Table I. Consumption of CFC-11 or CFC-12 in the Presence of *Clostridium pasteurianum* or Hematin

additions to	uptake over 24 h, % CFC-12	CFC-11
glucose medium		
C. pasteurianum	58.2 ± 7.7[a]	ND[b]
autoclaved cells	22.7 ± 11.7	ND
none	2.0 ± 0.5	ND
phosphate buffer		
hematin and cysteine	0	35.2 ± 4.3
autoclaved hematin and cysteine	0	28.0 ± 16.8
cysteine	0	0.3
hematin	0	0

[a] Values are mean ± standard deviation (n = 3), except for measurements with cysteine alone in which there were only 2 replicates. [b] CFC-11 could not be determined (ND) because an unknown component interfered with the chromatographic analysis. Initial concentrations of CFC-12 were ca. 1×10^{-9} atm and 1.7×10^{-9} atm for studies with *C. pasteurianum* and hematin, respectively. Initial CFC-11 concentration for hematin studies was ca. 1×10^{-9} atm.

through various enzymatic and nonenzymatic pathways to form $CHCl_3$, CH_2Cl_2, CH_3Cl, CH_4, CO, or CO_2 (*14, 19, 21, 23–25, 29*). No intermediates in CFC-11 and CFC-12 degradation were observed with the electron capture detector during the sediment incubations. After the studies reported here were completed, it was reported that, with titanium citrate as the reductant and CFC concentrations ca. 10^7 above atmospheric levels, corrinoids could catalyze the reductive dehalogenation of CFC-11 and CFC-12 with CO as the major end product (*14*). Any CO produced from the low concentrations of CFC-11 and CFC-12 employed in our study would not be detectable over the steady-state pool sizes of CO that are normally present in anaerobic sediments.

The stability of CFC-11 and CFC-12 under aerobic conditions has led to fruitful application of these compounds as conservative tracers of surface waters and groundwaters (*8,9*). However, the findings reported here suggest that caution be applied when one attempts to use these compounds as inert tracers in anaerobic environments. There is preliminary field evidence to support this conclusion. In the acetate-amended, anaerobic groundwater of a shallow, confined aquifer, 68% of the CFC-11 in the groundwater was removed within 2-m travel (**30**). Futhermore, the anaerobic groundwaters of a shallow aquifer in South Carolina that has been contaminated with jet fuel has significantly lower concentrations of CFC-11 and CFC-12 than nearby, uncontaminated aerobic groundwater (D. Vroblesky and J. Woodward, unpublished data). Degradation products of CFC-113 ($CFCl_2CF_2Cl$) such as $CHFClCF_2Cl$, $CFCl_2CHFCl$, and $CFClCF_2$ have been observed in anoxic groundwaters contaminated with CFC-113 (*31*).

CFC-11 and CFC-12 contained within refrigeration equipment or trapped in insulating foams may be disposed in anaerobic environments such as landfills. The potential for anaerobic degradation of CFC-11 and CFC-12 suggests that as this CFC-11 and CFC-12 leaches out of its containers, biological activity may prevent some of the waste CFC-11 and CFC-12 from entering the atmosphere.

In accordance with a recent report (*11*), the results presented here also suggest that CFC-11 and CFC-12 that have entered the atmosphere may be consumed in environments such as natural wetlands and rice paddies which contain anaerobic sediments in close proximity to the atmosphere. However, it would be premature to attempt to extrapolate global rates for uptake of atmospheric

CFC-11 and CFC-12 from the measurements reported here. Uptake rates in different sediments varied significantly even when the sediments were collected from a small geographic area and were incubated under similar conditions in the laboratory (Figure 3). There is also no indication of how rates of uptake in the bottle incubations compare with in situ rates in undisturbed sediments.

Furthermore, other anaerobic environments could contribute to CFC-11 and CFC-12 uptake. In fact, the studies reported here were initially stimulated by a report (*10*) which suggested that Australian termite mounds were consuming CFC-11 and CFC-12 from the atmosphere. Each of the mounds contained ca. 500 000 termites (*10*). The guts of termites are known to harbor a complex assemblage of anaerobic microorganisms (*32*) which might account for the CFC-11 and CFC-12 consumption within the termite mounds. Termites and other intestinal anaerobic microbial environments, such as those found in ruminants, are widespread and are already known to have a significant impact on the global methane budget (*33*).

The continued accumulation of CFC-11 and CFC-12 in the troposphere (*34*) demonstrates that CFC-11 and CFC-12 uptake in anaerobic habitats is minor in comparison to the current anthropogenic release of these compounds. Current global atmospheric models have large uncertainties in the estimated CFC-11 and CFC-12 tropospheric lifetimes (*34*, *35*). The extent of CFC-11 and CFC-12 uptake in anaerobic environments is probably much smaller than the calculated errors in current tropospheric lifetime estimates and thus would not be detected by such models. However, because of the long tropospheric lifetimes of CFC-11 and CFC-12 [ca. 100 years (*34*)], mechanisms which remove even a minor fraction of the tropospheric CFC-11 and CFC-12 will have a significant long-term impact on the amount of CFC-11 and CFC-12 which reaches the stratosphere (*10*, *36*). Thus, as models of CFC-11 and CFC-12 tropospheric lifetimes become more refined, and as the anthropogenic release of CFC-11 and CFC-12 is reduced, the extent of CFC-11 and CFC-12 uptake in anaerobic environments may be an important consideration for estimating long-term depletion of stratospheric ozone.

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Registry No. CFC-12, 75-71-8; CFC-11, 75-69-4; hematin, 14875-96-8.

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