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Production Potential of Greenhouse Gases Affected by Microplastics at Freshwater and Saltwater Ecosystems

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Abstract: Currently, microplastic pollution poses a great threat to diverse ecosystems. Microplastics can potentially change soil characteristics and impact soil microorganisms, and then affect the production of CO_2, CH_4 and other greenhouse gases. However, experimental study on different ecological soils is lacking. Herein, we experimentally analyzed the CO_2 and CH_4 production potential affected by four types of microplastics in freshwater (Poyang Lake in Jiangxi province, paddy soil in Hunan province) and saltwater (Salt marsh in Shandong province, mangrove soil in Fujian province) ecosystems. Microplastics promoted CO_2 production, of which polyethylene terephthalate (PET) had the greatest impact. In our study, the microplastics that had the greatest impact on CH_4 concentration emissions were high-density polyethylene (1276 umol·g^{-1}·L^{-1}), followed by polyvinyl chloride (384 umol·g^{-1}·L^{-1}), polyethylene terephthalate (198 umol·g^{-1}·L^{-1}), and polyamide (134 umol·g^{-1}·L^{-1}). In addition, the largest impact on CO_2 concentration emissions was displayed by polyethylene terephthalate (2253 umol·g^{-1}·L^{-1}), followed by polyvinyl chloride (2194 umol·g^{-1}·L^{-1}), polyamide (2006 umol·g^{-1}·L^{-1}), and high-density polyethylene (1522 umol·g^{-1}·L^{-1}). However, the analysis results based on one-way ANOVA showed that CO_2 emission was most significantly affected by soil properties rather than microplastics types. In comparison, the influencing factor on CH_4 production changed from soil types to the interaction between soil types and microplastics, and finally to the microplastics with the increase in incubation time. Further, by comparing CO_2 and CH_4 production and Global Warming Equivalent (GWE) affected by microplastics, freshwater ecosystems were more sensitive than saltwater. For all the soil types used in this study, high-density polyethylene had the greatest impact on CH_4 production potential. In conclusion, our study provided basic data for further understanding the effects of microplastics on soil greenhouse gas emissions from different sources.

Keywords: freshwater ecosystem; coastal ecosystem; soil; microplastics; CO_2; CH_4

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1. Introduction

Greenhouse gases (GHGs) are one of the important drivers of global climate change [1]. GHGs emitted by soil mainly include CO_2 and CH_4. According to the IPCC (Intergovernmental Panel on Climate Change), the increase in CO_2 concentration greatly affects the function and nature of terrestrial ecosystems [2]. Methane is the second largest greenhouse gas after CO_2, and although its atmospheric concentration is only 0.45% of CO_2, its warming potential is 28–34 times higher [3]. According to recent statistics (2008–2017) [4], greenhouse gas emissions are mainly from paddy fields, wetlands, oceans and the exploitation and combustion of fossil fuels. Studies have shown that natural factors include climate [5], vegetation [6], substrate availability [7], temporal and spatial differences, and anthropogenic factors including aerosols, nitrogen deposition, microplastics, and can all affect GHG production.

Furthermore, some substances added by endogenous and anthropogenic activities in the soil can affect greenhouse gas emissions. Examples include biochar, magnetite, and microplastics [8–11]. As a spongy substance, biochar can adsorb and store organic mineral nutrients for further feeding by microorganisms [4]. Studies have shown that biochar is beneficial for methane production due to its characteristics of microbial immobilization, pH buffering, availability of metal ions control and enzymatic processes [12–15]. Microbial immobilization is controlled by the adsorption capacity of the material, which in turn depends on the surface morphology, pore volume and size, hydrophobicity, and ion exchange capacity of biochar [16–18]. Especially on the rough surface with higher specific surface area [19], volatile fatty acids (VFA) are convenient to form biofilms [20], which provides sufficient feeding substrate for methanogenic archaea [21]. There was a positive correlation between pH value and pyrolysis temperature of biochar [22], and biochar synthesized below 700 °C increased methanogenesis [23]. In addition, biochar can alleviate acid and ammonium stress, which is conducive to the colonization of methanogens such as methanococcus, thus promoting the production of CH_4 in anaerobic environments [24]. In fact, there are three known CH_4 production pathways: hydrotropic methanogenesis (MIET-CO_2, DIET-CO_2), acetoclastic methanogenesis, and Methyl-trophic methanogenesis [25–27]. Magnetite is a common iron-bearing mineral with high electrical conductivity. It can improve CH_4 production of direct interspecific electron transfer (DIET) by accelerating intracellular electron transfer, interspecies electron exchange and enhancing redox capacity [28]. Currently microplastic pollution as some exogenous substance poses a great threat to diverse ecosystems.

Microplastics are an important contributor to the greenhouse gas cycle. For example, the presence of polyethylene terephthalate (PET) can affect CH_4 emissions. According to Han et al. [29], the addition of PET will increase soil redox potential (Eh). Due to the improvement of soil Eh, the abundance of pmoA and pmoB involved in CH_4 oxidation increased, while the abundance of mcrA and mcrB involved in CH_4 formation decreased. Moreover, microplastics can affect CO_2 emissions by influencing the microbial community structure in sediments [30–32]. In polyethylene-contaminated soil, bacterial abundance was much higher than fungal abundance [33]. Low molecular weight polyethylene (LDPE) can pass through the biofilm of microorganisms, and as such it can be absorbed and degraded by microorganisms [34]. High molecular weight polyethylene microplastics cannot directly cross the cell membrane into microorganisms due to the large molecular chain. Under the action of a series of abiotic factors such as light, temperature and biological factors, they are gradually transformed into small molecules, and the degraded small molecules or ethylene monomers are absorbed and degraded by microbial cells to generate CO_2 and CH_4, etc. [35]. Therefore, the impact of microplastics in soil on greenhouse gas emissions cannot be ignored.

At present, the research content of microplastics covers many fields. Most microplastics research focuses on oceans [36–38], beach sediments [39–41] and terrestrial waters [42,43]. The results of these studies attracted wide public attention. By contrast, terrestrial systems, especially soils, remain to be studied [44–46]. In particular, the influences of microplastics and the ecological environment on CH_4 and CO_2 are not clear yet. In this study, four kinds of microplastics (HDPE, polyvinyl chloride (PVC), polyamide (PA), PET), two saltwater (salt marsh, mangrove, and two freshwater (paddy soil, Poyang Lake)) were selected to investigate the effects of microplastics on CH_4 and CO_2 production in soils. As the largest freshwater lake in China, the submerged plant area in Poyang Lake is in an anaerobic environment all year round. Its distribution area accounts for nearly 50% of the total vegetation of Poyang Lake, which is regarded as an important source of CH_4 release [47]. Rice paddies are a complex ecosystem that both emits CH_4 and absorbs CO_2, and they play an important role in the global water–carbon cycle and carbon balance [48]. The Yellow River Delta wetland is a unique ecosystem due to the multiple influences of saline environment, freshwater environment and tidal action at the same time, and it is of great theoretical importance to study this area to reduce CH_4 release from coastal wetlands [49,50]. Mangroves, as one of the most carbon-intensive forest ecosystems, play an important role in the carbon biogeochemical cycling process. Therefore, we hypothesize that microplastics are a non-negligible influencing factor for greenhouse gas emissions in different ecosystems, and then proceed to collect soils from for representative sampling sites to study the impact of microplastics on their greenhouse gas emissions.

2. Materials and Methods

2.1. Samples Collecting

Soil used in this experiment was collected from the Dahuchi Conservation Station of Poyang Lake, the National Nature Reserve in Jiangxi (PY: 29°8′4.3″ N–29°8′6.79″ N, 115°58′56.09″ E–115°59′1.38″ E) [51], the Long-term Positioning Test Station of Ningxiang County from Hunan Province (PS: 28°07′ N, 112°18′ E) [52], from the Yellow River Delta Field Observation and Research Station of Coastal Wetland Ecosystem, Dongying city, Shandong Province (CJ: 37°34′45″ N, 118°56′39″ E) [53], and Minjiang Estuary Wetland (HS: 26°2′0″ N, 119°37′0″ E) [54]. The sedimentary material land of Poyang Lake is mainly divided into sandy soil and sandy loam, and the soil as a whole is alkaline [47]. The collected paddy soils were slab shale-developed paddy soils [55], the planting system used was an early rice—late rice winter leisure mode, and the soil nitrogen, phosphorus and potassium supply conditions at the sampling points were based on the expected crop demand [56]. The soils of a salt marsh are different due to the different parent matter and land formation time, and the distribution of fluvo-aquic and saline soils is the most widespread [49]. In mangroves, the pH of sediment pore water in tidal flat profile was basically neutral, and the frequency of flooding and electrical conductivity gradually increased. There is great spatial heterogeneity of iron and sulfur in high-, middle- and low-tidal flat profiles [57]. Four microplastics were applied to the soils: high-density polyethylene powder (HDPE), polyvinyl chloride (PVC), polyamide powder (PA), and polyethylene terephthalate (PET).

2.2. Experimental Process

The soils were mixed with water at a ratio of 1:4 (m:v), while impurities such as plant roots were removed. Nitrogen was injected to eliminate the internal air. The microcosm experiments consisted of a 5 mL soil–water mixture and 0.01 g microplastics (glass beads were added as control) in 12 mL serum vials. In order to ensure the uniformity of microplastics in the soil, shock and nitrogen blowing were used to distribute the microplastics evenly in the soil sample. Three cycles of vacuum/charging high-purity N_2 were applied to the system to create anaerobic conditions. All the experiments were performed in triplicate and kept at 30 °C in the dark without shaking.

On the basis of previous studies on GWP [3,58,59], this paper analyzed GWE. The gas concentration measured was calculated to obtain global warming equivalent (GWE, i.e., CO_2 equivalent), which was used to study the production of CO_2 and CH_4 under the condition of adding microplastics and to make a simple analysis of the influencing factors of global climate change. GWE of different ecosystems was calculated using the following formula (Formula (1)):

$$GWE = C_{CO2} \times 1 + C_{CH4} \times 34 \quad (1)$$

C_{CO2} represents the concentration of CO_2, and C_{CH4} represents the concentration of CH_4.

2.3. Statistical Analyses

The data analyses are implemented using the SPSS 26.0 software. To investigate the effects of different ecosystem soil types and microplastics on greenhouse gas production, one-way ANOVA and Tukey's honestly significant difference (HSD) test were performed. The graphs were drawn in Origin 2021.

3. Results and Discussion

3.1. Production of Greenhouse Gases in Freshwater Ecosystems Affected by Microplastics

3.1.1. Concentration of CO_2 in Poyang Lake and Paddy Soil

In the freshwater ecosystem, HDPE significantly increased soil CO_2 production from 1468 umol·g^{-1}·L^{-1} to 2713 umol·g^{-1}·L^{-1} within 49 d to 70 d in Poyang Lake. At the same time, PA and PET significantly increased the CO_2 production concentration of paddy soil on 49 d, after which it then decreased (Figure 1). Machado et al. [60] studied the effect of different types of microplastics on soil enzymes, finding that the addition of HDPE increased the activity of FDA hydrolase, while PET, PA, PS, etc. reduced the activity of microorganisms. Compared to the impact of various microplastics on CO_2, the effects of soil type on CO_2 were more significant (Table 1), and significant at the 70 days of culture except for 49 d ($o < 0.01$). Studies have shown that the main factors affecting soil CO_2 emission flux are soil temperature and soil moisture content [32]. For example, in the study of Poyang Lake wetland, the increase in soil water content will increase soil organic carbon content [61]. Anaerobic environment slows down microbial activity, reduces the utilization of activated organic carbon by microorganisms, and reduces carbon output [62]. Therefore, in freshwater ecosystems, under the condition of adding microplastics, the soil still has a greater impact on CO_2 production, and CO_2 is not sensitive to microplastics.

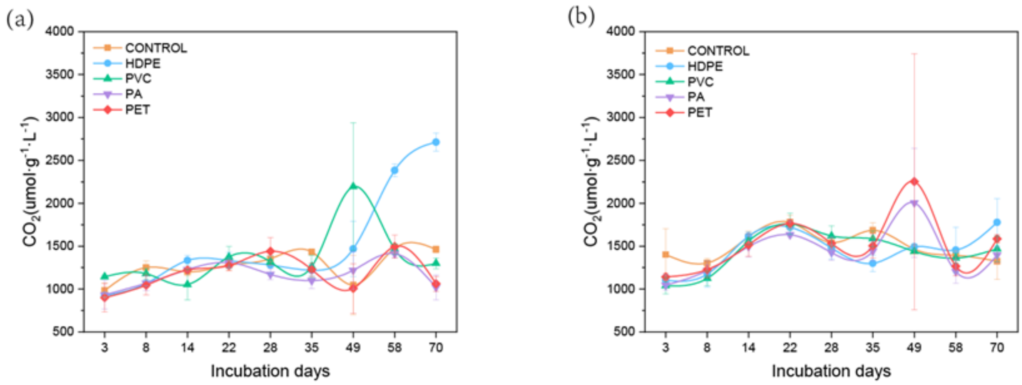

Figure 1. CO_2 concentrations in freshwater ecosystems after microplastics (HDPE, PVC, PA, PET) added. (**a**) Poyang Lake. (**b**) paddy soil.

Table 1. One-Way ANOVA of soil and microplastics on CO_2 yield.

Factors		Incubation Days								
		3	8	14	22	28	35	49	58	70
soil	F	22.786	9.551	20.834	20.574	14.064	16.234	2.402	26.778	13.032
	P	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.078	<0.01	<0.01
microplastics	F	0.565	1.663	1.087	1.318	1.221	2.111	0.174	11.045	10.621
	P	0.690	0.169	0.370	0.272	0.311	0.090	0.951	<0.01	0.010
soil × microplastics	F	0.779	0.446	0.964	0.363	1.208	1.450	1.210	4.020	7.348
	P	0.637	0.938	0.492	0.972	0.298	0.168	0.302	<0.01	<0.01

The F is the statistic of the F test, which is the ratio of the sum of squared deviations between and within groups to the degrees of freedom. The P means that the p value is the probability of a sample observation or a more extreme outcome when the null hypothesis is true. The smaller the p value, the more obvious the result. $p < 0.01$ indicates a significant difference.

3.1.2. Concentration of CH_4 in Poyang Lake and Paddy Soil

In Poyang Lake soil, the concentration of CH_4 was lower than 8.704 umol·g^{-1}·L^{-1} in the first 35 d of culture. When HDPE was added and cultured for 49 d, the concentration increased rapidly, and the highest concentration reached 1276 umol·g^{-1}·L^{-1} on 70 d (Figure 2a). In paddy soil, similar to Poyang Lake, the variation of CH_4 concentration was small, except for HDPE addition (Figure 2b). Paddy fields are one of the important sources of methane emissions, accounting for about 10% to 20% of the global total annually [38]. Studies have shown that soil characteristics and climatic factors are the main factors affecting CH_4 emission from paddy soils [6,63–65]. Table 2 showed that soil had a significant effect on CH_4 during the first 14 d of cultivation ($p < 0.01$). The source and abundance of microplastics are closely related to human activities [66]. Due to intensive and complex human activities in the Poyang Lake basins, microplastics in sediments are mainly debris [38]. With the extension of culture time, the significant influencing factors of CH_4 gradually changed from soil type to interaction between soil and microplastics, and finally, microplastics had the greatest influence (Table 2). Therefore, in the freshwater ecosystem, CH_4 was more sensitive to the addition of HDPE in Poyang Lake soil, but with a delayed response.

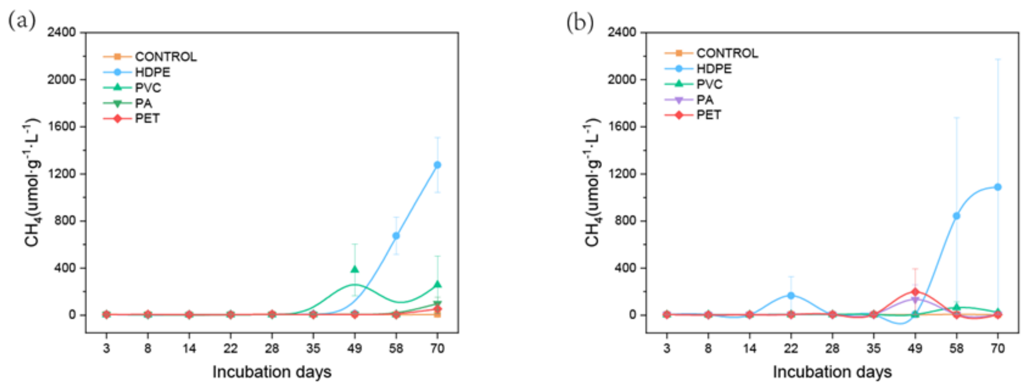

Figure 2. CH_4 concentrations in freshwater ecosystems after microplastics (HDPE, PVC, PA, PET) were added. (**a**) Poyang Lake (**b**) paddy soil.

Table 2. One-Way ANOVA of soil and microplastics on CH_4 yield.

Factors		Incubation Days								
		3	8	14	22	28	35	49	58	70
soil	F	164.071	18.658	8.898	1.154	2.340	5.823	1.728	1.386	2.591
	P	<0.01	<0.01	<0.01	0.334	0.081	0.001	0.170	0.254	0.069
microplastics	F	3.779	1.264	0.441	1.267	1.394	4.556	1.295	3.571	4.637
	P	0.008	0.293	0.778	0.291	0.245	0.003	0.281	0.011	0.002
soil × microplastics	F	5.139	2.922	1.002	1.105	2.502	4.981	2.192	1.217	1.575
	P	<0.01	0.003	0.457	0.371	0.009	<0.01	0.022	0.290	0.120

The F is the statistic of the F test, which is the ratio of the sum of squared deviations between and within groups to the degrees of freedom. The P means that the p value is the probability of a sample observation or a more extreme outcome when the null hypothesis is true. The smaller the p value, the more obvious the result. $p < 0.01$ indicates a significant difference.

3.2. Concentrations of Greenhouse Gases in Saltwater Ecosystems

3.2.1. Concentration of CO_2 in Intertidal and Mangrove Forests

In the intertidal soil, the CO_2 concentration increased rapidly after the third day of self-cultivation, and then the CO_2 concentration fluctuated between 548 umol·g^{-1}·L^{-1}–1446 umol·g^{-1}·L^{-1} (Figure 3a). The mangrove soil also fluctuated with the incubation days, and the CO_2 concentration ranged from 979 umol·g^{-1}·L^{-1} to 1534 umol·g^{-1}·L^{-1} (Figure 3b). As in freshwater ecosystems, CO_2 production was related to soil type, and was not significantly affected by the addition of microplastics. The distribution, transfer and accumulation of microplastics had little influence on the sampling sites. In addition, according to data analysis in this study, microplastics have no significant impact on the salt marsh and mangrove soil (Table 1). Soil texture significantly affects soil greenhouse gas emissions [66]. Therefore, CO_2 in saltwater ecosystem is not sensitive to microplastics, like when it is in a freshwater ecosystem, and is greatly affected by soil type.

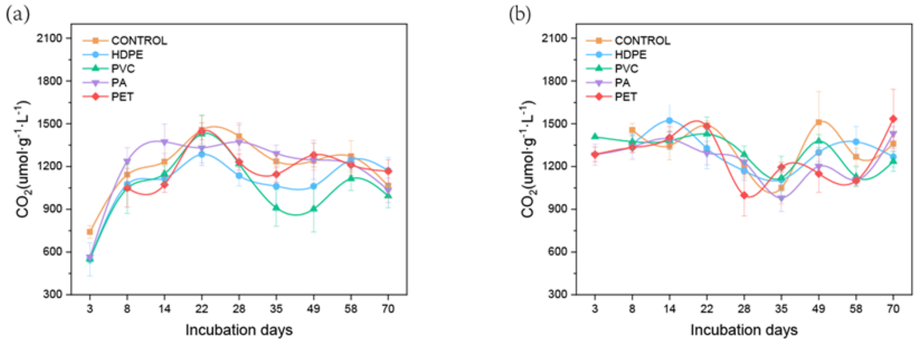

Figure 3. CO_2 concentration in saltwater ecosystem after microplastics (HDPE, PVC, PA, PET) were added. (**a**) Salt marsh (**b**) mangrove forest.

3.2.2. Concentration of CH_4 in Intertidal and Mangrove Forests

The concentrations of CH_4 were lowest in saltwater ecosystems. Compared with mangroves, the addition of HDPE had an effect on CH_4 concentration in salt marsh, and the concentration increased the most at the 35th day of culture (Figure 4a). The concentration of CH_4 produced by mangrove soil after microplastics addition fluctuated little and produced low concentration (Figure 4b). CH_4 emissions from intertidal wetlands are affected by plant biomass, water level, temperature, salinity and SO_4^{2-} content [65]. It was found that microplastics affected the process of soil methane production to a certain extent [6,67]. In particular, microplastics, as carriers of other pollutants, have large comparative area and strong hydrophobicity, all of which can interfere with or inhibit methanogenesis in collaboration with other pollutants [68,69]. For example, PVC microplastics can dissolve bisphenol A in the anaerobic digestion process, thus weakening the reaction activity of key enzymes in the reaction system and inhibiting the process of sludge hydrolysis, acidification and CH_4 production [70]. As an important sink of microplastics and pollutants, mangroves produce complex pollution from combined environmental pollutants on their surfaces in addition to the plastic particles themselves [41]. Mangrove wetlands, as coastal zone wetlands, contain a certain amount of salinity in the tidal water, and salinity further affects CH_4 fluxes by influencing the activity of methanogens [71]. A study by some scholars found that CH_4 fluxes in marshes were significantly inhibited above a certain range of salinity (salinity > 18×10^{-3}). In this study, the addition of HDPE has a certain degree of influence on the CH_4 production of intertidal soil in saltwater ecosystems, while mangroves are insensitive to microplastics addition.

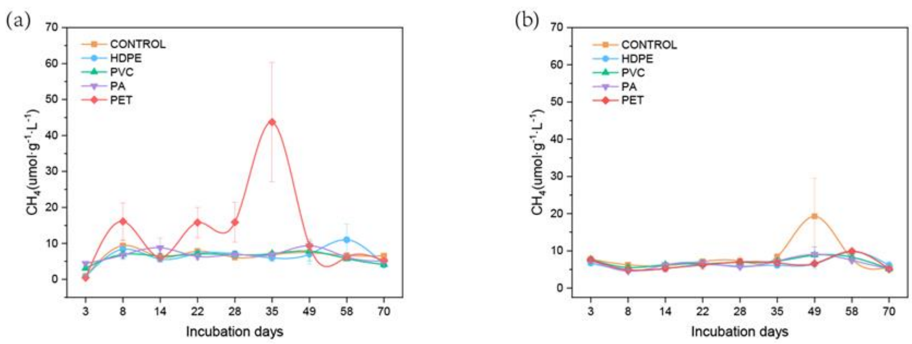

Figure 4. CH_4 concentrations in saltwater ecosystem after microplastics (HDPE, PVC, PA, PET) were added. (**a**) salt marsh (**b**) mangrove forest.

3.3. Global Warming Equivalent in Different Ecosystems

The increasingly serious problem of global warming, caused by greenhouse gas emissions, has been widely discussed [35]. CH_4 emissions during the growing season were converted into a CO_2 equivalent by multiplying CH_4 concentration by 34 (based on a 100-year time horizon).

This study investigated the relative trends of GWE in brackish and freshwater ecosystems affected by microplastics. Studies have shown that HDPE has a significant effect on GWE in freshwater ecosystems, with GWE values increasing significantly after 49 days of culture. CH_4 concentration in paddy soil increased significantly on 22 d and remained stable on 35 d when HDPE was added. The GWE of HDPE increased from 17.36 to 387.94 after 49 days of culture (Figure 5b). In Poyang Lake soil, there was a steady fluctuation trend from 35 days to 35 days. PVC fluctuated significantly after 35 days of culture. The GWE of HDPE increased from 19.11 to 460.98 after 49 days of culture (Figure 5a). Different from freshwater ecosystem, GWE fluctuated greatly in saltwater ecosystem from the initial stage of culture. In the salt marsh, the upward trend was most obvious on the 8th day of culture, and then tended to a steady fluctuation state. On the 35th day of culture, excluding HDPE, which increased significantly, the other three microplastics showed a downward trend. Among these, PVC decreased most obviously (Figure 6a). In mangrove forests, the overall fluctuation range is large. The minimum value appeared on the 35th day of PA cultivation, GWE was 12.25; the maximum value was the GWE of PET, 17.07 at day 70 of culture (Figure 6b). In addition, the GWE maximum value for freshwater ecosystems is 11.23 higher than the maximum value for saltwater ecosystems. Compared with saltwater GWE, it was found that the greenhouse effect of soil in freshwater system was more significant than that in saltwater under the condition of microplastics. In addition, HDPE has the highest methane concentration in soil compared with PVC, PA and PET, and so HDPE has the most significant impact on greenhouse gases.

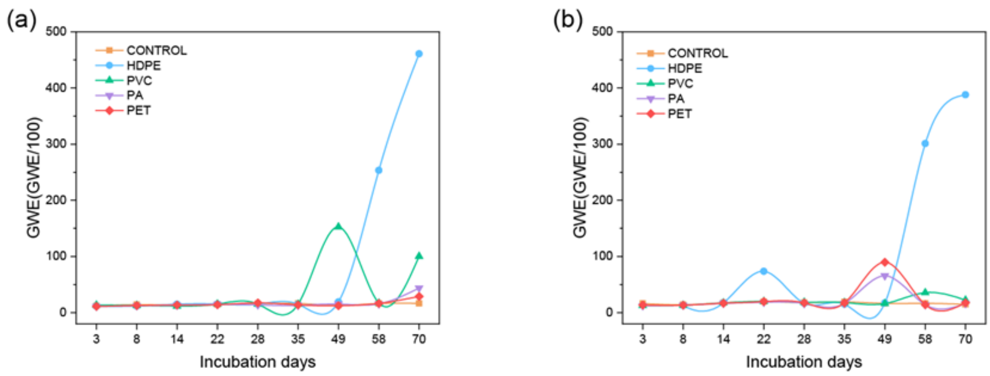

Figure 5. GWE in freshwater ecosystems (**a**) Poyang Lake (**b**) paddy soil.

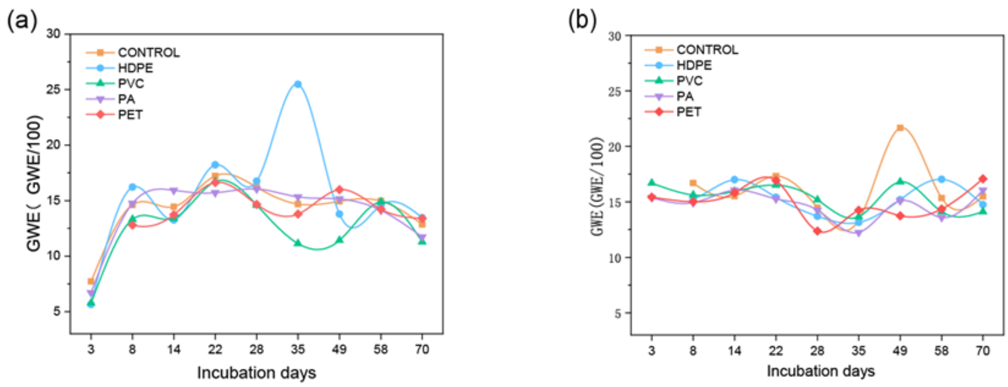

Figure 6. GWE in saltwater ecosystems (**a**) salt marsh (**b**) mangrove forest.

4. Conclusions

In this study, we demonstrated that CO_2 production was most significantly affected by soil characteristics (Table 1). However, we observed that CH_4 production changed from being most affected by soil to being most impacted by interactions between soil and microplastics with time elapsing, and finally was most significantly affected by microplastics (Table 2). In the soil of different ecosystems, the highest concentrations of CH_4 and CO_2 were found in Poyang Lake (PY), which were 1276 umol·g^{-1}·L^{-1} and 2713 umol·g^{-1}·L^{-1}, respectively (Figure 1a, Figure 2a). Furthermore, PET had the most significant effect on CO_2, and HDPE had the most significant effect on CH_4. However, mangrove soil was not sensitive to the addition of microplastics. The concentration of CO_2 emitted from mangroves ranged from 980 to 1534 umol·g^{-1}·L^{-1} (Figure 3b), and the concentration of CH_4 was even lower, ranging from 5 to 19 umol·g^{-1}·L^{-1} (Figure 4b). Compared with the saltwater ecosystem, CO_2 and CH_4 production in freshwater ecosystem increased sharply. The GWE maximum value for freshwater ecosystems is 11 times higher than the maximum value for saltwater ecosystems (Figure 5). Therefore, the GWE of a freshwater ecosystem was higher than that of a saltwater ecosystem, which may produce more greenhouse gases and contributed more to global climate change. Finally, it is hoped that the data analysis of CH_4 and CO_2 in different ecosystems of the four microplastics, HDEP, PVC, PA and PET, will provide value for future GHG-related research.

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