

# Effects of Carbon Dioxide Enrichment and Nitrogen Addition on Inorganic Carbon Leaching in Subtropical Model Forest Ecosystems

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## ABSTRACT

Soil mineral weathering may serve as a sink for atmospheric carbon dioxide (CO<sub>2</sub>). Increased weathering of soil minerals induced by elevated CO<sub>2</sub> concentration has been reported previously in temperate areas. However, this has not been well documented for the tropics and subtropics. We used model forest ecosystems in open-top chambers to study the effects of CO<sub>2</sub> enrichment alone and together with nitrogen (N) addition on inorganic carbon (C) losses in the leachates. Three years of exposure to an atmospheric CO<sub>2</sub> concentration of 700 ppm resulted in increased annual inorganic C export through leaching below the 70 cm soil profile. Compared to the control without any CO<sub>2</sub> and N treatments, net biocarbonate C (HCO<sub>3</sub><sup>-</sup>-C) loss increased by 42%, 74%, and 81% in the high

CO<sub>2</sub> concentration treatment in 2006, 2007, and 2008, respectively. Increased inorganic C export following the exposure to the elevated CO<sub>2</sub> was related to both increased inorganic C concentrations in the leaching water and the greater amount of leaching water. Net annual inorganic C (HCO<sub>3</sub><sup>-</sup>-C and carbonate C: CO<sub>3</sub><sup>2-</sup>-C) loss via the leaching water in the high CO<sub>2</sub> concentration chambers reached 48.0, 49.5, and 114.0 kg ha<sup>-1</sup> y<sup>-1</sup> in 2006, 2007, and 2008, respectively, compared with 33.8, 28.4, and 62.8 kg ha<sup>-1</sup> y<sup>-1</sup> in the control chambers in the corresponding years. The N addition showed a negative effect on the mineral weathering. The decreased inorganic C concentration in the leaching water and the decreased leaching water amount induced by the high N treatment were the results of the adverse effect. Our results suggest that tropical forest soil systems may be able to compensate for a small part of the atmospheric CO<sub>2</sub> increase through the accelerated processing of CO<sub>2</sub> into HCO<sub>3</sub><sup>-</sup>-C during soil mineral weathering, which might be transported in part into ground water or oceans on geological timescales.

**Key words:** carbon dioxide; inorganic carbon loss; mineral weathering; N addition; soil CO<sub>2</sub> flux; subtropical area.

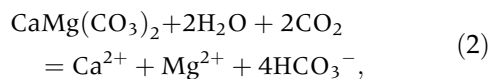
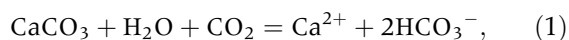
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## INTRODUCTION

Increasing atmospheric CO<sub>2</sub> concentration has been one of the most pronounced global changes for the past 100 years. To prevent or at least limit the potential adverse impacts of rising CO<sub>2</sub> concentrations, it is important to stabilize the atmospheric CO<sub>2</sub> concentration. Berner and Kothavala (2001) pointed out that the weathering of soil carbonate minerals may serve as a sink for increasing atmospheric CO<sub>2</sub>. Through soil CO<sub>2</sub> flux (including autotrophic and heterotrophic respiration), CO<sub>2</sub> is released and transitorily kept in soils, and in turn, partly dissolves in the soil solution to form carbonic acid that is available for mineral weathering. As the result of carbonate weathering by carbonic acid (equations 1, 2), bicarbonate and base cations are released. This dissolved C can then be sequestered in shallow groundwater reservoirs or returned to the ocean reservoir via rivers at the geological time scale (Williams and others 2003).



Under elevated atmospheric CO<sub>2</sub>, increased soil CO<sub>2</sub> flux has been demonstrated to affect mineral weathering (Andrews and Schlesinger 2001; Williams and others 2003). Andrews and Schlesinger (2001) conducted an experiment in the Duke forest, and reported that an increase of 55% in atmospheric CO<sub>2</sub> over 2 years resulted in a 33% increase in the flux of dissolved inorganic C to groundwater. Using open-top chambers, Williams and others (2003) determined the effect of elevated atmospheric CO<sub>2</sub> on mineral weathering reactions in mid-latitude forest soils, and found that the CO<sub>2</sub> treatment had a significant effect on the concentration of dissolved inorganic C, which was 12% higher in elevated than ambient CO<sub>2</sub> chambers. However, all these experiments were conducted in temperate forests. There has been little work done in tropical forest ecosystems. The effect of elevated CO<sub>2</sub> on inorganic C leaching in old and highly weathered soils is not well understood, especially in southern China.

It is widely assumed that elevated atmospheric CO<sub>2</sub> leads to reduced stomatal conductance (Saxe and others 1998) and diffusive conductance (Pearson and others 1995; Niklaus and others 1998). Reductions of diffusive and stomatal conductance translate usually into decreased rates of canopy transpiration and increased soil moisture in

CO<sub>2</sub>-enriched plots (Bunce 2004). Higher soil moisture usually causes a higher fraction of precipitation to flow away through streamflow and deep seepage. If CO<sub>2</sub> enrichment can also increase the weathering of carbonate minerals in forest soils of tropical and subtropical regions, together with the high precipitation and streamflow in these regions, inorganic C leaching could be an important C sink under the global CO<sub>2</sub> enrichment background.

Atmospheric N deposition is a serious problem in southern China. Currently, this leads to high N deposition in precipitation in some forests (30–73 kg N ha<sup>-1</sup> y<sup>-1</sup>) (Ren and others 2000; Mo and others 2006). The N deposition in Guangzhou City of southern China increased from 46 kg N ha<sup>-1</sup> y<sup>-1</sup> in 1988 to 73 kg N ha<sup>-1</sup> y<sup>-1</sup> in 1990 (Ren and others 2000). It was reported that increased plant growth under a CO<sub>2</sub> enrichment environment depends on N availability (Luo and others 2006). What would be then the effects of elevated atmospheric CO<sub>2</sub> with the increased N deposition on the soil mineral weathering in tropical and subtropical forests?

In this study, we used open-top chambers to study the effect of elevated atmospheric CO<sub>2</sub> alone and together with N addition in southern China on the dynamics of inorganic C in the soil leaching water over 3 years after the treatments started. We hypothesized: (1) elevated atmospheric CO<sub>2</sub> would increase soil moisture, and then increased soil water leaching in the tropical area; (2) elevated atmospheric CO<sub>2</sub> would increase soil mineral weathering, which would lead to increased inorganic C leaching; and (3) inorganic C leaching under elevated CO<sub>2</sub> would be altered by the N supply.

## METHODS

### Open-Top Chamber Design

Model forest ecosystems were established in 10 open-top chambers (Figure 1). These 3-m diameter chambers consisted of two parts, a 3-m high aboveground part and a 0.7-m deep belowground part. The aboveground part was delimited by impermeable and transparent plastic sheets that were wrapped around the chambers. The tops of these chambers were completely open with no cover. The light intensity in the chambers was 97% of the full light and there was no spectral change. Moreover, the measured rainfall intensity was identical inside and outside of the chambers and the temperature was not significantly different

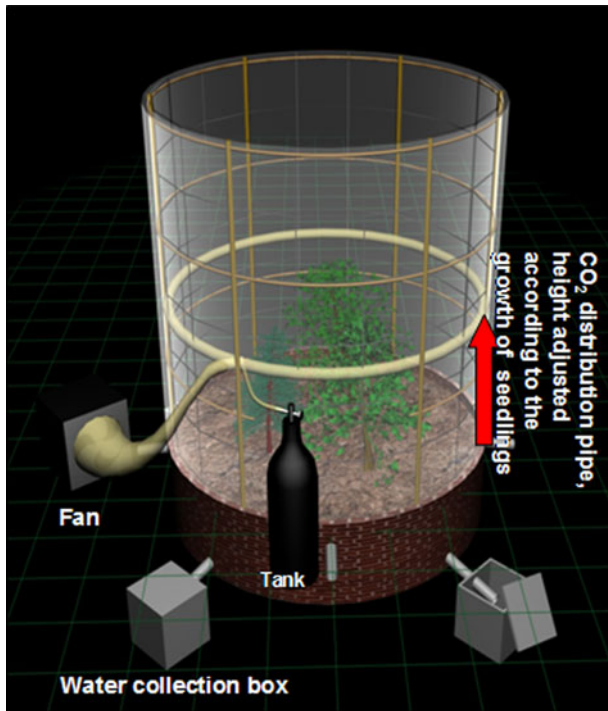


Figure 1. Open-top chamber in our experiment.

either. The belowground part was delimited by brick walls that prevented any lateral or vertical water and/or element fluxes with the outside surrounding soil. Three holes at the bottom of the walls were connected to stainless steel water collection boxes. Holes were capped by a 2-mm plastic net to prevent losses other than those of leachates. The additional CO<sub>2</sub> coming from the tank was distributed in each chamber by a transparent pipe with pinholes. The pipe entwines the inner wall of each chamber in a snake shape and from 2.5 to 0.5 m in height. The distance between two pinholes is 1 cm. To ensure that CO<sub>2</sub> was equally distributed in the entire chamber, the pipe was connected to a fan. The CO<sub>2</sub> flux from the tank was controlled by a flowmeter and CO<sub>2</sub> concentrations on the five planes (2.5, 2, 1.5, 1.0, and 0.5 m in height) in the chambers were periodically monitored using a Licor-6400 (LI-COR Inc., Lincoln, Nebraska, USA). There were three monitoring points on each plane. Soil moisture was recorded every week on several random points per chamber using time-domain reflectometry (TDR).

## Experiment Design

The study was carried out in Guangzhou City, Guangdong Province, China (23°20' N and 113°30' E). The area has a monsoon climate, characterized by a mean annual total solar radiation of

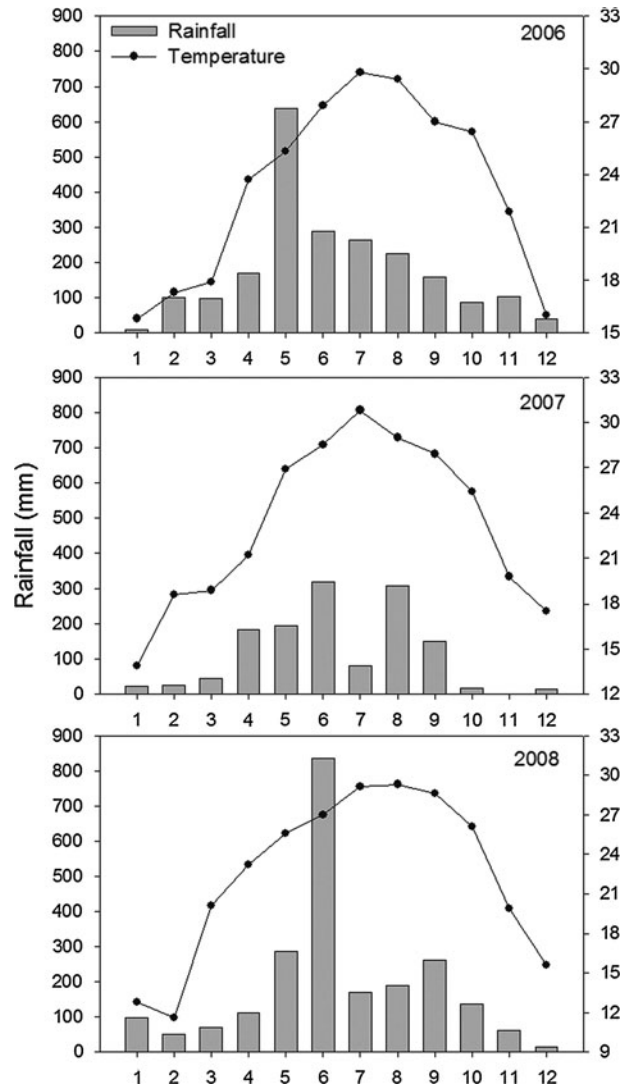


Figure 2. Monthly rainfall and mean temperature in Guangzhou City, China, during the experimental period from 2006 to 2008.

4367.2–4597.3 MJ m<sup>-2</sup> in the visible waveband and a mean annual temperature of 21.5°C. The annual precipitation ranges from 1600 to 1900 mm, and the mean relative humidity is 77%. There are two main seasons, a wet/rainy season from April to September and a dry season from October to March. Rainfall and air temperature during the experimental period are shown in Figure 2.

In March 2005, we collected the soil from a nearby ever-green broad-leaved forest after harvesting. The soil type was ultisol overlying sandstone and shale bedrocks, with a pH value below 4.5 in all the soil layers. Soil organic C is low in this sandy soil. The primary chemical properties were shown in Liu and others (2008). The soil was collected as three different layers (0–20-, 20–40-, and 40–70-cm depth)

that were homogenized separately and used to fill the belowground part of the chambers correspondingly. One- to two-year-old seedlings grown in a nursery were transplanted in the chambers without damaging the roots. All the chambers were planted with 48 randomly located seedlings with 8 seedlings for each of the following six species: *Castanopsis hystrix* Hook.f. & Thomson ex A.DC, *Syzygium hancei* Merr. et Perry, *Pinus massoniana* Lambert, *Schima superba* Gardn. and Champ., *Acmena acuminatissima* (Blume) Merr. et Perry, and *Ormosia pinnata* (Lour.) Merr. These species were selected because they are native and the most widely distributed tree species in southern China. As trees were growing fast, one tree per species was harvested at the end of each year to avoid excessively crowded conditions in each chamber.

The open-top chambers used in the experiment were located in an open area where they were exposed to full light and rain. From April 2005, the chambers were exposed to the different treatments. Three chambers received a high CO<sub>2</sub> and high N treatment (CN), three chambers received a high CO<sub>2</sub> treatment (CC), two chambers a high N treatment (NN), and finally two chambers were used as a control (CK) and did not receive high CO<sub>2</sub> or high N treatment. The high CO<sub>2</sub> treatments were achieved by supplying additional CO<sub>2</sub> from a tank until a CO<sub>2</sub> concentration of 700 ppm was reached in the chambers. The high N addition treatments were achieved by spraying seedlings one time a week for a total amount of NH<sub>4</sub>NO<sub>3</sub> at 100 kg N ha<sup>-1</sup> y<sup>-1</sup>. No other fertilizer was used. Because the walls of the chambers in the belowground parts blocked lateral and vertical water fluxes, the seedlings were watered with tap water. All other chambers received the same amount of water as the CK chambers.

## Sample Collection and Measurement

To determine soil C content, soil samples were collected for each chamber in July 2005 and November 2008 (the end of soil solution collection). Soil samples were randomly collected from 0–20, 20–40, and 40–60 cm soil layers in each chamber. Each sample consisted of three cores (2.5-cm diameter) by the same depth in each chamber. The composite samples were air dried, gently mixed, and sieved (<2 mm). Dead roots and plant residues were picked out. Soil organic C was determined following the Walkley Black's wet digestion method (Nelson and Sommers 1982) and inorganic C was measured using colorimetric titration, in which samples were acidified by HCl to

dissolve carbonate minerals and release CO<sub>2</sub>, and then using NaOH to titrate the excess HCl. Weighted percent inorganic C was then obtained by calculation.

Leaching water sample collection started in 2006 in the second year of the treatment applications and continued for three years. Soil leachates were collected at the bottom of the chamber belowground walls in stainless steel boxes. During the dry season, they were collected after each rainfall. During the wet season, they were collected one time a week. Each time, the exact volume of total leachates was measured and 100 ml per box was collected for chemical analysis. The Ca<sup>2+</sup> and Mg<sup>2+</sup> concentrations in the leachates were measured using inductively coupled plasma atomic emission spectroscopy. Both CO<sub>3</sub><sup>2-</sup>-C and HCO<sub>3</sub><sup>-</sup>-C were measured by titration with HCl after the addition of BCP indicator (Vuai and Tokuyama 2007). To determine C input from rainfall, rain water was collected in an open area near the chambers and sent for the same analyses as leachates.

To measure soil CO<sub>2</sub> flux (including autotrophic root respiration and heterotrophic microbial respiration), in April 2006, four PVC circular collars (10-cm diameter) were permanently installed in each chamber; each was inserted about 5 cm into the mineral soil between the growing plants. From 26 May 2006, soil CO<sub>2</sub> flux measurements were made once a week using an infrared gas analysis system (IRGA, LICOR 6400, LiCor Inc, Lincoln, Nebraska, USA). To avoid extremely high temperatures at noon, soil CO<sub>2</sub> fluxes were determined in the morning (09:00–12:00). The LICOR 6400 chamber (with a foam gasket) was placed on the PVC collars making an air-tight seal. Soil CO<sub>2</sub> flux of a collar was determined three times repeatedly by measuring the rate of CO<sub>2</sub> increase in the LICOR 6400 respiration chamber. The soil CO<sub>2</sub> flux was based on the average of the data from the four locations in each chamber (they differed by less than 5% at any measurement period). Soil moisture was recorded at the same time when measuring the soil CO<sub>2</sub> flux. It was determined on several random points around each collar using a PMKit (Tang and others 2006).

The growth of five species (*Castanopsis hystrix* Hook.f. & Thomson ex A.DC, *Syzygium hancei* Merr. et Perry, *Schima superba* Gardn. and Champ., *Acmena acuminatissima* (Blume) Merr. et Perry, and *Ormosia pinnata* (Lour.) Merr.) was measured as the increment in plant dimensions over time to determine how the species responded to the various CO<sub>2</sub> and N treatments. Plant height and basal diameter were measured at the time of planting in early



March 2005. They were assessed seven times later in August 2005, November 2005, May 2006, September 2007, January 2008, August 2008 and January 2009, respectively. Plant height was measured from the soil-stem surface to the tip of the apical bud and the diameter was assessed at the soil surface.

## Data Analysis

Data analyses were carried out using the SAS software (SAS Institute Inc., Cary, North Carolina, USA). For each variable, the normality of the distribution was tested by a Shapiro–Wilk test. We chose  $\alpha$  equal to 0.05. Analyzed data consisted of monthly concentrations and amounts of inorganic C in the leachates, monthly amount of leaching water, soil CO<sub>2</sub> flux and soil moisture. Monthly leaching water volumes were calculated by adding each individual amount of leaching water during the whole month. Monthly inorganic C leached was calculated by adding each individual ion concentration measurement factored by their respective amount of leaching water. Standardized average monthly ion concentrations in the leachates were calculated by dividing monthly ion amounts by monthly volumes of the leachates. Annual net loss amounts were calculated by adding monthly amounts but minus the ion inputs from the rainfall. Data were analyzed using the following mixed linear model:

$$\begin{aligned} \text{Dependent variables} &= C + N + C * N \\ &+ \text{season} + \text{month (season)} + C * \text{season} \\ &+ N * \text{season} + C * \text{month (season)} \\ &+ N * \text{month (season)} + \epsilon \end{aligned}$$

where C is the effect of the CO<sub>2</sub> treatments (ambient or 700 ppm), N is the effect of the N treatment (ambient or high deposition), season is the effect of seasons (wet season from March to September, and dry season during the other months), month (season) is the effect of months nested in their respective season, \* is the interaction between the factors, and  $\epsilon$  is the experimental error. The effect of the chambers nested inside the C and N treatments was added as a random variable. The chamber and the interaction of C \* N \* season were not significantly affected for the measurement data and therefore were excluded from the Results section. When the effects were significant, they were further analyzed using a Tukey multiple comparison test performed on least square means. Additionally, simple correlations were performed on the whole set of data using the Pearson correlation coefficients.

To evaluate the effect of treatments on plant growth at different measurement occasions, repeated measures of ANOVA were used. When the effects were significant, they were further analyzed using a Tukey multiple comparison test.

## RESULTS

### Soil C Content

Soil organic C and inorganic C were determined at the beginning (July 2005) and at the end of the experiment (November 2008). The higher soil organic C was observed at the end of the experiment when compared to the initial data in all the treatments. The treatments had a significant effect on soil organic C content, with the greater contents in the CN and NN treatment chambers compared to the other chambers ( $P < 0.05$ , Table 1). As for inorganic C, neither treatments nor sampling times affected inorganic C contents.

### Soil Moisture and Soil CO<sub>2</sub> Flux

Soil CO<sub>2</sub> flux and soil moisture were recorded once a week. Soil moisture was significantly affected by the CO<sub>2</sub> treatment, N treatment, sampling season ( $P < 0.0001$  for all) and their interactions. Soil moisture showed a strong seasonal pattern with a higher value observed in the wet season (April–September), compared with that in the dry season (October–March) for all treatments. The elevated CO<sub>2</sub> treatment increased soil moisture remarkably, with greater soil moisture in the CC and CN chambers when compared to the control chambers. However, the N treatment decreased significantly the soil moisture, with the lowest soil moisture in the NN treatment chambers (Table 2). With time, the negative effect of the N treatment became more significant. Soil CO<sub>2</sub> fluxes varied with the different measuring times, with higher values in the wet season as soil CO<sub>2</sub> fluxes were positively related to soil moisture ( $P < 0.0001$ ). The CN treatment showed the highest soil CO<sub>2</sub> flux rates, followed by the CC treatment, and then the NN and CK treatments (Table 2). Both CO<sub>2</sub> and N treatments positively affected soil CO<sub>2</sub> flux rates ( $P < 0.0001$  for both). Among the years, the highest soil CO<sub>2</sub> flux rates were in 2008, and the lowest were in 2007.

### Leaching Water Volume and pH

The volumes of leaching water varied throughout the year with the greater amounts during the wet season (from April to September). The monthly volumes of leaching water were consistent with the

**Table 1.** Comparison of Inorganic and Organic Carbon (C) Contents (Means  $\pm$  Standard Deviations) of Open-Top Chamber Soils at the Beginning (Initial, July 2005) and at the End of the Experiment (Final, November 2008)

Treatments	Soil layers	Soil organic C (% means $\pm$ SD)		Soil inorganic C (% means $\pm$ SD)	
		Initial	Final	Initial	Final
CN	0–20	0.450 $\pm$ 0.035	1.555 $\pm$ 0.070	0.019 $\pm$ 0.003	0.014 $\pm$ 0.005
	20–40	0.360 $\pm$ 0.006	0.992 $\pm$ 0.036	0.016 $\pm$ 0.003	0.014 $\pm$ 0.007
	40–60	0.220 $\pm$ 0.025	0.680 $\pm$ 0.041	0.019 $\pm$ 0.006	0.018 $\pm$ 0.006
CC	0–20	0.427 $\pm$ 0.033	1.259 $\pm$ 0.153	0.019 $\pm$ 0.003	0.013 $\pm$ 0.002
	20–40	0.297 $\pm$ 0.032	0.812 $\pm$ 0.067	0.016 $\pm$ 0.003	0.010 $\pm$ 0.003
	40–60	0.140 $\pm$ 0.005	0.451 $\pm$ 0.030	0.019 $\pm$ 0.006	0.011 $\pm$ 0.004
NN	0–20	0.500 $\pm$ 0.090	1.498 $\pm$ 0.181	0.019 $\pm$ 0.003	0.018 $\pm$ 0.002
	20–40	0.225 $\pm$ 0.015	0.851 $\pm$ 0.011	0.016 $\pm$ 0.003	0.017 $\pm$ 0.003
	40–60	0.180 $\pm$ 0.030	0.595 $\pm$ 0.051	0.019 $\pm$ 0.006	0.018 $\pm$ 0.004
CK	0–20	0.425 $\pm$ 0.003	1.283 $\pm$ 0.097	0.019 $\pm$ 0.003	0.017 $\pm$ 0.004
	20–40	0.240 $\pm$ 0.020	0.853 $\pm$ 0.126	0.016 $\pm$ 0.003	0.020 $\pm$ 0.005
	40–60	0.180 $\pm$ 0.010	0.462 $\pm$ 0.106	0.019 $\pm$ 0.006	0.026 $\pm$ 0.008

Treatments are: CK = control; NN = high N treatment; CC = elevated CO<sub>2</sub> concentration treatment; CN = elevated CO<sub>2</sub> concentration treatment + high N treatment.

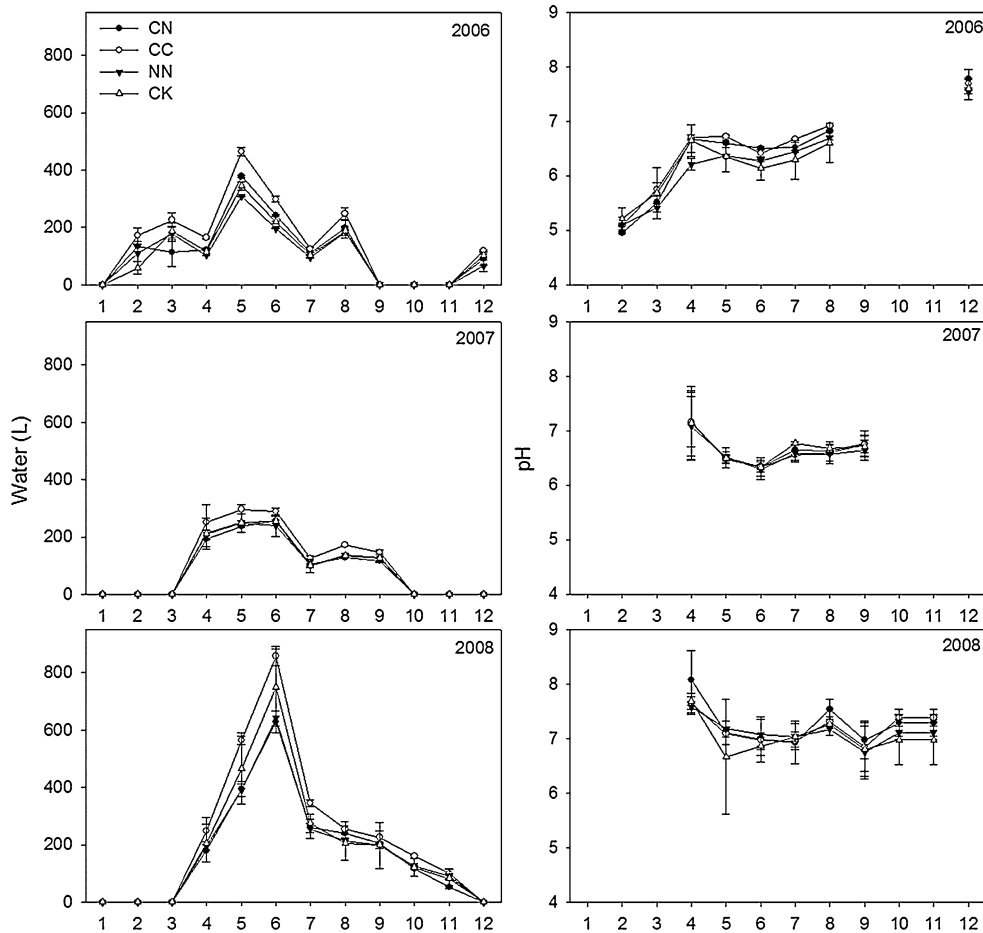
**Table 2.** Effects of Treatments on Soil Moisture and Soil CO<sub>2</sub> Flux (Means  $\pm$  Standard Deviations)

	2006				2007				2008			
	CN	CC	NN	CK	CN	CC	NN	CK	CN	CC	NN	CK
Soil moisture (%)												
Year	22.50a	21.97a	19.67b	20.62b	21.47a	22.00a	18.31c	20.17b	21.55ab	22.45a	18.49c	20.58b
	$\pm$ 4.99	$\pm$ 4.77	$\pm$ 5.01	$\pm$ 4.50	$\pm$ 5.82	$\pm$ 5.59	$\pm$ 6.57	$\pm$ 6.60	$\pm$ 6.78	$\pm$ 6.93	$\pm$ 7.07	$\pm$ 6.88
Wet	25.48a	24.14a	22.48b	22.45b	23.92a	24.42a	21.94b	23.64a	26.48b	27.51a	23.84c	25.61b
	$\pm$ 4.23	$\pm$ 4.87	$\pm$ 4.70	$\pm$ 4.57	$\pm$ 5.52	$\pm$ 5.41	$\pm$ 6.31	$\pm$ 6.25	$\pm$ 3.75	$\pm$ 3.89	$\pm$ 3.68	$\pm$ 3.54
Dry	19.28a	19.69a	16.57b	18.66a	18.98a	19.57a	14.70c	16.70b	16.85ab	17.65a	13.53c	15.95b
	$\pm$ 3.56	$\pm$ 3.43	$\pm$ 3.19	$\pm$ 3.51	$\pm$ 5.02	$\pm$ 4.64	$\pm$ 4.52	$\pm$ 4.89	$\pm$ 5.58	$\pm$ 5.66	$\pm$ 5.72	$\pm$ 5.89
Soil CO <sub>2</sub> flux ( $\mu$ mol CO <sub>2</sub> m <sup>-2</sup> s <sup>-1</sup> )												
Year	3.72a	3.33b	2.80c	2.53d	3.51a	3.18b	2.51c	2.45c	4.11a	3.43b	2.67c	2.61c
	$\pm$ 1.41	$\pm$ 1.19	$\pm$ 0.97	$\pm$ 0.76	$\pm$ 1.48	$\pm$ 1.54	$\pm$ 1.61	$\pm$ 0.92	$\pm$ 1.88	$\pm$ 1.65	$\pm$ 1.51	$\pm$ 1.16
Wet	4.60a	4.09b	3.02c	2.89c	4.25a	4.12a	2.95b	2.80b	5.21a	4.53b	3.75c	3.27d
	$\pm$ 1.31	$\pm$ 1.05	$\pm$ 0.97	$\pm$ 0.74	$\pm$ 1.65	$\pm$ 1.61	$\pm$ 1.09	$\pm$ 1.07	$\pm$ 1.73	$\pm$ 1.56	$\pm$ 1.46	$\pm$ 1.25
Dry	2.85a	2.51b	2.57b	2.16c	2.76a	2.29b	2.11c	2.14c	3.09a	2.43b	1.72c	2.02d
	$\pm$ 0.85	$\pm$ 0.70	$\pm$ 0.92	$\pm$ 0.57	$\pm$ 0.74	$\pm$ 0.72	$\pm$ 1.88	$\pm$ 0.62	$\pm$ 1.37	$\pm$ 0.96	$\pm$ 0.70	$\pm$ 0.65

Treatments with the same superscript letter are not significantly different from each other ( $P > 0.05$ ). Treatments are: CK = control; NN = high N treatment; CC = elevated CO<sub>2</sub> concentration treatment; CN = elevated CO<sub>2</sub> concentration treatment + high N treatment; year = the means of the whole year; wet = the means in the wet season; dry = the means in the dry season.

rainfall amounts during the whole experiment period (Figures 2, 3). This means that higher monthly rainfall led to greater monthly leaching water. There was a higher annual water leaching amount in 2008, followed by 2006 and then 2007. The high CO<sub>2</sub> treatment led to increased leaching water volumes (Table 4), whereas the high N treatment reduced the volumes (Tables 3, 4). Consequently, the greatest water volumes were found in the chambers exposed to the high CO<sub>2</sub>

treatment (CC) over the 3 years. There was a strong interactive effect of the high CO<sub>2</sub> and N treatment ( $P < 0.01$  in 2006,  $P < 0.001$  in 2007 and 2008). In 2006, the water volumes in the leachates were in the order: CC > CN > NN > CK. However, in both 2007 and 2008, the greatest water volumes were found in the CC chambers, followed by CK and finally the NN and CN treatments, indicating that with time the N treatments had significant effects on the amount of soil water



**Figure 3.** Monthly volumes (left column) and pH values (right column) of leachates under different CO<sub>2</sub> and N treatments. Error bars are standard deviations ( $n = 9$  for CC and CN,  $n = 6$  for NN and CK). Treatments are: CK = control; NN = high N treatment; CC = elevated CO<sub>2</sub> concentration treatment; CN = elevated CO<sub>2</sub> concentration treatment + high N treatment.

**Table 3.** Effects of Treatments on Annual Volumes of Leaching Water and Annual Amounts of DIC (Dissolved Inorganic C) in Leachates

	2006				2007				2008			
	CN	CC	NN	CK	CN	CC	NN	CK	CN	CC	NN	CK
Annual amounts (L ha <sup>-1</sup> y <sup>-1</sup> )												
Water × 10 <sup>-4</sup>	215b	281a	170c	166d	147c	181a	150c	153b	294c	390a	299c	326b
Annual amounts(kg ha <sup>-1</sup> y <sup>-1</sup> )												
CO <sub>3</sub> <sup>2-</sup> -C	0.43a	0.34b	0.15c	0.2c	Not detected				0.55b	0.79a	0.25c	0.17c
HCO <sub>3</sub> <sup>-</sup> -C	38.7b	47.7a	29.4c	33.6c	40.9b	49.5a	29.4c	28.4c	68.5b	113.2a	54.0b	62.6b
% Increase over the control												
Water	30	69	2	0	-4	18	-2	0	-10	20	-8	0
CO <sub>3</sub> <sup>2-</sup> -C	116	72	-24	0	Not detected				217	367	50	0
HCO <sub>3</sub> <sup>-</sup> -C	15	42	-12	0	44	74	4	0	9	81	-14	0

Treatments with the same superscript letter are not significantly different from each other ( $P > 0.05$ ). Treatments are: CK = control; NN = high N treatment; CC = elevated CO<sub>2</sub> concentration treatment; CN = elevated CO<sub>2</sub> concentration treatment + high N treatment.

leached. The volumes of leachates were 69%, 18%, and 20% greater in the CC treatment in 2006, 2007, and 2008, respectively, compared to the control treatment (Table 3).

In 2006, pH values of leaching water increased significantly, from 4.48 in February to 7.79 in

December (Figure 3). In both 2007 and 2008, the pH values in the leachates were relatively stable. However, there were no significant differences between the treatments in both 2006 and 2007. The chambers exposed to the elevated CO<sub>2</sub> had higher pH values in the leachates ( $P = 0.008$ ) than

**Table 4.** Effects of CO<sub>2</sub> Treatment (C), N Treatment (N), Season and Month (season) and their Interactions on Concentrations and Monthly Amounts of Water and DIC (Dissolved Inorganic C, CO<sub>3</sub><sup>2-</sup>-C and HCO<sub>3</sub><sup>-</sup>-C) in Leachates

Variables	Year	C	N	C * N	Season	C * season	N * season	Month (season)	C * month (season)	N * month (season)
Concentrations										
CO <sub>3</sub> <sup>2-</sup> -C	2006	3.4	1.41	1.4	15.5***	2.0	0.81	10.7***	2.4*	1.2
	2008	15.5***	0.42	0.3	24.5***	7.7**	1.02	8.0***	3.1**	0.2
HCO <sub>3</sub> <sup>-</sup> -C	2006	7.8**	0.4	0	19.4***	1.5	0.1	7.3***	0.4	0.4
	2007	55.5***	2.2	2.9				26.0***	2.4*	0.9
	2008	8.4**	8.2**	0.2	18.1***	1.1	0.1	2.8*	0.6	0.3
Monthly amounts										
Water	2006	86.0***	14.8***	19.1**	61.2***	0.9	0.2	79.4***	1.6	1.0
	2007	22.6***	69.0***	37.4***				353.5****	1.0	0.1
	2008	8.9**	62.9***	19.3***	1338.3****	3.7	11.2**	319.4***	0.7	9.0***
CO <sub>3</sub> <sup>2-</sup> -C	2006	4.4*	0.3	0.6	19.8***	2.5	0.2	14.2***	3.3**	1.0
	2008	17.6***	1.4	1.7	0.7	0.4	0.5	18.0***	3.3**	0.4
HCO <sub>3</sub> <sup>-</sup> -C	2006	9.3**	4.0*	0.55	31.0***	2.6	0.1	23.3***	0.9	0.4
	2007	68.4***	16.0***	12.4***				63.2***	6.9***	1.9
	2008	17.1***	17.0***	5.6*	137.4***	6.5*	3.8	28.7***	2.4*	1.5

Numbers are F values. Asterisks indicate the level of significance (no asterisk = not significant, \*  $P < 0.05$ , \*\*  $P < 0.01$ , \*\*\*  $P < 0.001$ ).

the chambers under the ambient CO<sub>2</sub> in 2008. The pH values in the leachates were positively related to both leaching water volume and HCO<sub>3</sub><sup>-</sup>-C concentrations.

### Inorganic C in the Leachates

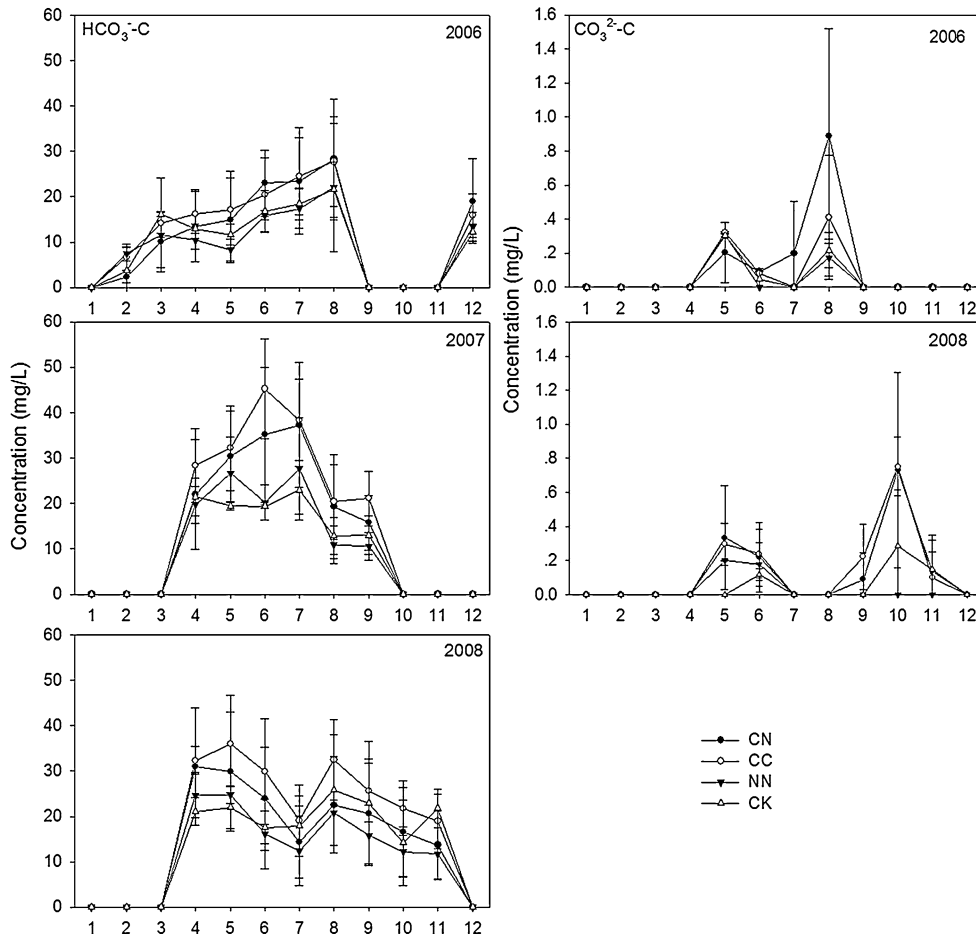
The HCO<sub>3</sub><sup>-</sup>-C concentrations in the leachates varied throughout the whole experimental period (Figure 4). Sampling time affected the HCO<sub>3</sub><sup>-</sup>-C concentrations, with reduced HCO<sub>3</sub><sup>-</sup>-C concentrations during the wet season (Figure 4). Among the 3 years, concentrations of HCO<sub>3</sub><sup>-</sup>-C in 2007 showed the highest values, followed by 2008 and then 2006 for the CC, CN, and CK treatments, whereas there was no significant difference between the years for the NN treatment. The HCO<sub>3</sub><sup>-</sup>-C concentrations in the leachates were positively affected by the high CO<sub>2</sub> treatment and negatively affected by the high N treatment (Table 4). There were no statistically significant differences among the treatments in 2006. However, the greater HCO<sub>3</sub><sup>-</sup>-C concentrations were shown in the CC and CN treatments than the NN and CK treatments ( $P < 0.0001$ ) in 2007. In 2008, the HCO<sub>3</sub><sup>-</sup>-C concentrations in the treatments followed the order: CC > CN = CK > NN ( $P = 0.004$ ). The HCO<sub>3</sub><sup>-</sup>-C concentrations and Ca and Mg concentrations were positively correlated to each other.

The monthly amounts of HCO<sub>3</sub><sup>-</sup>-C in the leachates also varied throughout the whole

experimental period. There were more leaching amounts of HCO<sub>3</sub><sup>-</sup>-C in 2008 than the other sampling years. The CO<sub>2</sub> and N treatments affected significantly the monthly amounts of HCO<sub>3</sub><sup>-</sup>-C in the leachates ( $P < 0.001$  for both in 2007 and 2008, Table 4). The combined effect of the high CO<sub>2</sub> and N treatments on the HCO<sub>3</sub><sup>-</sup>-C concentrations was the apparent interaction ( $P < 0.001$  in 2007 and  $P < 0.05$  in 2008). Compared to the other treatments, the CC treatment showed the greatest monthly amounts of HCO<sub>3</sub><sup>-</sup>-C in the leachates, and the NN treatment had the lowest monthly amounts. The annual net HCO<sub>3</sub><sup>-</sup>-C leaching losses were 47.7, 49.5, and 113.2 kg ha<sup>-1</sup> y<sup>-1</sup> in the CC treatment in 2006, 2007, and 2008, respectively, which were 42%, 74%, and 81% higher than those of the control (Table 3). The annual net HCO<sub>3</sub><sup>-</sup>-C leaching losses were 38.7, 40.9, and 68.5 kg ha<sup>-1</sup> y<sup>-1</sup> in the CN treatment in 2006, 2007, and 2008, respectively (Table 3). The N treatment reduced the positive effect of high CO<sub>2</sub> treatment on the HCO<sub>3</sub><sup>-</sup>-C loss in the leachates.

Compared to the HCO<sub>3</sub><sup>-</sup>-C, CO<sub>3</sub><sup>2-</sup>-C concentrations and amounts in the leachates were low. They could not be detected in 2007. Both concentrations and monthly amounts of CO<sub>3</sub><sup>2-</sup>-C in the leachates were affected by the CO<sub>2</sub> treatment and the sampling time. The N treatment had no effects on CO<sub>3</sub><sup>2-</sup>-C (Table 4). The CC and CN treatments exhibited higher CO<sub>3</sub><sup>2-</sup>-C concentrations and monthly leaching amounts than CK and NN





**Figure 4.** Concentrations of  $\text{HCO}_3^-$ -C (left column) and  $\text{CO}_3^{2-}$ -C (right column) in leachates under various  $\text{CO}_2$  and N treatments. Error bars are standard deviations ( $n = 9$  for CC and CN,  $n = 6$  for NN and CK). Treatments are: CK = control; NN = high N treatment; CC = elevated  $\text{CO}_2$  concentration treatment; CN = elevated  $\text{CO}_2$  concentration treatment + high N treatment. We could not detect the  $\text{CO}_3^{2-}$ -C concentration in 2007.

treatments. The annual net  $\text{CO}_3^{2-}$ -C leaching losses were 0.34 and 0.79  $\text{kg ha}^{-1} \text{y}^{-1}$  in the CC treatment in 2006 and 2008, respectively, which were 72% and 367% higher than those of the control (Table 3). The annual net  $\text{CO}_3^{2-}$ -C leaching losses were 0.43 and 0.55  $\text{kg ha}^{-1} \text{y}^{-1}$  in the CN treatment in 2006 and 2008, respectively (Table 3). The  $\text{CO}_3^{2-}$ -C concentrations were positively correlated to the Ca and Mg concentrations ( $R^2 = 0.31$  and  $R^2 = 0.37$ ,  $P < 0.05$ , respectively).

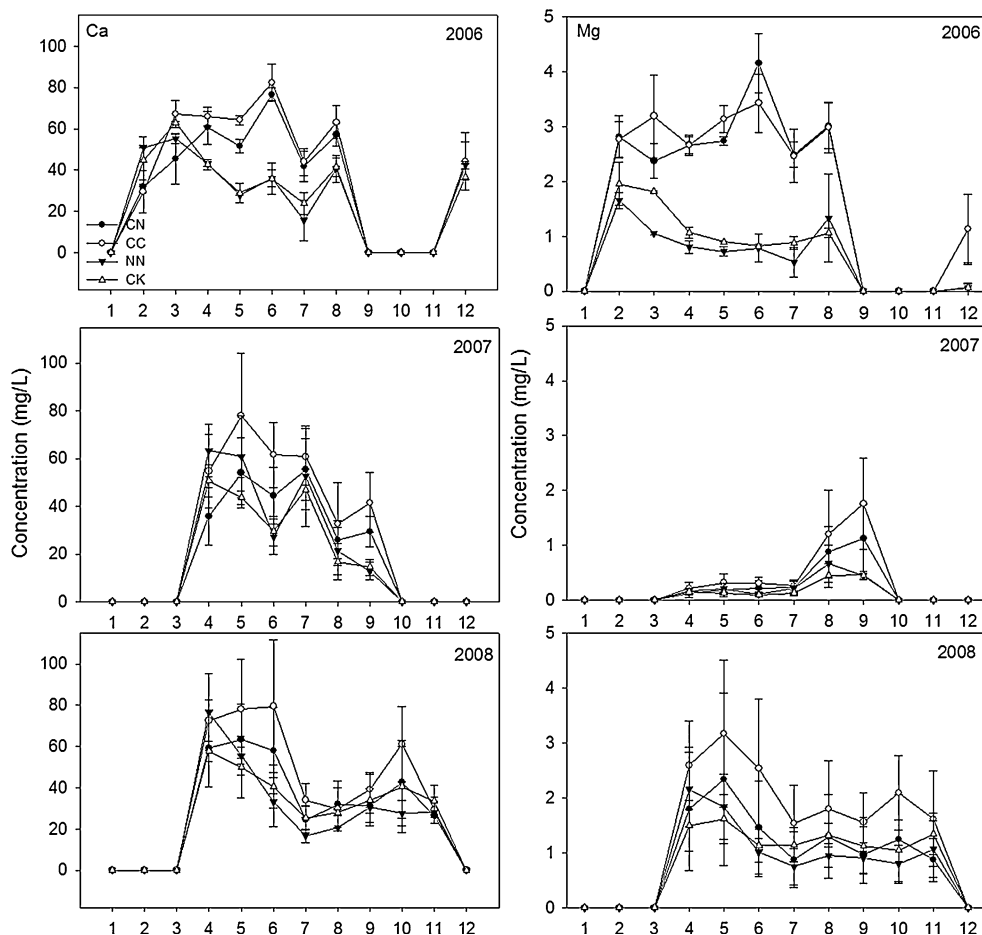
Annual net inorganic C leaching losses were calculated by summing up the C losses from  $\text{HCO}_3^-$ -C and  $\text{CO}_3^{2-}$ -C. Annual inorganic C leaching losses were highest in the CC treatments with 48.0, 49.5, and 114.0  $\text{kg ha}^{-1} \text{y}^{-1}$  in 2006, 2007, and 2008, respectively, compared with 33.8, 28.4, and 62.8  $\text{kg ha}^{-1} \text{y}^{-1}$  in the CK treatment in the corresponding years. There were 86.5 and 24.1  $\text{kg ha}^{-1}$  increases of inorganic C loss in the CC and CN treatments compared to the CK treatment during the 3 years. However, there was a decrease of 11.75  $\text{kg ha}^{-1}$  in inorganic C loss in the NN treatment when compared to the CK treatment.

## Ca and Mg in the Leachates

Like the concentration of  $\text{HCO}_3^-$ -C in the leachates, concentrations of Ca and Mg in the leachates were also significantly affected by both  $\text{CO}_2$  and N treatments ( $P < 0.001$ ). The CC treatment had greater concentrations of Ca and Mg in the leachates than the other treatments during the experimental period (Figure 5). The N treatment reduced the positive effect of  $\text{CO}_2$  on the Ca and Mg leaching losses. The Ca and Mg concentrations were positively correlated to the  $\text{HCO}_3^-$ -C concentrations ( $R^2 = 0.55$  and  $R^2 = 0.35$ ,  $P < 0.05$ , respectively).

## Tree Growth

Tree growth as basal diameter and height in our experiment is shown in Figure 6. Treatments did not affect the basal diameter of trees during the whole experiment period. However, the height of trees followed the order: CN > NN = CC > CK. Elevated  $\text{CO}_2$  and high N treatment increased the tree height, but the intensity of the treatment effect



**Figure 5.** Concentrations of Ca (*left column*) and Mg (*right column*) in leachates under various CO<sub>2</sub> and N treatments. Error bars are standard deviations ( $n = 9$  for CC and CN,  $n = 6$  for NN and CK). Treatments are: CK = control; NN = high N treatment, CC = elevated CO<sub>2</sub> concentration treatment; CN = elevated CO<sub>2</sub> concentration treatment + high N treatment.

changed with time (Figure 6). In the short term (the first 6 months), there was no significant difference among the treatments. From the end of 2005 to the end of 2007, the CN treatment showed greater height than the other treatments. During the fourth growth year, there was no significant difference among the treatments again.

## DISCUSSION

### Soil C

Among the treatments, greater soil organic C was observed in the CN treatment when compared to the other treatments. This result is in accordance with the previous studies (Luo and others 2006), which showed additional N supply enhanced the effects of CO<sub>2</sub> on C accumulations in the soil pools. The high CO<sub>2</sub> treatment alone did not affect soil organic C content, and this supports the findings of Schlesinger and Lichter (2001) and Williams and others (2003), and adds to the uncertainty about whether the soil C pool would act as a net sink for atmospheric CO<sub>2</sub> under future increasing atmospheric CO<sub>2</sub> concentrations (Cox and others 2000).

Neither treatments nor sampling times affected soil inorganic C contents; this is in accordance with the finding of Williams and others (2003), where the higher dissolved inorganic C leaching induced by the elevated CO<sub>2</sub> than control did not change the soil inorganic C contents which were low, like those in our soils.

### Soil Moisture and Soil CO<sub>2</sub> Flux

Under elevated atmospheric CO<sub>2</sub>, higher soil moisture has been reported previously (Niklaus and others 1998; Morgan and others 2004). Increased soil moisture was also shown in the high CO<sub>2</sub> treatment chambers in our experiment. The decreased soil moisture in the N treatment was due to the increased plant growth under the N fertilizer condition (Figure 6; Duan and others 2009).

Increased soil CO<sub>2</sub> fluxes induced by the CO<sub>2</sub> treatment have been found in many studies (King and others 2001; Pendall and others 2001; Gill and others 2002; Suwa and others 2004; Bernhardt and others 2006). Increased root biomass and soil organic matter resulted in increased root and microbial respiration (de Graaff and others 2006).

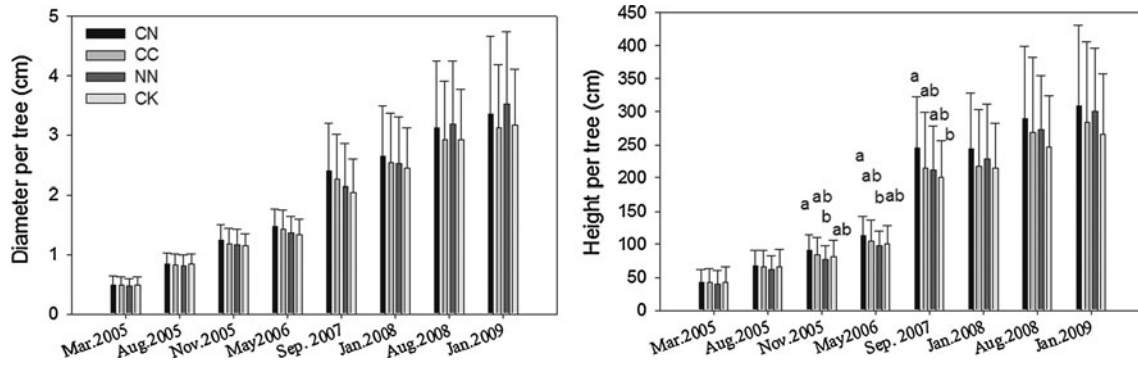


Figure 6. Growth of basal diameter (left column) and height (right column) of trees exposed to the various CO<sub>2</sub> and N treatments. Treatments are: CK = control; NN = high N treatment; CC = elevated CO<sub>2</sub> concentration treatment; CN = elevated CO<sub>2</sub> concentration treatment + high N treatment. Error bars are standard deviations ( $n = 75$  for CC and CN,  $n = 47$  for NN and CK). Different letters indicate significant differences at the confidence level of  $P < 0.05$  among the treatments.

This is the reason for the higher soil CO<sub>2</sub> fluxes under elevated CO<sub>2</sub> (de Graaff and others 2006). Greater soil organic matter was also shown in the CN treatments in our experiment. We also found increased tree growth in this treatment's chambers (Figure 6). The N treatment increased N availability in the soils for faster plant growth, which led to greater soil organic matter in the CN treatment than in the CC treatment. This would also translate to greater soil CO<sub>2</sub> flux rates in the CN treatment than in the CC treatment. Increased soil moisture in the CN and CC chambers also led to higher soil CO<sub>2</sub> fluxes in these treatments.

### Variations of Leaching Water Volumes and pH in Leachates

In our study, the volume of leachates varied greatly. There were three main reasons for these variations. The main and most obvious one was the rainfall. The volume of leachates showed a monthly pattern that closely matched the monthly rainfall ( $R^2 = 0.49$ ,  $P < 0.05$ ). The second factor for the variations was the high CO<sub>2</sub> treatments. High CO<sub>2</sub> concentrations in the CC and CN treatments resulted in a shift of the leaching pattern imposed by the rainfall toward greater leaching rates. This led to annual leaching volumes 69% and 30% greater in the CC and CN treatments, respectively, in 2006 and 18% and 20% in the CC treatment in 2007 and 2008, respectively, compared with those for the control. Increased leaching volumes in these treatments were probably related to the decrease of other water outputs such as lower evaporation and plant transpiration. Indeed, these two treatments also

showed the highest soil moisture contents. Nelson and others (2004) also found that elevated CO<sub>2</sub> increased soil moisture in a long-term field study and might lead to increased water drainage. They suggested that higher soil moisture under elevated CO<sub>2</sub> could cause a greater fraction of precipitation to be lost by run off or deep seepage, which is in accordance with our findings. Finally, the high N treatment was the third factor for the leachate volume variations. High N addition caused a decrease in leachate volume and with time, this decrease became more obvious. This is the consequence of better tree growth following N fertilization (Duan and others 2009). During the low rainfall months, the volume of leachates showed reduced differences between the treatments because less water was available for leaching due to increased plant water use.

In 2006, pH values of leaching water increased significantly. There are three reasons which would lead to the increase in soil water pH values. First, the rise in soil water pH was probably linked to the reduction in NO<sub>3</sub> leaching (Liu and others 2008). Secondly, high precipitation amounts and temperatures since April led to higher rates of weathering of soil minerals and soil organic matter decomposition, which produced more cations and accelerated the exchange of H<sup>+</sup> with cations. Generally, the seedlings would grow faster during the pioneer stage, which would also facilitate the weathering of soil minerals and soil organic matter decomposition. The chambers exposed to elevated CO<sub>2</sub> had higher pH values in the leachates than the chambers under ambient CO<sub>2</sub> in 2008. This is due to the higher weathering of carbonate minerals in these chambers.

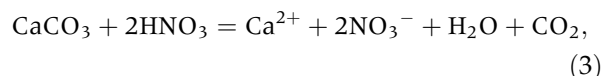
## Variations of Inorganic C Leaching Losses

In our study, the significant increases in  $\text{CO}_3^{2-}$ -C and  $\text{HCO}_3^-$ -C concentrations in the leachates that occurred in the high  $\text{CO}_2$  treatment chambers were due to the higher soil  $\text{CO}_2$  fluxes in the treatment. This result is consistent with the findings in the studies of Andrews and Schlesinger (2001) and Williams and others (2003). Increased concentrations of Ca and Mg were found in the high  $\text{CO}_2$  treatments, which also indicated that more chemical weathering of carbonate minerals happened in the high  $\text{CO}_2$  treatment chambers. The increased pH values in the leachate of the CC treatment in 2008 also support this result.

We noted that there are a few studies showing higher  $\text{HCO}_3^-$ -C concentrations in the soil water induced by elevated  $\text{CO}_2$  in forest ecosystems (Andrews and Schlesinger 2001; Williams and others 2003). But none of these could report the exact total amount of inorganic C ( $\text{CO}_3^{2-}$ -C and  $\text{HCO}_3^-$ -C) losses induced by the elevated  $\text{CO}_2$  due to experiment design limitations. In addition to the fact that there was a higher inorganic C concentration in the leachates in the high  $\text{CO}_2$  treatment chambers, we also found that there were greater leaching water amounts in the CC treatment chambers. This led to increased inorganic C leaching exports over the 3 years in these chambers. Compared to the CK chambers, higher soil  $\text{CO}_2$  together with the increased soil moisture evidenced in our study, created favorable conditions for chemical weathering (Vuai and Tokuyama 2007). Bicarbonate and free base cations are byproducts of carbonate mineral weathering and an increase of  $\text{HCO}_3^-$ -C loss is the sign of accelerated weathering.

With time, the high N treatment reduced the inorganic C loss with the leaching water. Compared to the CK chambers, lower inorganic C losses were found in the NN treatment chambers. Meanwhile, compared to the CC treatment chambers, decreased inorganic C losses were found in the CN treatment chambers. This was related to the lower inorganic C concentrations and less leaching water amounts in these chambers. Less leaching water amount was due to the increase in plant growth in these chambers (Figure 6; Duan and others 2009), which led to increased transpiration and hence decreased leachate volume. There are two reasons that would decrease inorganic C concentrations in the CN and NN treatments when compared to the CC and CK treatments, respectively. First, this is related to the massive atmospheric N deposition that already occurs in our research area. Plant roots could not

take up the continued addition of N fertilizer, which led to nitric acid production (Liu and others 2008). Nitric acid would generate protons that could be consumed in the weathering of carbonate minerals (equation 3), and this process might dominate the mineral weathering reaction; less bicarbonate would be then produced.



The lower pH values of the leachates found in the CN treatment, when compared to the CC treatment in 2008, supported this hypothesis. Secondly, increased plant growth in the CN and NN treatments (Figure 6; Duan and others 2009) consumed more soil water, which decreased the reactions shown in equations (1) and (2).

Increasing atmospheric  $\text{CO}_2$  and N deposition are two primary and concurrent changes in subtropical China. Elevated  $\text{CO}_2$  and N addition affected each other in stimulating plant growth (Duan and others 2009), which resulted in complex interactive effects on soil  $\text{CO}_2$  flux in our study shown in Deng and others (2010). High  $\text{CO}_2$  treatment increased soil  $\text{CO}_2$  fluxes and soil moisture contents, and then increased the reactions shown in equations (1) and (2), which led to more inorganic C loss. High N treatment decreased soil moisture and then the leaching water amount, which resulted in lower inorganic C loss. Thus, the positive effect of the high  $\text{CO}_2$  treatment on inorganic C loss was offset by the high N treatment, indicating a strong interactive effect of these two factors on inorganic C loss. They should be evaluated in combination in subtropical forest ecosystems in China where atmospheric  $\text{CO}_2$  and N deposition have been increasing simultaneously and remarkably.

## Importance of Inorganic C Losses for Mitigating Atmospheric $\text{CO}_2$ in Tropical Areas

Recently, the uncertainty about whether the soil C pool will act as a net sink for atmospheric  $\text{CO}_2$  under elevated  $\text{CO}_2$  has been reported by some studies (Cox and others 2000; Schlesinger and Lichten 2001; Hagedorn and others 2003; Lichten and others 2005). New sinks for anthropogenic atmospheric  $\text{CO}_2$  should be sought. Chemical weathering through the conversion of  $\text{CO}_2$  into its dissolved form which can then be sequestered in water has been considered as a net sink of atmospheric  $\text{CO}_2$  (Smedberg and others 2006; Oh and Raymond 2006; Gilfillan and others 2009).



After 2 years of observation in the Duke FACE forest, Andrews and Schlesinger (2001) reported that an increase of 55% in atmospheric CO<sub>2</sub> over 2 years resulted in a 33% increase in the flux of dissolved inorganic C to groundwater. However, after another 3-year study at the same research site, Oh and others (2007) showed that carbonic acid weathering was increased by less than that suggested by Andrews and Schlesinger (2001) as the results of linear mixed-effects models and soil heterogeneity studies showed. There was no significant interaction effect of elevated CO<sub>2</sub> concentration treatment and time detected in soil water chemistry over the 5 years of sample collection. This result is contrary to the findings in our study. Using the same models (linear mixed models), we found that there was a significant interaction effect of elevated CO<sub>2</sub> concentration treatment and sampling time on inorganic C loss via leaching water (Table 4). This could be due to the climate and soil type differences between the two study sites. Our results suggest that tropical forest soil systems may be able to compensate for a small part of the atmospheric CO<sub>2</sub> increase through accelerated processing of CO<sub>2</sub> into HCO<sub>3</sub><sup>-</sup>-C.

Unlike the previous studies, we found that the greater inorganic C loss induced by elevated CO<sub>2</sub> was related to both the increased concentration of inorganic C and the increased leaching water amount. When studying the effects of elevated CO<sub>2</sub> on soil weathering in the future, the change of soil water amount induced by the elevated CO<sub>2</sub> concentration treatment should be considered. In subtropical areas, where rainfall is not distributed evenly throughout the year, with extreme amounts of precipitation in wet seasons, this would increase the weathering of carbonate minerals in the soils in these areas.

There was about a 3 g C m<sup>-2</sup> y<sup>-1</sup> increase of inorganic C loss at elevated compared to ambient CO<sub>2</sub> conditions in our experiment. Using meta-analytic methods, Luo and others (2006) have summarized the results of 104 studies on plant biomass production from free air CO<sub>2</sub> enrichment (FACE) and open-top chamber experiments. They concluded that the averaged rate of C accumulation in land ecosystems is approximately 100 g C m<sup>-2</sup> y<sup>-1</sup> more at elevated than ambient CO<sub>2</sub>. Compared with the increased C size in plant, litter, and soil pools at elevated than ambient CO<sub>2</sub>, the C sink via weathering of soil carbonate is small, however, C stored in plants would finally enter into the ground and decompose at a relatively short temporal scale, and soil C sequestration would not be unlimited (Paustian and others 1997; Six and others 2002).

Atmospheric C that has been consumed in the chemical weathering process is exported as bicarbonate in part to the sea, where C is stored as CaCO<sub>3</sub> at the geological time scale (Smedberg and others 2006). Scaling the inorganic C (HCO<sub>3</sub><sup>-</sup>-C and CO<sub>3</sub><sup>2-</sup>-C) loss estimates for the model forest ecosystems under elevated CO<sub>2</sub> in our open-top chambers to the global tropical forests suggests a net increased sink of 0.26 × 10<sup>14</sup> g C y<sup>-1</sup>. Therefore, changes in soil water leaching of inorganic C loss are likely to buffer the changes in geologic processes that alter atmospheric CO<sub>2</sub>. However, as there is an artificial restriction of the chambers that we used in the study to the plant rooting zone, it is possible that we overestimate the effect of high CO<sub>2</sub> treatment on soil weathering.

## CONCLUSION

Elevated atmospheric CO<sub>2</sub> concentration resulted in increased soil weathering processes as indicated by the increased HCO<sub>3</sub><sup>-</sup>-C and mineral cations measured in the soil leaching water collected at the 70-cm depth. Elevated CO<sub>2</sub> concentration also increased soil moisture contents and resulted in greater volumes of leaching water during the high rainfall events. As a consequence, the elevated CO<sub>2</sub> concentration treatment caused higher inorganic C (HCO<sub>3</sub><sup>-</sup>-C and CO<sub>3</sub><sup>2-</sup>-C) losses by leaching. The annual net HCO<sub>3</sub><sup>-</sup>-C leaching losses were 47.7, 49.5, and 113.2 kg ha<sup>-1</sup> y<sup>-1</sup> in the chambers exposed to the elevated CO<sub>2</sub> concentration treatment in 2006, 2007, and 2008, respectively. The N treatment reduced the positive effect of the elevated CO<sub>2</sub> concentration treatment on the inorganic C loss in the leachates. The annual net HCO<sub>3</sub><sup>-</sup>-C leaching losses were 38.7, 40.9, and 68.5 kg ha<sup>-1</sup> y<sup>-1</sup> in the chambers exposed to both elevated CO<sub>2</sub> concentration and N treatments in 2006, 2007, and 2008, respectively. Increased inorganic C loss with the leaching water was indirectly due to the higher soil CO<sub>2</sub> fluxes and increased soil moisture contents under the elevated CO<sub>2</sub> concentration. The extreme amount of precipitation in wet seasons under the monsoon climate in subtropical areas would increase the inorganic C losses. The consumption of CO<sub>2</sub> to produce HCO<sub>3</sub><sup>-</sup>-C through soil chemical weathering may act as a net sink for CO<sub>2</sub> in subtropical areas. However, high N deposition in these areas would decrease the positive effect of elevated CO<sub>2</sub> concentrations on the consumption of atmospheric CO<sub>2</sub>/losses of HCO<sub>3</sub><sup>-</sup>-C via belowground water. There was a strong interactive effect of the high CO<sub>2</sub> and N treatment on inorganic C loss.



## ACKNOWLEDGMENTS

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