Impact of different fertilizers on ~~the~~ carbonate weathering in a typical karst area, Southwest China: a field column experiment

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Abstract: Carbonate weathering, as a significant vector for the movement of carbon both between and within ecosystems, ~~are~~ is strongly influenced by ~~anthropogenic perturbations such as~~ agricultural fertilization,~~. —~~ since the addition of fertilizers tends to change the chemical characteristics of soil such as the pH~~ value~~. Different fertilizers may exert a different impact on carbonate weathering, but ~~their~~ these discrepancies~~differences~~ are as of yet not ~~still~~ well-known~~ so far~~. In this study, a field column experiment was ~~employed~~ conducted to explore the responses of carbonate weathering to the addition of different fertilizers~~ addition~~. ~~The eleven different treatments with three replicates including control, NH_4NO_3, NH_4HCO_3, $NaNO_3$, NH_4Cl, $(NH_4)_2CO_3$, $Ca_3(PO_4)_2$, $(NH_4)_3PO_4$, fused calcium-magnesium phosphate fertilizer (Ca-Mg-P), Urea and K_2CO_3 were established in this column experiment, where limestone and dolostone tablets were buried at the bottom of each to determine the weathering amount and ratio of carbonate in soil.~~ We compared 11 different treatments, including a control treatment, using 3 replicates per treatment. Carbonate weathering was assessed by measuring the weight loss of limestone and dolostone tablets buried at the bottom of ~~the~~soil-filled columns. The results show~~ed~~ that the addition of urea, NH_4NO_3, NH_4HCO_3, NH_4Cl and $(NH_4)_2CO_3$ distinctly increased carbonate weathering, which was attributed to the nitrification of NH_4~~$^+$, ~~$^+$. ~~and t~~The addition of $Ca_3(PO_4)_2$, Ca-Mg-P and K_2CO_3 induced carbonate precipitation due to the common ion effect. ~~Whereas the~~The addition of $(NH_4)_3PO_4$ and $NaNO_3$ ~~addition~~ did not significantly impact ~~significantly on~~ carbonate weathering. The results of

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NaNO$_3$ treatment ~~seem to be raising~~raise a new question: the negligible~~little~~ impact of nitrate on carbonate weathering may result in ~~the~~ overestimation of the impact of N-fertilizer on CO$_2$ consumption by carbonate weathering at the regional/global scale, if the effects of NO$_3$ and NH$_4$ are not distinguished. ~~Moreover, in order to avoid misunderstanding more or less, the statement that nitrogenous fertilizer can aid carbonate weathering should be replaced by ammonium fertilizer.~~

Keywords: Carbonate weathering; Column experiment; Nitrogenous fertilizer; Phosphate fertilizer; Southwest China

1. Introduction

Carbonate weathering plays a significant role in consumption of ~~the elevated~~ atmospheric CO$_2$ (Kump et al., 2000; Liu et al., 2010; ~~Liu et al.,~~ 2011)~~(Kump et al., 2000; Liu et al., 2010; 2011)(Kump et al., 2000; Liu et al., 2010; 2011)~~. ~~The r~~Riverine hydro-chemical composition, such as the ratio of HCO$_3^-$ ~~and~~ to Ca^{2+} + Mg^{2+}, is usually employed as an indicator to estimate the CO$_2$ consumption by natural carbonate weathering at the regional/global scale (Hagedorn and Cartwright, 2009; Li et al., 2009)~~(Hagedorn and Cartwright, 2009; Li et al., 2009)(Hagedorn and Cartwright, 2009; Li et al., 2009)~~. However, fluvial alkalinity may also be produced by other processes including the reaction between carbonates and the protons derived from: (i) ~~from~~ the nitrification of N-fertilizer ~~a disturbance to CO$_2$ consumption estimation is introduced because the fluvial alkalinity, Ca^{2+} and Mg^{2+} may also be~~

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~~produced due to the reaction between carbonate and the protons which can originate from the nitrification processes of N-fertilizer~~ (Barnes and Raymond, 2009; Chao et al., 2011; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-~~w~~Wickmann et al., 2009; Semhi and Suchet, 2000; West and McBride, 2005)~~(Barnes and Raymond, 2009; Chao et al., 2011; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-wickmann et al., 2009; Semhi and Suchet, 2000; West and McBride, 2005)(Barnes and Raymond, 2009; Chao et al., 2011; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-wickmann et al., 2009; Semhi and Suchet, 2000; West and McBride, 2005)~~~~,~~; (ii) ~~from the~~ sulfuric acid (Lerman and Wu, 2006; Lerman et al., 2007; Li et al., 2008; ~~Li et al.,~~ 2009)~~(Lerman and Wu, 2006; Lerman et al., 2007; Li et al., 2008; Li et al., 2009)(Lerman and Wu, 2006; Lerman et al., 2007; Li et al., 2008; Li et al., 2009)~~~~,~~; (iii) ~~from~~ organic acid secreted by microorganisms (Lian et al., 2008)~~(Lian et al., 2008)(Lian et al., 2008)~~;~~,~~ ~~as well as~~and (iv) ~~from~~ acidic soil (Chao et al., 2014;~~ Chao et al.,~~ 2017)~~(Chao et al., 2014)(Chao et al., 2014)~~. Given ~~the~~ that atmospheric CO_2 is not ~~the~~ a unique weathering agent, differentiating the agent of carbonate weathering is ~~more and more significant to enable~~important for the accurate budgeting of ~~the~~ net CO_2 consumption by carbonate weathering, especially in agricultural areas where mineral fertilizers are used~~area~~.

The ~~world~~ global average annual increase in mineral fertilizer consumption was 3.3 % from 1961 to 1997, and FAO's study predicts a 1 % increase per year until 2030 (FAO, 2000). ~~For~~ In China, the consumption of chemical fertilizer increased

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from 12.7 Mt in 1980 to 59.1 Mt in 2013 (Fig. 1). The ~~Increasing~~ increasing consumption of ~~chemical~~ mineral fertilizers is a significant disturbance factor ~~of~~ in carbonate weathering and the carbon cycle. ~~Many~~ Several studies have ~~showed~~ shown that nitrogen fertilizer additions ~~aided~~ increased ~~in the dissolution of lime~~ weathering rates, and also increased the total export of DIC from agricultural watersheds (Barnes and Raymond, 2009; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-~~w~~Wickmann et al., 2009; Probst, 1986; Semhi and Suchet, 2000; West and McBride, 2005)~~(Barnes and Raymond, 2009; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-wickmann et al., 2009; Probst, 1986; Semhi and Suchet, 2000; West and McBride, 2005)(Barnes and Raymond, 2009; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-wickmann et al., 2009; Probst, 1986; Semhi and Suchet, 2000; West and McBride, 2005)~~. According to ~~the estimation from~~estimates by Probst (1988) and Semhi et al. (2000), the contribution of N-fertilizers to carbonate dissolution ~~represents~~ was 30 % and 12-26 %~~, respectively,~~ ~~on~~in two small agricultural carbonate basins in south-western France, the Girou and the Gers, respectively (~~subtributary and~~ tributar~~y~~ies of the Garonne ~~river~~River~~, respectively~~). ~~For~~ In ~~lager basin level, such as~~ the Garonne ~~river~~ River ~~basin~~Basin, which is a large~~r~~ basin (52,000 km^2), this contribution was estimated at 6 % by Semhi et al. (2000). ~~At national and global scales,~~ Perrin et al. (2008) estimated that the ~~deficit~~ contribution of ~~CO$_2$ uptake due to~~ N-fertilizer ~~addition~~ (usually in form of NH$_4$NO$_3$) represents up to 5.7-13.4 % and ~~only~~ 1.6-3.8 %~~–~~ ~~of the total CO$_2$ flux~~

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~~naturally consumed by~~~~to~~of the carbonate dissolution~~,~~ ~~for~~in France and across the ~~on a~~ ~~g~~global ~~scale~~, respectively.

The~~se estimated results~~ estimates described above ~~were~~are ~~usually~~largely based on calculations that assumed~~ing that~~ a single type of fertilizer (e.g. $(NH_4)_2SO_4$, NH_4NO_3, or NH_4Cl) was used throughout the whole basin that was considered. However~~were usually based on a hypothesis of individual fertilizer (e.g. $(NH_4)_2SO_4$, NH_4NO_3, or NH_4Cl) input into an agricultural basin. Nevertheless~~, in actual agricultural practices, ~~at an agricultural basin,~~ different fertilizers are usually added for different crops~~ in actual agricultural practices~~. The impact of these fertilizers on carbonate weathering and riverine chemical composition may be different. ~~For~~ In the case of nitrogenous fertilizer, 100 % NO_3^- produced after the addition $(NH_4)_2SO_4$ and NH_4Cl is derived from the nitrification of NH_4^+, whilst comparatively, it is only 50 % after the addition of NH_4NO_3. ~~The d~~Differences ~~of~~ in NO_3^- sources may ~~cause the~~produce an evaluated deviation ~~of~~ in the impact of N-fertilizer addition on CO_2 consumption by carbonate weathering~~. Because~~, since the addition of different N-fertilizers (e.g. $(NH_4)_2SO_4$, NH_4NO_3, NH_4Cl, $NaNO_3$ or urea) may result in different contributions to carbonate weathering and relative products such as HCO_3^-, Ca^{2+} and Mg^{2+}. For phosphate fertilizer, the coprecipitation of phosphate ions with calcium carbonate may inhibit carbonate weathering (Kitano et al., 1978)~~(Kitano et al., 1978)(Kitano et al., 1978)~~. We ~~suppose~~ assume that the response of carbonate weathering to the addition of different fertilizers, such as N-fertilizer (NH_4 and NO_3), P-fertilizer and Ca/Mg fertilizer, may display differences, which ~~is~~ are so far poorly known, ~~so far~~ but likely significant. Here we sought to fully ~~well~~ understand the agricultural ~~force~~ impact on natural carbonate weathering, and to accurately evaluate

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the CO_2 consumption via carbonate weathering in agricultural areas.

~~Moreover, Tt~~The carbonate-rock-tablet test is used to determine the weathering rate of carbonate rock/mineral from the laboratory to the field (Chao et al., 2011; Chao et al., 2014; Dreybrodt et al., 1996; Gams, 1981; Gams, 1985; Jiang and Yuan, 1999; Liu and Dreybrodt, 1997; Plan, 2005; Trudgill, 1975)~~(Chao et al., 2014; Chao et al., 2011; Dreybrodt et al., 1996; Gams, 1981; Gams, 1985; Jiang and Yuan, 1999; Liu and Dreybrod, 1997; Plan, 2005; Trudgill, 1975)(Gams, 1981; Chao et al., 2011; Trudgill, 1975; Chao et al., 2014; Dreybrodt et al., 1996; Gams, 1985; Jiang and Yuan, 1999; Liu and Dreybrod, 1997; Plan, 2005)~~. In the laboratory, the carbonate-rock-tablet is employed to study the kinetics of calcite dissolution/precipitation (Dreybrodt et al., 1996; Liu and Dreybrodt, 1997)~~(Dreybrodt et al., 1996; Liu and Dreybrod, 1997)(Dreybrodt et al., 1996; Liu and Dreybrod, 1997)~~ and determine the rate of carbonate mineral weathering in the soil column (Chao et al., 2011)~~(Chao et al., 2011)(Chao et al., 2011)~~. However, in the field, it is also used to observe the rate of carbonate weathering and estimate~~d~~ CO_2 consumption ~~by carbonate weathering~~ (Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013; Plan, 2005)~~(Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013; Plan, 2005)(Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013; Plan, 2005).~~ ~~Although~~ Liu (2011) argued that the carbonate-rock-tablet test may lead to ~~the~~ deviations ~~of~~ in estimated CO_2 consumption by carbonate weathering at the regional/global scale, in ~~the~~ cases ~~of~~where there are insufficient representative data (Liu, 2011)~~(Liu, 2011)(Liu, 2011);~~. ~~our results show that~~yet ~~i~~It is nonetheless ~~a~~the preferred ~~option~~method for the ~~condition controlled contrast~~comparative or stimulated experiment (Chao et al., 2011; ~~Chao et al.,~~ 2014; ~~Chao et al.,~~ 2017) ~~(Chao et al., 2014; Chao et al., 2011)(Chao et al., 2011; Chao et al., 2014);~~. ~~Where the result~~

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~~from the carbonate-rock-tablet test is consistent to the major element geochemical data of leachates from soil column(Chao et al., 2011).~~

~~Thus~~Therefore~~, in order to observe their difference between the impacts of different fertilizer addition on carbonate weathering in soil, a ~~A field column experiment that involved embedding carbonate~~–~~-rock-~~–~~tablets ~~with eleven different treatments ~~was carried out in a typical karst area of southwest China, in order to observe the impact~~s~~ of different fertilizer additions on carbonate weathering in soil.

2. Materials and Methods

2.1 The study site

This study was carried out in a typical karst area, namely the ~~HuaXi ~~Huaxi ~~district ~~District of Guiyang ~~city~~City, Guizhou ~~province~~Province, SW China (26°23′N, 106°40′E, 1094 m ~~asl~~ASL). Guiyang, the capital city of Guizhou Province, is located in the central part of ~~The ~~the ~~Province~~province, covering an area from 26°11′00″ to 26°54′20″N and 106°27′20″ to 107°03′00″E (~~about~~approximately 8,000 km^2), with elevations ranging from 875 to 1655 m ~~above mean sea level~~ASL. Guiyang has a population of more than 1.5 million people, a ~~high ~~wide diversity of karstic landforms, ~~a ~~high elevations and low latitude, with a subtropical warm-moist climate, and an average annual ~~average ~~temperature of 15.3 ℃ and annual precipitation of 1200 mm (Lang et al., 2006)~~(Lang et al., 2006)(Lang et al., 2006)~~. A monsoonal climate often results in high precipitation during summer, with~~ and ~~ much less during winter, although the humidity is often high ~~during ~~throughout most of the year (Han and Jin, 1996)~~(Han and Jin, 1996)(Han and Jin, 1996)~~. Agriculture is a major land use in order to produce the vegetables and foods in the suburbs of Guiyang (Liu et al., 2006)~~(Liu~~

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~~et al., 2006)(Liu et al., 2006)~~. The consumption of chemical fertilizer increased from ~~0.8~~ 150 kg/ha ~~Mt~~ in 1980 to ~~1.0~~190 kg/ha ~~Mt~~ in 2013 (GBS, 2014)~~(GBS, 2014)(GBS, 2014)~~.

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2.2 Soil properties

The soil used in this column experiment was sampled from the B horizon (below 20 cm in depth) of yellow-brown soil ~~in~~from ~~dug from~~ a cabbage-corn or capsicum-corn rotation plantation in Huaxi ~~d~~District~~,~~. ~~It~~The soil was air-dried, ground to pass through a 2-mm sieve, mixed thoroughly and used for the soil columns. The soil pH (V_{soil}:V_{water} = 1:2.5) ~~were~~ was determined by pH meter. The chemical characteristics of the soil, in~~-~~cluding organic matter (OM), NH_4-N, NO_3-N, available P, available K, available Ca, available Mg, ~~available S and~~ available Fe, and available S were determined according to the Agro Services International (ASI) ~~M~~method (Hunter, 1980)~~(Hunter, 1980)(Hunter, 1980)~~~~,~~. OM was determined using an ~~where the~~ extracting solution ~~used for O.M.~~ contain~~ed~~ing 0.2 mol l^{-1} NaOH, 0.01 mol l^{-1} EDTA, 2 % methanol, and 0.005 % Superfloc 127~~,~~. NH_4-N, NO_3-N, available Ca, and Mg were determined ~~based~~ using ~~on~~ an ~~extraction~~ extracting solution of~~by~~ 1 mol l^{-1} KCl solution, whereas available K, P and Fe were determined using an ~~extracted by~~ extracting solution containing 0.25 mol l^{-1} $NaHCO_3$, 0.01 mol l^{-1} EDTA, 0.01 mol l^{-1} NH_4F, and 0.005 % Superfloc 127~~,~~. Finally, ~~and~~ available S was determined using an extracting solution of ~~ed by~~ 0.1 mol l^{-1} $Ca(H_2PO_4)_2$ and 0.005 % Superfloc 127. The results are shown in Table 1.

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2.3 Soil column and different fertilization treatments

In order to test the hypothesis that the ~~responses of the~~ impact of different chemical fertilizers on carbonate weathering may be different, columns (Ø = 20 cm, H= 15 cm) were constructed from 20~~-~~ cm diameter polyvinylchloride (PVC) pipe (Fig. 2). A hole (Ø = 2 cm) ~~were~~ was ~~established~~ placed at the bottom of each column to discharge soil water from the ~~of~~ soil column. A polyethylene net ~~mesh~~ (Ø 0.5 mm) was placed in the bottom of the columns to prevent ~~the~~ soil loss ~~of the filter material~~. A 2 cm thick filter ~~sand~~ layer, ~~with 2 cm thickness~~ including gravel, coarse sand and fine sand, was spread ~~on~~over the net.~~-~~ Two different carbonate rock tablets were buried in the bottom of each soil column (Fig~~-~~. 2). ~~According to~~Based on the common kinds of chemical fertilizers and the main objective of this study, eleven fertilization treatments, each with three replicates, were set up in the field column experiment. There are ~~were set up~~: (1) control without fertilizer (CK); (2) 43g NH_4NO_3 fertilizer (CF); (3) 85g NH_4HCO_3 fertilizer (NHC); (4) 91g $NaNO_3$ fertilizer (NN); (5) 57g NH_4Cl fertilizer (NCL); (6) 51g $(NH_4)_2CO_3$ fertilizer (NC); (7) 52g $Ca_3(PO_4)_2$ fertilizer (CP); (8) 15g $(NH_4)_3PO_4$ fertilizer (NP); (9) 44g fused calcium-magnesium phosphate fertilizer (Ca-Mg-P); (10) 32g Urea fertilizer (U); and (11) 10g K_2CO_3 fertilizer (PP). ~~To shorten the experiment time and enhance the effect of fertilization, the added amount of fertilizers in these treatments motioned above was increased to 30 times than its local practical amount (N fertilizer: 160 kg N ·ha^{-1}; P fertilizer: 50 kg P ·ha^{-1}; K fertilizer: 50 kg K ·ha^{-1}).~~ ~~The~~An aliquot of 6 kg of soil was weighed (bulk density = 1.3 g/cm^3), mixed ~~perfect~~throughly with one of the above fertilizers, ~~respectively,~~ and filled into its own column. This process was repeated for all three

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replicates of the 11 fertilizer treatments. The~~se~~ soil columns were placed at the field experiment site in Huaxi District, Guiyang~~ of Southwestern China~~ for a whole year.

2.4 The rate of carbonate weathering

Two different kinds of carbonate rock tablets (2 cm × 1 cm × 0.5 cm in size) were ~~established ~~placed in the bottom of each soil column to ~~explore ~~examine the rate of carbonate weathering in the soil. The two different kinds of carbonate rock collected from the karst area of Huaxi ~~d~~District were: (1) limestone with 60-65 % micrite, 30-35 % microcrystalline calcite, and 2-3 % pyrite; and (2) dolostone with 98-99 % ~~power ~~fine crystalline dolomite, ~~3-5% microcrystalline calcite, ~~1 % pyrite, and ~~little~~ trace quantities organic matter. All ~~of ~~the tablets were ~~baked ~~heated at 80 ℃ for 4 hours, ~~then ~~weighed in a 1/10000 electronic balance in the laboratory, ~~tied to a ~~labeled by tying a label with fishing line, and then buried at the bottom of each soil column. After a whole year, ~~–T~~the~~y~~ tablets were ~~taken out~~removed carefully, rinsed, baked and weighed~~ after a whole year~~.

The amount of weathering ~~carbonate weathering ~~(A~~c~~*w*), the ratio of ~~carbonate~~ weathering ~~–~~(R~~c~~*w*) and the rate of ~~carbonate ~~weathering (R*a*~~c~~*w*) for limestone and dolomite were calculated according to the weight difference of the tablets using the following formulas:

$$A\cancel{c}w = (Wi\text{-}Wf) \tag{1}$$

$$R\cancel{c}w = (Wi\text{-}Wf)/\ Wi \tag{2}$$

$$Ra\cancel{c}w = (Wi\text{-}Wf)/(S*T) \tag{3}$$

where W*i* is the initial weight of the carbonate~~-~~ rock~~-~~ tablets, W*f* is the~~ir~~ final weights,

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S is the surface area of carbonate ~~weathering~~ rock tablets, and T is the length of the experimental period.

2.5 Statistical analysis

Statistical analysis was performed using IBM SPSS 20.0 (Statistical Graphics C~~r~~orp, Princeton, USA). All results of carbonate weathering were reported as the means ± standard deviations (SD)~~means standard errors (SE)~~ for the three replicat~~ion~~es. ~~as~~~~Statistical analysis was performed using IBM SPSS 20.0 (Statistical Graphics Crop, Princeton, USA).~~

3. Results

3.1 ~~The w~~Weathering rate of carbonate under different fertilized treatments

~~weathering of under different treatments~~The ~~amount (AR~~*c*~~w),~~ and ~~the ratio (~~R*ac*w ~~) and the rate (Racw)~~ of limestone and dolostone~~carbonate weathering~~~~we~~are listed –in Table 2. ~~were listed~~ The results showed that ~~in Table 2, and the Rcw were plotted in Fig. 3. The results in Table 2 and Fig. 3 The results showed T~~the ~~Acw,~~ R~~c~~*w* ~~and Racw~~ of ~~carbonate weathering~~limestone ~~and dolomite weathering~~ under urea, NH_4NO_3, NH_4Cl, $(NH_4)_2CO_3$~~NH_4HCO_3, NH_4Cl~~ and NH_4HCO_3 ~~$(NH_4)_2CO_3$~~ treatments were 8.48 ±0.96, 6.42 ±0.28, 5.54 ±0.64, 4.44 ±0.81 and 4.48 ±0.95 ‰ ~~(mean ±SD, $p<0.05$) positive~~, ~~and much bigger~~significantly greater than that under the control treatment 0.48 ±0.14 ‰ (see Fig. 3~~,~~). In addition, the observed R*w* of ~~as observed in~~ dolo~~mite~~stone were ~~(~~ 6.59 ±0.67, 5.30 ±0.87, 4.77 ±0.78, 4.94 ±1.91 and 3.22 ±0.87 ‰ respectively, under these same five fertilization treatments, in contrast to ~~vs.~~

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-0.31 ± 0.09 ‰ in the control treatment~~)~~. This ~~suggesting~~ ~~manifested~~clearly demonstrates that the addition of these five fertilizers ~~can aid and~~ increased the rate of ~~the chemical weathering of~~ carbonate weathering~~.~~ .

The re~~st~~maining treatments ~~had~~made no significant differences in the R*w* and R~~*c*~~*aw* of limestone and dolo~~mite~~stone in comparison ~~with~~ to the control treatment (Fig. 3). ~~In~~ In the $(NH_4)_3PO_4$ treatment, the ~~A*cw*~~R*w*~~, and R*acw*~~ were only 1.08 ± 0.34 ‰, ~~0.0028g~~ and ~~0.0007~~0.75 ± 0.21‰~~g~~ for limestone and dolomite, respectively, while the R*aw* were 4.00 ± 1.15 g m^{-2} a^{-1} ~~1.08‰~~ and 1.00 ± 1.01~~0.75~~ g m^{-2} a^{-1}~~‰~~ for limestone and dolomite, respectively~~,~~. These values are less than those under the other four NH_4-fertilizers, as mentioned above. The ~~A*cw*, R*cw*~~ R*w* and R*a*~~*c*~~*w* in the $NaNO_3$ treatments failed to show ~~a remarkable~~ notable differences with the control treatment, ~~implying~~ exhibiting little effect of the $NaNO_3$ fertilizer addition on carbonate weathering (Fig. 3). –

~~However, e~~Except for the R~~*c*~~*w* of limestone approaching zero in the $Ca_3(PO_4)_2$ treatment~~ approaching zero~~, all the values of ~~the A*cw*,~~ R~~*c*~~*w* and R~~*ac*~~*aw*~~ of two different carbonate~~ in Ca-Mg-P~~ and~~, K_2CO_3 and $Ca_3(PO_4)_2$ treatments showed ~~a~~ negative values~~,~~. This ~~indicating~~ indicates that the addition of Ca-Mg-P~~-~~, K_2CO_3 and $Ca_3(PO_4)_2$ fertilizers ~~can lead~~led to ~~the~~ precipitation at the surface of the carbonate mineral, which can be explained by common ion effect.

3.2 ~~The c~~Comparison of limestone of dolomite

In Fig. 4, we plotted the R*w* of~~In order to compare~~ ~~The result of~~ limestone ~~with~~vs.

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dolo~~mite~~stone tablets in a~~, we plotted Fig. 4, a~~ linear corre~~c~~lation diagram, in order to compare the weathering responses of ~~with the Rw of limestone vs. dolostone tablets~~limestone with dolostone. The results show~~s~~ that the R*w* of limestone and dolostone exhibit~~s~~ a high positive correla~~c~~tion (R^2=0.9773~~,~~; see Fig. 4), ~~suggest~~indicating that the weathering of ~~ANOVA was use the~~ limestone and dolostone are similar under different treatments~~ were similar~~. Thus, we will explain the results ~~with~~ in terms of carbonates, rather than~~ instead of~~ by way of the individual dolostone and limestone. ~~was used to determine the differences of weathering rate between limestone and dolostone.~~

4. Discussion

~~4.1 The carbonate rock tablet test: the validation of this experiment~~

~~The carbonate-rock-tablet test is used to determine the weathering rate of carbonate rock/mineral from laboratory to field (Gams, 1981; Chao et al., 2011; Trudgill, 1975; Chao et al., 2014; Dreybrodt et al., 1996; Gams, 1985; Jiang and Yuan, 1999; Liu and Dreybrod, 1997; Plan, 2005). In laboratory, the carbonate-rock-tablet is employed to study the kinetics of calcite dissolution/precipitation (Dreybrodt et al., 1996; Liu and Dreybrod, 1997) and determine the rate of carbonate mineral weathering in soil column (Chao et al., 2011). However, in field, it is also used to observe the rate of carbonate weathering and estimated CO$_2$ consumption by carbonate weathering (Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013; Plan, 2005). Although Liu (2011) argue that the carbonate-rock-tablet test may lead to the deviation of~~

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~~estimated CO_2 consumption by carbonate weathering at the regional/global scale in the case of insufficient representative data (Liu, 2011), our results show that it is a preferred option for the condition controlled contrast or stimulated experiment (Chao et al., 2011; Chao et al., 2014), where the result from the carbonate-rock-tablet test is consistent to the major element geochemical data of leachates from soil column(Chao et al., 2011).~~

~~In this study, every procedure to establish soil column with carbonate rock tablets in the bottom of each was strictly same, including the size of column, the preparation and column filling of soil sample, the setting and test of carbonate rock tablets, etc. Moreover, three replicates of each treatment were designed. We consider the experiment design can meet the objective of this study and the results of carbonate-rock-tablet test are therefore valid and credible.~~

4.1 ~~The k~~Kinetics of carbonate dissolution/precipitation: controlling factors

Experimental studies of carbonate dissolution kinetics have shown metal carbonate weathering usually depends upon three parallel reactions occurring at the carbonate interface (Chou et al., 1989; Plummer et al., 1978; Pokrovsky et al., 2009)~~(Chou et al., 1989; Plummer et al., 1978; Pokrovsky et al., 2009)~~:

$$MeCO_3 + H^+ \leftrightarrow Me^{2+} + HCO_3^- \qquad (4)$$

$$MeCO_3 + H_2CO_3 \leftrightarrow Me^{2+} + 2HCO_3^- \qquad (5)$$

$$MeCO_3 \leftrightarrow Me^{2+} + CO_3^{2-} \qquad (6)$$

where Me = Ca, Mg. As Eq. (5) describes, atmospheric/soil CO_2 is usually ~~regard~~considered to be ~~as~~ the natural weathering agent of carbonate. In watersheds

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with calcite- and dolomite-containing bedrock, H_2CO_3 formed in the soil zone usually reacts with carbonate minerals, resulting in dissolved Ca, Mg, and HCO_3^- as described in Eq. (5) (Andrews and Schlesinger, 2001; Shin et al., 2014)~~(Andrews and Schlesinger, 2001; Shin et al., 2014)~~. Although it has been proven that the reaction of carbonate dissolution is mainly controlled by the amount of rainfall (Amiotte Suchet et al., 2003; Egli and Fitze, 2001; Kiefer, 1994)~~(Amiotte Suchet et al., 2003; Egli and Fitze, 2001; Kiefer, 1994)~~, we consider that the effect of rainfall is equal in each soil column, and hence is disregarded ~~unconsidered~~ as a controlling factor in this study. ~~The~~ Eq. (4) suggests that the proton from other origins, such as the nitrification processes of NH_4^+, as mentioned in the ~~i~~Introduction section, can play the role of weathering agent in agricultural areas. In this study, the urea, NH_4NO_3, NH_4HCO_3, NH_4Cl, and $(NH_4)_2CO_3$ amendments increased (10 to 17-fold) the natural weathering rate ~~of~~from 2.00 g m^{-2} a^{-1} ~~from~~for limestone tablets in the control treatment (~~t~~Table 2). Thus, these increases are strongly rela~~tive~~ted to the effect of ~~the~~ proton release~~d~~ from the nitrification of NH_4^+. ~~On~~In ~~the~~ contra~~ry~~st, ~~the~~ carbonate precipitation will occur ~~as~~ due to the backward reaction of ~~the~~ Eq. (5) in the following cases: (1) the degassing of dissolved CO_2~~,~~; (2) soil evapotranspiration; or (3) the common ion effect: the increase of Ca^{2+}, Mg^{2+} or CO_3^{2-} in a weathering-system with equilibrium between water and calcite (Calmels et al., 2014; Dreybrodt, 1988)~~(Calmels et al., 2014; Dreybrodt, 1988)~~.

4.2 ~~The m~~Main reactions and effects in different treatments

The main reactions and effects of every treatment in this study ~~we~~are listed in

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Table 3.

(1) ~~The n~~Nitrification in NH_4-fertilizer: NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea

In urea ($CO(NH_2)_2$) treatment, the enzyme urease rapidly hydrolyzes the urea-N ($CO(NH_2)_2$) to NH_4^+ ions (Eq. (7)) when urea is applied to the soil (Soares et al., 2012)~~(Soares et al., 2012)~~.

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$$CO(NH_2)_2 + 3H_2O \rightarrow 2NH_4^+ + 2OH^- + CO_2 \qquad (7)$$

Although the study ~~from~~of Singh et al. showed that ~~a~~ part of NH_4^+ may be lost as ammonia (NH_3) and subsequently as nitrous oxide (N_2O) (Singh et al., 2013)~~(Singh et al., 2013)~~, ~~yet~~ the ~~rest~~remaining ammonium (NH_4^+) is mainly oxidized during nitrification in soil by autotrophic bacteria ~~(,~~ ~~like~~such as Nitrosomonas,~~) during nitrification,~~ resulting in nitrite NO_2^- and H^+ ions. Nitrite is~~,~~ –in turn~~,~~ oxidized by another bacterium, such as Nitrobacter, resulting in nitrate (NO_3^-) (Eq. (8)) (Perrin et al., 2008)~~(Perrin et al., 2008)~~.

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$$NH_4^+ + 2O_2 \rightarrow NO_3^- + H_2O + 2H^+ \qquad (8)$$

The protons (H^+) produced by nitrification can be neutralized in two ways:

(i) either by exchange process with base cations in the soil exchange complex (Eq. (9))

$$Soil - Ca + 2H^+ \rightarrow Soil - 2H^+ + Ca^{2+} \qquad (9)$$

or (ii) ~~or~~ via carbonate mineral dissolution (Eq.(10))

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$$Ca_{(1-x)}Mg_xCO_3 + H^+ \rightarrow (1-x)\,Ca^{2+} + xMg^{2+} + HCO_3^- \qquad (10)$$

Consequently, ~~after~~by combining Eq. (8) and Eq. (10) ~~are combined~~, carbonate weathering by protons produced by nitrification ~~is~~can supposedly be expressed as ~~to becomes~~ (Eq. 11) (See details in Perrin et al., 2008 and Gandois et al., 2011).

$$2Ca_{(1-x)}Mg_xCO_3 + NH_4^+ + 2O_2 \rightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 2HCO_3^- \quad (11)$$

As discussed above, provided that the loss as ammonia (NH_3) and nitrous oxide (N_2O) after hydrolyzation is ~~unconsidered~~disregarded in this study, the final equation of carbonate weathering in NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea treatments will be as follow~~ed~~s~~ as~~, respectively:

$$2Ca_{(1-x)}Mg_xCO_3 + NH_4NO_3 + 2O_2 \rightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + 2NO_3^- + H_2O + 2HCO_3^- \quad (12)$$

$$2Ca_{(1-x)}Mg_xCO_3 + NH_4HCO_3 + 2O_2 \rightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 3HCO_3^- \quad (13)$$

$$2Ca_{(1-x)}Mg_xCO_3 + NH_4Cl + 2O_2 \rightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + NO_3^- + Cl^- + H_2O + 2HCO_3^- \quad (14)$$

$$3Ca_{(1-x)}Mg_xCO_3 + (NH_4)_2CO_3 + 4O_2 \rightarrow 3(1-x)\ Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 2H_2O + 4HCO_3^- \quad (15)$$

$$3Ca_{(1-x)}Mg_xCO_3 + CO(NH_2)_2 + 4O_2 \rightarrow 3(1-x)\ Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 4HCO_3^- \quad (16)$$

(2) No effect of NO_3-fertilizer treatment: $NaNO_3$ treatment

In the $NaNO_3$ treatment, the reaction occurs according to ~~as~~ Eq. (17), indicating that the addition of NO_3-fertilizer does not significantly influence carbonate weathering.

$$Ca_{(1-x)}Mg_xCO_3 + NaNO_3 + CO_2 + H_2O \rightarrow (1-x)\ Ca^{2+} + xMg^{2+} + Na^+ + NO_3^- + 2HCO_3^- \quad (17)$$

(3) ~~The c~~Common ion effect: K_2CO_3 treatment

In the K_2CO_3 treatment, CO_3^{2-} and HCO_3^- will be produced after the addition of K_2CO_3 according to Eq. (18)~~ after adding K_2CO_3~~, hence resulting in carbonate

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precipitation as described in Eq. (19), due to the common ion effect.

$$K_2CO_3 + H_2O \rightarrow 2K^+ + HCO_3^- + OH^- \qquad (18)$$

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$$(1-x)\ Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O \qquad (19)$$

(4) Complex effects: Nitrification versus ~~I~~inhibition effect of PO_4 in $(NH_4)_3PO_4$ treatments

~~For~~In the $(NH_4)_3PO_4$ treatment, the reaction of carbonate weathering will occur according to Eq. (11) due to the nitrification of NH_4^+ ionized from the $(NH_4)_3PO_4$ fertilizer~~ will occur the nitrification~~. ~~Whilst t~~The PO_4^{3-} anion will exert an inhibition to calcite dissolution~~;~~, as calcium orthophosphate (Ca-P) precipitation is produce~~s~~d on the surface of calcite after the addition of PO_4^{3-} in soil (reaction: Ca + PO_4 → Ca-P), resulting in inhibiti~~ng~~ion of the calcite dissolution~~ of calcite~~.

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(5) Complex effects: Common ion effect versus ~~I~~inhibition effect of PO_4 in $Ca_3(PO_4)_2$ and Ca-Mg-P treatments

In the $Ca_3(PO_4)_2$ and Ca-Mg-P treatments, on the one hand, ~~the~~ $Ca_{(1-x)}Mg_xCO_3$ is produced~~s~~ when the concentrations of Ca^{2+} (or/and Mg^{2+}) increases~~ as~~ according to ~~following~~ Eq. (19). On the other hand, the inhibition effect of phosphate will cause ~~that~~ calcium phosphate precipitation to be produce~~s~~d on the surface of carbonate minerals after the addition of P in soil (reaction: Ca + PO_4 → Ca-P), ~~correspondingly~~ resulting in inhibiti~~ng~~ion the carbonate precipitation.

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4.3 ~~The d~~Difference between NH_4^+ and NO_3^- in impacts on carbonate weathering and ~~the~~ implication on the estimation of CO_2 consumption

In order to further compare the differenceS between NH_4^+ and NO_3^- effects on carbonate weathering, the initial molar amount of fertilizer-derived NH_4 per unit in

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every treatment were calculated, and are listed in Table 4. The results show that the amount of NH_4^+ hydrolyzed from urea is 1.06 mole, while NH_4^+ ionized from NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and $(NH_4)_3PO_4$ is 0.54 mole, 1.08 mole, 1.07 mole, 1.06 mole, and 0.03 mole, respectively (Table 4). The R*w* of limestone tablets and the initial amount of NH_4^+ per treatment are plotted in Fig. ~~4~~5. A distinct relationship between them is observed, in that~~:~~ the R*w* values in NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea treatments are~~ bigg~~ larger than in the control treatment, where the initial amount of NH_4^+ ~~displays~~yields similar results (Fig. ~~4~~5). This suggests that carbonate weathering in NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea treatments are mainly attributed to the dissolution reaction described as Eq. (11). This process of carbonate weathering by protons released from nitrification has been proven by many studies, from the laboratory to the field (Barnes and Raymond, 2009; Bertrand et al., 2007; Biasi et al., 2008; Chao et al., 2011; Errin et al., 2006; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Semhi and Suchet, 2000; West and McBride, 2005)~~(Barnes and Raymond, 2009; Bertrand et al., 2007; Biasi et al., 2008; Chao et al., 2011; Errin et al., 2006; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Semhi and Suchet, 2000; West and McBride, 2005)~~. We have noted that the R*w* values in NH_4HCO_3 and $(NH_4)_2CO_3$ treatments are ~~lower~~less than~~ even~~ half ~~of ~~those in urea treatment ~~in~~ despite~~ of~~ adding the same amount of fertilizer-derived NH_4 (~~about ~~approximately 1.07 mole). This is probably because the two fertilizers, NH_4HCO_3 and $(NH_4)_2CO_3$, are easier to decompose and produce ~~the ~~NH_3 and CO_2 gases ~~as following~~according to Eq. (20) and (21), resulting in ~~the ~~amounts of fertilizer-derived NH_4 ~~of~~that are lower than 1.07 moles.

$$NH_4HCO_3 \rightarrow NH_3\uparrow + H_2O + CO_2\uparrow \qquad (20)$$

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$$(NH_4)_2CO_3 \rightarrow 2NH_3\uparrow + H_2O + CO_2\uparrow \qquad (21)$$

The Aw and Rw in the $(NH_4)_3PO_4$ treatment, unlike in other NH_4-fertlizer treatments, ~~had not~~do not show a significant increase compar~~ing~~ed to~~ wi~~ the control treatment, which is not only owing to the low amount of added NH_4^+ in $(NH_4)_3PO_4$ treatment (0.3 mole~~,~~; see Table 4), but also more or less relative to the inhibition of phosphate (Chien et al., 2011; Wang et al., 2012)~~(Chien et al., 2011; Wang et al., 2012)~~. After the addition of $(NH_4)_3PO_4$ in soil, calcium orthophosphate (Ca-P) precipitation will form on calcite surfaces, which is initiated with the aggregation of clusters leading to the nucleation and subsequent growth of Ca-P phases, at various pH values and ionic strengths relevant to soil solution conditions (Chien et al., 2011; Wang et al., 2012)~~(Chien et al., 2011; Wang et al., 2012)~~.

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However, in Fig. 3, there is no significant different between the Rw ~~without significant difference with control treatment~~ in the $NaNO_3$ treatment compared to the control treatment, indicating~~es~~ that the addition of NO_3-fertilizer does not significantly influence carbonate weathering.

A notable issue herein is that the $NaNO_3$ treatment produces the same amount of NO_3^- (1.07 mole) as other NH_4 fertilizer (NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea), but it fails to impact on carbonate weathering, which ~~is ~~rais~~ing~~es a new problem. Eq. (5), usually considered as an expression for the natural weathering process of carbonate, is an important reaction~~ for~~ in understanding the kinetics process of carbonate dissolution in carbonate-dominated areas, where the molar ratio of HCO_3^- and Me^{2+} in the river ~~as an indicator~~ is usually used as an indicator to make estimat~~ion~~es of CO_2 consumption by carbonate weathering at the regional/global scale (Hagedorn and Cartwright, 2009; Li et al., 2009)~~(Hagedorn and Cartwright, 2009; Li et al., 2009)~~. ~~At~~In agricultural areas, the relationship between $(Ca+Mg)/HCO_3^-$ and

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NO_3^- is usually employed to estimate the contribution of N-fertilizer to riverine Ca^{2+}, Mg^{2+}, and alkalinity (Etchanchu and Probst, 1988; Jiang, 2013; Jiang et al., 2009; Perrin et al., 2008; Semhi and Suchet, 2000)~~(Etchanchu and Probst, 1988; Jiang, 2013; Jiang et al., 2009; Perrin et al., 2008; Semhi and Suchet, 2000)~~. In these studies, the nitrification described ~~as~~in Eq. (8) is usually considered as the unique origin of NO_3^-. According to the results of the $NaNO_3$ treatment in this study, the contribution of protons from nitrification to carbonate weathering may be overestimated, if anthropogenic NO_3^- is neglected, since the anthropogenic NO_3^- does not release the proton described ~~as~~in Eq. (8). For NH_4NO_3 fertilizer, ~~the (~~Eq. (12)~~)~~ shows that ~~the~~ two moles of Ca^{2+}+Mg^{2+}, NO_3^-, and HCO_3^- will be produced when one mole NH_4NO_3 reacts with 2 moles of carbonate, where only half of the NO_3^- originates from nitrification described as Eq. (8). This will result in a double overestimation o~~n~~f the contribution of ~~the~~ nitrification to carbonate weathering, and thus thereby mislead the estimation of CO_2 consumption ~~therein~~.

At regional scales, ~~I~~if different fertilizers are ~~added~~ simultaneously added to an agricultural area, the estimation of CO_2 consumption by carbonate weathering might became more complicated, since the mole ratios of Ca+Mg, HCO_3^-~~–~~, and/or NO_3^- between different fertilization treatments ~~is~~are different (see Table 3). Thus, the related anthropogenic inputs (e.g. Ca+Mg, NH_4, NO_3^-, HCO_3^-, etc.) need to be investigated to more accurately estimate the impact of fertilization on carbonate weathering and its CO_2 consumption.

4.4 The comparison with other studied results

The R*w* and R*aw* of limestone in the control treatment in this study ~~is~~were 0.48 ~~%~~‰ and 2.00 g m^{-2} a^{-1}, respectively.~~ which~~ These are ~~is~~ generally consistent with ~~the~~ observations of 0.51-32.97 g m^{-2} a^{-1} (for R*aw*) in Nongla, Guangxi, a karst area of

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Southwestern China (Zhang, 2011)~~(Zhang, 2011)~~, and with the results of 0.05-5.06 ‰ (for R*w*) and 1.08-136.90 g m^{-2} a^{-1} (for R*aw*) from the north slope of the Hochschwab ~~m~~Massif in Austr~~al~~ia (Plan, 2005)~~(Plan, 2005)~~, as determined –using the limestone tablet method. But the R*aw* of 2.00 g m^{-2} a^{-1} is lower than the results ~~(~~of 7.0-63.5 g m^{-2} a^{-1} for R*aw*~~)~~ from Jinfo Mount~~i~~ain in Chongqing~~ of~~, China (Zhang, 2011)~~(Zhang, 2011)~~. These differences in carbonate weathering are mainly attributed to the different types of carbonate rock tablets, climate, micro-environments of soil, etc. The R*aw* of limestone in the N-fertilizer~~s~~ treatments ~~is~~are 20.57-34.71 g m^{-2} a^{-1}, similar to the weathering rate of carbonate in ~~O~~orchard (32.97 g m^{-2} a^{-1}) at Nongla, Manshan, Guangxi~~ of~~, China, wh~~ere~~ich usually involves~~ in~~ fertilization activities.

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At larger scales, such as ~~–like~~ watersheds, the weathering rate is usually estimated by using the riverine hydro-chemical method, which is inconsistent with the results from the carbonate-rock-tablet test. ~~The estimation of~~ Zeng, et al. (2014) estimate~~views~~ that the carbon sink intensity calculated by the carbonate~~-~~-rock~~-~~-tablet test is only one sixth of that estimated ~~by~~ using the riverine hydro-chemical method, due to its own limits in methodology (Zeng et al., 2014)~~(Zeng et al., 2014)~~. The results from Semhi, et al. (2000) show~~s~~ th~~e~~at weathering rates of carbonate rock ~~by~~ using riverine hydro-chemical method are– ~~about~~approximately 77.5 g m^{-2} a^{-1} and 50.4 g m^{-2} a^{-1} in the upstream and downstream, respectively, of the Garonne river, France~~, respectively~~, which are ~~about~~approximately 25-35 ~~and 2-3~~ times greater than that in the control treatment (2.00 g m^{-2} a^{-1} for natural weathering rate-) and 2-3 times greater than in the N-fertilizer treatment (20.57-34.71 g m^{-2} a^{-1} for anthropic weathering rate) in this study. The global natural weathering rate of carbonate reported by Amiotte Suchet, et al. (2003) is 47.8 g m^{-2} a^{-1}, which is much higher than that we observed. Thus, we conclude that it is difficult to compare ~~between~~ the results

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from the carbonate-rock-tablet test and the riverine hydro-chemical method. The carbonate-rock-tablet test is suitable for ~~the~~ research on the ~~condition controlled~~comparative ~~contrast~~ or s~~t~~imulated experiments, while the riverine hydro-chemical method is appropriate for ~~the~~ regional investigations and estimations. According to the estimation from Yue et al. (2015), ~~T~~the enhanced HCO_3^- flux due to nitrification of NH_4^+ at Houzhai catchment of Guizhou ~~p~~Province would be 3.72 × 10^5 kg C/year and account for 18.7 % of this flux in the entire catchment(Yue et al., 2015)~~(Yue et al., 2015)~~. This is similar to estimates from other small agricultural carbonate basins (12–26 %) in ~~S~~southwest France (Perrin et al., 2008; Semhi and Suchet, 2000)~~(Perrin et al., 2008; Semhi and Suchet, 2000)~~.

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5. Conclusions

The impact of the addition of different fertilizers (NH_4NO_3, NH_4HCO_3, $NaNO_3$, NH_4Cl, $(NH_4)_2CO_3$, $Ca_3(PO_4)_2$, $(NH_4)_3PO_4$, Ca-Mg-P, ~~U~~urea, and K_2CO_3) on carbonate weathering was studied in a field column experiment ~~with~~using carbonate -rock~~-~~-tablets~~ at its bottom of each~~. The amount of weathering ~~amount~~ and the ratio of weathering of carbonate rock tablets showed that the addition of urea, NH_4NO_3, NH_4HCO_3, NH_4Cl, and $(NH_4)_2CO_3$ distinctly increased carbonate weathering, which was attributed to the nitrification of NH_4^+, ~~and~~while the addition of $Ca_3(PO_4)_2$, Ca-Mg-P and K_2CO_3 induced carbonate precipitation due to the common ion effect. ~~While t~~The addition of $(NH_4)_3PO_4$ and $NaNO_3$ ~~addition~~ did not impact significantly on carbonate weathering, where the former can be attributed to the low added amount of (NH4)$_3$PO$_4$,~~–~~ and may be related to the inhibition of phosphate, ~~and~~ while the latter seemed to ~~be~~ rais~~ing~~e a new question. The ~~little~~minor impact of nitrate on carbonate weathering may result in the overestimation of the impact of N-fertilizer on CO_2 consumption by carbonate weathering at the regional/global scale, if the effects of NO_3 and NH_4 are not distinguished. Thus, the related anthropogenic inputs (e.g. Ca+ Mg, NH_4, NO_3^-, HCO_3^-, etc.) need to be investigated to more accurately estimate the impact of fertilization on carbonate weathering and its consumption of CO_2 ~~consumption~~. ~~**4.2 The kinetics and controlled factors of carbonate weathering**~~

~~Experimental studies of carbonate dissolution kinetics have shown metal carbonate weathering usually depends upon three parallel reactions occurring at the carbonate interface (Chou et al., 1989; Plummer et al., 1978; Pokrovsky et al., 2009):~~

~~$MeCO_3 + H^+ \rightarrow Me^{2+} + HCO_3^-$ (4)~~

~~$MeCO_3 + H_2CO_3 \rightarrow Me^{2+} + 2HCO_3^-$ (5)~~

~~$MeCO_3 \rightarrow Me^{2+} + CO_3^{2-}$ (6)~~

~~where Me=Ca, Mg. As Eq. (5) describes, atmospheric/soil CO_2 is usually regard as the natural weathering agent of carbonate, whereas many studies have exposed that carbonate weathering can occur due to the reaction (Eq. (4)) between carbonate and the other proton contributors, as mentioned in introduction section: s which can originate from the nitrification processes of N-fertilizer~~H_4^+ ~~(Semhi and Suchet, 2000; West and McBride, 2005; Oh and Raymond, 2006; Hamilton et al., 2007; Perrin et al., 2008; Barnes and Raymond, 2009; Pierson-wickmann et al., 2009; Chao et al., 2011; Gandois et al., 2011), from the sulfuric acid~~ acid, ~~(Lerman and Wu, 2006; Lerman et al., 2007; Li et al., 2008; Li et al., 2009), from organic acid secreted by microorganisms (Lian et al., 2008), as well~~ and ~~as from acidic soil (Chao et al.,~~

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~~In field, carbonate dissolution is mainly controlled by the amount of rainfall (Amiotte Suchet et al., 2003; Egli and Fitze, 2001; Kiefer, 1994), as well as impacted of soil CO_2 (Andrews and Schlesinger, 2001). We consider that the effect of rainfall on each soil column is same. In this study, the urea, NH_4NO_3, NH_4HCO_3, NH_4Cl and $(NH_4)_2CO_3$ amendment increased (10 to 17-fold) the natural weathering rate of 2.00 g m^{-2} a^{-1} from limestone tablets in control treatment (table 2). These increases may be, in the one hand, attributed to the effect of the proton released from the nitrification of NH_4^+. On the other hand, it may be, in theory, related to enhanced microbiogenic CO_2 due to nitrogenous nutrients stimulation (Eq(5)), because fertilizer application can increase soil CO_2 flux (Sainju et al., 2008; Bhattacharyya et al., 2013), the increased CO_2 can enhance carbonate dissolution rate at near neutral or alkali pH (Andrews and Schlesinger, 2001).~~

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~~According to the added amount of different fertilization treatment, the molar amount of added nitrogen nutrient in $NaNO_3$ treatment is 1.07mol, much bigger than in NH_4NO_3, equivalent to NH_4HCO_3 and NH_4Cl treatment. However, the *Acw* and *Rcw*, and *Racw* of $NaNO_3$ treatment is far less (Fig. 3 and table 2), inhibiting that the increases of carbonate weathering rate in urea, NH_4NO_3, NH_4HCO_3, NH_4Cl and $(NH_4)_2CO_3$ amendment have no distinct relationship with enhanced microbiogenic CO_2 due to nitrogenous fertilizer amendment.~~

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~~4.3 The effect of nitrification of NH_4-fertilizer~~

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~~In urea ($CO(NH_2)_2$) treatment, the enzyme urease rapidly hydrolyzes the urea-N ($CO(NH_2)_2$) to NH_4^+ ions (Eq. (7)) when urea is applied to the soil (Soares et al.,~~

~~2012).~~

~~$$CO(NH_2)_2 + 3H_2O \longrightarrow 2NH_4^+ + 2OH^- + CO_2 \qquad (7)$$~~

~~Table 3 shows that the amount of NH_4^+ hydrolyzed from urea is 1.06 mol, while NH_4^+ ionized from NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and $(NH_4)_3PO_4$ is 0.54 mol, 1.08 mol, 1.07 mol, 1.06 mol and 0.03 mol, respectively (Table 3). Although the study from Singh et al showed that a part of NH_4^+ may be lost as ammonia (NH_3) and subsequently as nitrous oxide (N_2O) (Singh et al., 2013), yet the rest ammonium (NH_4^+) is mainly oxidized in soil by autotrophic bacteria (like Nitrosomonas) during nitrification, resulting in nitrite NO_2^- and H^+ ions. Nitrite is, in turn, oxidized by another bacterium, such as Nitrobacter, resulting in nitrate (NO_3^-) (Eq. (8)) (Perrin et al., 2008).~~

~~$$NH_4^+ + 2O_2 \longrightarrow NO_3^- + H_2O + 2H^+ \qquad (8)$$~~

~~The protons (H^+) produced by nitrification can be neutralized in two ways:~~

~~(i) either by exchange process with base cations in the soil exchange complex (Eq. (9))~~ ~~$$Soil—Ca + 2H^+ \longrightarrow Soil—2H^+ + Ca^{2+} \qquad (9)$$~~

~~(ii) or via carbonate mineral dissolution (Eq.(10))~~

~~$$Ca_{(1-x)}Mg_xCO_3 + H^+ \longrightarrow (1-x)\ Ca^{2+} + xMg^{2+} + HCO_3^- \qquad (10)$$~~

~~Consequently, after Eq. (8) and Eq. (10) are combined, carbonate weathering by protons produced by nitrification is supposed to becomes (Eq. 11) (See details in Perrin et al., 2008 and Gandois et al., 2011).~~

~~$$2Ca_{(1-x)}Mg_xCO_3 + NH_4^+ + 2O_2 \longrightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 2HCO_3^- \qquad (11)$$~~

~~The *Rcw* of limestone tablets and the initial concentration of NH_4^+ are plotted in~~

~~Fig. 4. A distinct relationship between them is observed: the A*cw* and R*cw* in NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea treatments are bigger than in control treatment, where the initial concentration of NH_4^+ displays similar results (Fig. 4). This suggests that carbonate weathering in NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea treatments are mainly attributed to the dissolution reaction described as Eq. (11). This process of carbonate weathering by protons from nitrification has been proven by many studies, from laboratory to field (Semhi and Suchet, 2000; Bertrand et al., 2007; Oh and Raymond, 2006; Errin et al., 2006; Hamilton et al., 2007; Biasi et al., 2008; Perrin et al., 2008; Barnes and Raymond, 2009; Chao et al., 2011; West and McBride, 2005; Gandois et al., 2011). According to the estimation from Yue et al. (2015), The enhanced HCO_3^- flux due to nitrification of NH_4^+ at Houzhai catchment of Guizhou province would be 3.72×10^5 kg C/year and account for 18.7% of this flux in the entire catchment(Yue et al., 2015). This is similar to estimates from other small agricultural carbonate basins (12-26%) in Southwest France (Semhi and Suchet, 2000; Perrin et al., 2008).~~

~~As discussed above, provided that the loss as ammonia (NH_3) and nitrous oxide (N_2O) after hydrolyzation is unconsidered in this study, the final equation of carbonate weathering in NH_4NO_3, NH_4HCO_3, NH_4Cl, $(NH_4)_2CO_3$ and urea treatments will be followed as, respectively:~~

~~$2Ca_{(1-x)}Mg_xCO_3 + NH_4NO_3 + 2O_2 \longrightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + 2NO_3^- + H_2O + 2HCO_3^-$ ———— (12)~~

~~$2Ca_{(1-x)}Mg_xCO_3 + NH_4HCO_3 + 2O_2 \longrightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 3HCO_3^-$ ———— (13)~~

~~$2Ca_{(1-x)}Mg_xCO_3 + NH_4Cl + 2O_2 \rightarrow 2(1-x)\ Ca^{2+} + 2xMg^{2+} + NO_3^- + Cl^- + H_2O + 2HCO_3^-$ (14)~~

~~$3Ca_{(1-x)}Mg_xCO_3 + (NH_4)_2CO_3 + 4O_2 \rightarrow 3(1-x)\ Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 2H_2O + 4HCO_3^-$ (15)~~

~~$3Ca_{(1-x)}Mg_xCO_3 + CO(NH_2)_2 + 4O_2 \rightarrow 3(1-x)\ Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 4HCO_3^-$ (16)~~

~~The A*cw* and R*cw* in $(NH_4)_3PO_4$ treatment, unlike in other NH_4-fertlizer treatments, had not a significant increase comparing with control treatment, which is not only owing to the low amount of added NH_4^+ in $(NH_4)_3PO_4$ treatment (0.3 mol, see Table 3) but also relative to the inhibition of phosphate. After the addition of $(NH_4)_3PO_4$ in soil, calcium orthophosphate (Ca-P) precipitation will form on calcite surface which is initiated with the aggregation of clusters leading to the nucleation and subsequent growth of Ca-P phases, at various pH values and ionic strengths relevant to soil solution conditions (Chien et al., 2011; Wang et al., 2012).~~

~~**4.4 Little/no effect of NO_3-fertilizer on carbonate weathering and its implication to the evaluation of CO_2 consumption by carbonate weathering**~~

~~In Fig. 3, the A*cw* and R*cw* without significant difference with control treatment in $NaNO_3$ treatment indicates that the addition of NO_3-fertilizer does not significantly influence carbonate weathering. This result is raising a new problem.~~

~~Eq. (5), usually as an expression for the natural weathering process of carbonate, is an important reaction for understanding the kinetics process of carbonate dissolution in carbonate-dominated areas, where the molar ratio of HCO_3^- and Me^{2+} in the river as an indicator is usually used to make estimations of CO_2 consumption by carbonate weathering at the regional/global scale (Hagedorn and Cartwright, 2009; Li et al., 2009). At agricultural areas, the relationship between $(Ca+Mg)/HCO_3^-$ and NO_3^-~~

~~is usually employed to estimate the contribution of N-fertilizer to riverine Ca^{2+}, Mg^{2+} and alkalinity (Etchanchu and Probst, 1988; Jiang, 2013; Jiang et al., 2009; Perrin et al., 2008; Semhi and Suchet, 2000). In these studies, the nitrification described as Eq. (8) is usually considered as the unique origin of NO_3^-. According to the result of $NaNO_3$ treatment in this study, the contribution of protons from nitrification to carbonate weathering may be overestimated if anthropogenic NO_3^- is neglected, since the anthropogenic NO_3^- does not release the proton described as Eq. (8). For NH_4NO_3 fertilizer, the (Eq. (12)) show that the two moles of Ca^{2+}+Mg^{2+}, NO_3^- and HCO_3^- will be produced when one mole NH_4NO_3 react with 2 moles of carbonate, where only half of NO_3^- originate from nitrification described as Eq. (8). This will result in doubled overestimation on the true contribution of the nitrification to CO_2 consumption by carbonate weathering.~~

~~At regional scales, If different fertilizers are added to an agricultural area, the estimation of CO_2 consumption by carbonate weathering might became more complicated, since the mole ratio of Ca+Mg, HCO_3^- and/or NO_3^- between different fertilization treatment is different (see Eq. (8)-(12)). Thus, the related anthropogenic inputs (e.g. Ca+Mg, NH_4, NO_3^-, HCO_3^-, etc.) need to be investigated to more accurately estimate the impact of fertilization on carbonate weathering and its CO_2 consumption. Moreover, the statement that nitrogenous fertilizer can aid carbonate weathering may result in misunderstanding more or less, it should not be nitrogenous fertilizer but, rather, ammonium fertilizer.~~

~~5. Conclusion~~

~~The impact of the addition of different fertilizer (NH_4NO_3, NH_4HCO_3, $NaNO_3$, NH_4Cl, $(NH_4)_2CO_3$, $Ca_3(PO_4)_2$, $(NH_4)_3PO_4$, Ca-Mg-P, Urea and K_2CO_3) on carbonate weathering was studied in a field column experiment with carbonate rock tablets at its~~

~~bottom of each. The weathering amount and ratio of carbonate rock tablets showed that the addition of urea, NH_4NO_3, NH_4HCO_3, NH_4Cl and $(NH_4)_2CO_3$ distinctly increased carbonate weathering, which was attributed to the nitrification of NH_4^+, and the addition of $Ca_3(PO_4)_2$, Ca-Mg-P and K_2CO_3 induced carbonate precipitation due to common ion effect. While the $(NH_4)_3PO_4$ and $NaNO_3$ addition did not impact significantly on carbonate weathering, where the former can be attributed to low added amount of $(NH4)_3PO_4$, may be related to the inhibition of phosphate, and the latter seemed to be raising a new question. The little impact of nitrate on carbonate weathering may result in the overestimation of impact of N-fertilizer on CO_2 consumption by carbonate weathering at the regional/global scale if the effect of NO_3 and NH_4 are not distinguished. Thus, the related anthropogenic inputs (e.g. Ca+ Mg, NH_4, NO_3^-, HCO_3^-, etc.) need to be investigated to more accurately estimate the impact of fertilization on carbonate weathering and its CO_2 consumption. Moreover, in order to avoid misunderstanding more or less, the statement that nitrogenous fertilizer can aid carbonate weathering should be replaced by ammonium fertilizer.~~

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Table 1 Chemical composition of soil

Parameter	Unit	Values
pH	-	6.94
Content of particles ~~(~~<0.01mm~~)~~	%	74
Content of particles ~~(~~<0.001mm~~)~~	%	45
Organic matter	%	0.99
NH_4^+-N	mg/kg	339.87
NO_3^--N	mg/kg	569.05
Available P	mg/kg	8.18
Available K	mg/kg	56.88
Available Ca	mg/kg	3041.06
Available Mg	mg/kg	564.83
Available S	mg/kg	100.72
Available Fe	mg/kg	24.41

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Table 2 Carbonate weathering under different fertilizer treatments

Treatment	Limestone		Dolostone	
	~~Acw~~ Rw / ‰~~g~~	Raw / $g\ m^{-2}\ a^{-1}$ ~~Rcw /‰~~	~~Acw~~ Rw / ‰~~g~~	Raw / $g\ m^{-2}\ a^{-1}$ ~~Rcw /‰~~
Control	0.~~0014~~48 ± 0.14~~a~~	2.00 ± 0.58~~a0.48~~	-0.~~0011~~31 ± 0.09~~a~~	-1.57 ± 0.86~~a-0.31~~
NH_4NO_3	~~0.0174~~6.42 ± 0.28~~c~~	24.86 ± 2.01~~b~~ ~~6.42~~	~~0.0144~~5.30 ± 0.87~~c~~	20.57 ± 1.15~~b5.30~~
NH_4HCO_3	~~0.0147~~4.44 ± 0.81~~b~~	21.00 ± 3.45~~b~~ ~~4.44~~	~~0.0096~~3.22 ± 0.87~~b~~	13.71 ± 3.88~~b3.22~~
$NaNO_3$	~~0.0031~~0.86 ± 0.17~~a~~	4.43 ± 1.73~~a0.86~~	0.~~0022~~53 ± 0.26~~a~~	3.14 ± 1.73~~a0.53~~
NH_4Cl	~~0.0149~~5.54 ± 0.64~~bc~~	21.29 ± 2.45~~b~~ ~~5.54~~	~~0.0131~~4.77 ± 0.78~~bc~~	18.71 ± 0.86~~b~~ ~~4.77~~
$(NH_4)_2CO_3$	~~0.0144~~4.48 ± 0.95~~bc~~	20.57 ± 4.46~~b~~ ~~4.84~~	~~0.0186~~4.94 ± 1.91~~bc~~	26.57 ± 7.62~~b4.94~~
$Ca_3(PO_4)_2$	~~0.0003~~0.01 ± 0.04~~a~~	0.43 ± 0.86~~a0.01~~	-0.~~0013~~55 ± 0.25~~a~~	-1.86 ± 1.29~~a-0.55~~
$(NH_4)_3PO_4$	~~0.0028~~1.08 ± 0.34~~a~~	4.00 ± 1.15~~a1.08~~	~~0.0007~~0.75 ± 0.21~~a~~	1.00 ± 1.01~~a0.75~~
Ca-Mg-P	~~-0.0013~~-0.31 ± 0.12~~a~~	-1.86 ± 0.43~~a-0.31~~	-0.~~0022~~97 ± 0.38~~a~~	-3.14 ± 0.72~~a-0.97~~
Urea	~~0.0243~~8.48 ± 0.96~~d~~	34.71 ± 4.32~~c~~ ~~8.48~~	~~0.0185~~6.59 ± 0.67~~d~~	26.43 ± 2.73~~c6.59~~
K_2CO_3	-0.~~0008~~26 ± 0.15~~a~~	-1.14 ± 0.58~~a-0.26~~	-0.~~0018~~59 ± 0.15~~a~~	-2.57 ± 0.43~~a-0.59~~

~~Acw - the amount of carbonate weathering;~~ R~~c~~w - the ratio of carbonate weathering; Ra~~c~~w - the rate of carbonate weathering; ~~Acw~~ Rw =1000 (Wi-Wf)/Wi~~;~~ and ~~Rcw = (Wi-Wf)/Wi,~~ Ra~~c~~w = (Wi-Wf)/(S*T), where Wi is the initial weight of the carbonate rock tablet~~s~~, and Wf is the~~ir~~ final weight. S is the surface area of carbonate ~~weathering~~ rock tablet~~s~~ (~~In this study~~here, we used ~~a same~~ S = 7 cm^2 for every tablet~~s~~), and T is the experiment period. Values are reported as means ± standard deviations, n = 3. ~~Values in each column followed by different letters are significantly (p <0.05) different based on one-way ANOVA.~~

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~~Table 3: The main reaction and effects in fertilized treatments, and the potential nitrogenous transformation (The amount of generated NH_4^+ NO_3^-) at the initial phase of the experiment~~

~~Treatment~~	~~Main reactions and effects~~
~~1.Control~~	~~$Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O \rightarrow (1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^-$~~
~~2.NH_4NO_3~~	~~$2Ca_{(1-x)}Mg_xCO_3 + NH_4NO_3 + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + 2NO_3^- + H_2O + 2HCO_3^-$~~
~~3. H_4HCO_3~~	~~$2Ca_{(1-x)}Mg_xCO_3 + NH_4HCO_3 + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 3HCO_3^-$~~
~~4. $NaNO_3$~~	~~$Ca_{(1-x)}Mg_xCO_3 + NaNO_3 + CO_2 + H_2O \rightarrow (1-x) Ca^{2+} + xMg^{2+} + Na^+ + NO_3^- + 2HCO_3^-$~~
~~5. NH_4Cl~~	~~$2Ca_{(1-x)}Mg_xCO_3 + NH_4Cl + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_3^- + Cl^- + H_2O + 2HCO_3^-$~~
~~6. $(NH_4)_2CO_3$~~	~~$3Ca_{(1-x)}Mg_xCO_3 + (NH_4)_2CO_3 + 4O_2 \rightarrow 3(1-x) Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 2H_2O + 4HCO_3^-$~~
~~7. $Ca_3(PO_4)_2$~~	~~(1) Common ion effect: The $Ca_{(1-x)}Mg_xCO_3$ produces when the concentrations of Ca^{2+} and Mg^{2+} increases~~ ~~$(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$~~ ~~(2) Inhibition of phosphate to calcite precipitation: calcium phosphate precipitation produces on the surface of calcite after the addition of PO_4^{3-} in soil, resulting in inhibiting the precipitation of calcite~~
~~8. $(NH_4)_3PO_4$~~	~~(1) $2Ca_{(1-x)}Mg_xCO_3 + NH_4^+ + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 2HCO_3^-$~~ ~~(2) Inhibition of phosphate to calcite dissolution: calcium orthophosphate (Ca-P) precipitation produces on surface of calcite after the addition of PO_4^{3-} in soil, resulting in inhibiting the dissolution of calcite~~
~~9. Ca-Mg-P~~	~~(1) Common ion effect: The $Ca_{(1-x)}Mg_xCO_3$ produces when the concentrations of Ca^{2+} and Mg^{2+} increases~~ ~~$(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$~~ ~~(2) Inhibition of phosphate to calcite precipitation: calcium phosphate precipitation produces on the surface calcite after the addition of P in soil, resulting in inhibiting the precipitation of calcite~~
~~10. Urea~~	~~$3Ca_{(1-x)}Mg_xCO_3 + CO(NH_2)_2 + 4O_2 \rightarrow 3(1-x) Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 4HCO_3^-$~~
~~11. K_2CO_3~~	~~Common ion effect: The $Ca_{(1-x)}Mg_xCO_3$ produces when the concentration of HCO_3^- increases~~ ~~(i) $(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$~~ ~~(ii) $K_2CO_3 + H_2O \rightarrow 2K^+ + HCO_3^- + OH^-$~~

Table 3~~:~~ –The main reaction and effects in the~~se~~ 11 fertiliz~~ed~~er treatments

Treatment	Main reactions and effects
1. Control	$Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O \rightarrow (1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^-$
2. NH_4NO_3	$2Ca_{(1-x)}Mg_xCO_3 + NH_4NO_3 + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + 2NO_3^- + H_2O + 2HCO_3^-$
3. NH_4HCO_3	$NH_4HCO_3 \rightarrow NH_3\uparrow + H_2O + CO_2\uparrow$ $2Ca_{(1-x)}Mg_xCO_3 + NH_4HCO_3 + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 3HCO_3^-$
4. $NaNO_3$	$Ca_{(1-x)}Mg_xCO_3 + NaNO_3 + CO_2 + H_2O \rightarrow (1-x) Ca^{2+} + xMg^{2+} + Na^+ + NO_3^- + 2HCO_3^-$
5. NH_4Cl	$2Ca_{(1-x)}Mg_xCO_3 + NH_4Cl + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_3^- + Cl^- + H_2O + 2HCO_3^-$
6. $(NH_4)_2CO_3$	$(NH_4)_2CO_3 \rightarrow 2NH_3\uparrow + H_2O + CO_2\uparrow$ $3Ca_{(1-x)}Mg_xCO_3 + (NH_4)_2CO_3 + 4O_2 \rightarrow 3(1-x) Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 2H_2O + 4HCO_3^-$
7. $Ca_3(PO_4)_2$	(1) $(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$ (2) $Ca + PO_4 \rightarrow Ca\text{-}P$
8. $(NH_4)_3PO_4$	(1) $2Ca_{(1-x)}Mg_xCO_3 + NH_4^+ + 2O_2 \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_3^- + H_2O + 2HCO_3^-$ (2) $Ca + PO_4 \rightarrow Ca\text{-}P$
9. Ca-Mg-P	(1) $(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$ (2) $Ca + PO_4 \rightarrow Ca\text{-}P$
10. Urea	$3Ca_{(1-x)}Mg_xCO_3 + CO(NH_2)_2 + 4O_2 \rightarrow 3(1-x) Ca^{2+} + 3xMg^{2+} + 2NO_3^- + 4HCO_3^-$
11. K_2CO_3	(i) $(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$

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(ii) $K_2CO_3 + H_2O \rightarrow 2K^+ + HCO_3^- + OH^-$

Note: (1) Common ion effect: The $Ca_{(1-x)}Mg_xCO_3$ produced~~s~~ when the concentrations of Ca^{2+}, ~~–~~Mg^{2+} and/or HCO_3^- increases (for ~~T~~treatment 7, 9 and 11): $(1-x)\ Ca^{2+} + xMg^{2+} + 2HCO_3^- \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$; (2) Inhibition of ~~phosphate to~~ calcite dissolution/precipitation by phosphate: calcium orthophosphate (Ca-P) precipitation produced~~s~~ on the surface of calcite after the addition of PO_4^{3-} in soil, resulting in the inhibit~~ing~~ion of the dissolution/precipitation of calcite (for ~~T~~treatment 7, 8 and 9): $Ca + PO_4 \rightarrow Ca\text{-}P$

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Table 4~~:~~ –The amount of fertilizer-derived NH_4^+ at the initial phase of the experiment and the potential nitrogenous transformation (NH_4^+-NO_3^-)

Treatment	Molecular mass g/mol	Amount of added fertilizer /g	Molar amount /mole	Amount of fertilizer-derived NH_4^+ /mole	The maximum of N products /mole
NH_4NO_3	80	43	0.54	0.54	1.08
NH_4HCO_3	79	85	1.08	1.08	1.08
$NaNO_3$	85	91	1.07	0.00	1.07
NH_4Cl	53.5	57	1.07	1.07	1.07
$(NH_4)_2CO_3$	96	51	0.53	1.06	1.06
$Ca_3(PO_4)_2$	310	52	0.17	0.00	0.00
$(NH_4)_3PO_4$	149	15	0.10	0.30	0.30
Ca-Mg-P	~~w~~nd	44	~~w~~nd	0.00	0.00
Urea	60	32	0.53	1.06	1.06
K_2CO_3	138	10	0.07	0.00	0.00

~~w~~nd=~~without~~no data~~;~~. The amount of added fertilizer (g) divided by its molecular mass (g/mol) ~~wa~~is the molar amount of fertilizer (mole)~~;~~. ~~Gadf- gram amount of added fertilizers (g); Maafof added fertilizers (mol).~~The amounts of fertilizer-derived NH_4^+ is calculated by their own ionization or hydrolysis processes. The maximum of N products is estimated by their main reactions in ~~t~~Table 3.

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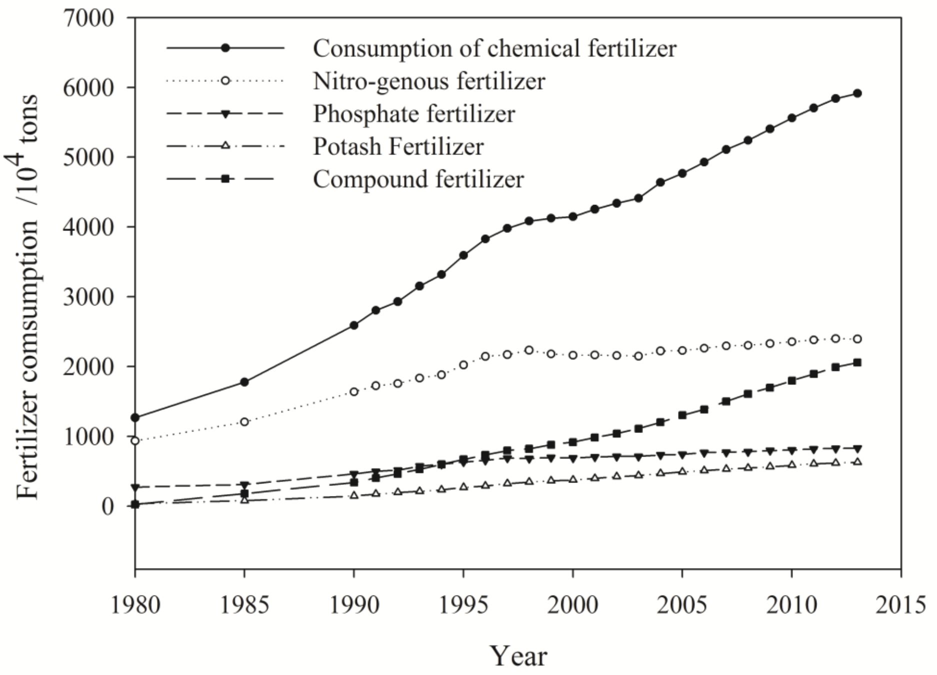


Fig. 1 The change ~~of~~ in chemical fertilizer consumption in China during the 1980-2013 period

The data were collected from National Bureau of Statistics of the People's Republic of China (NBS, 2014)~~(NBS, 2014)(NBS, 2014)~~ (http://www.stats.gov.cn/tjsj/ndsj/)

域代码已更改

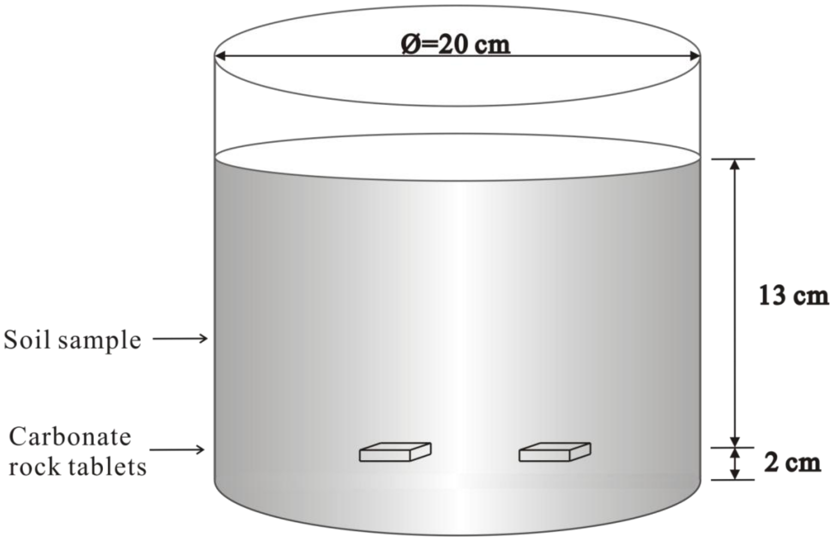
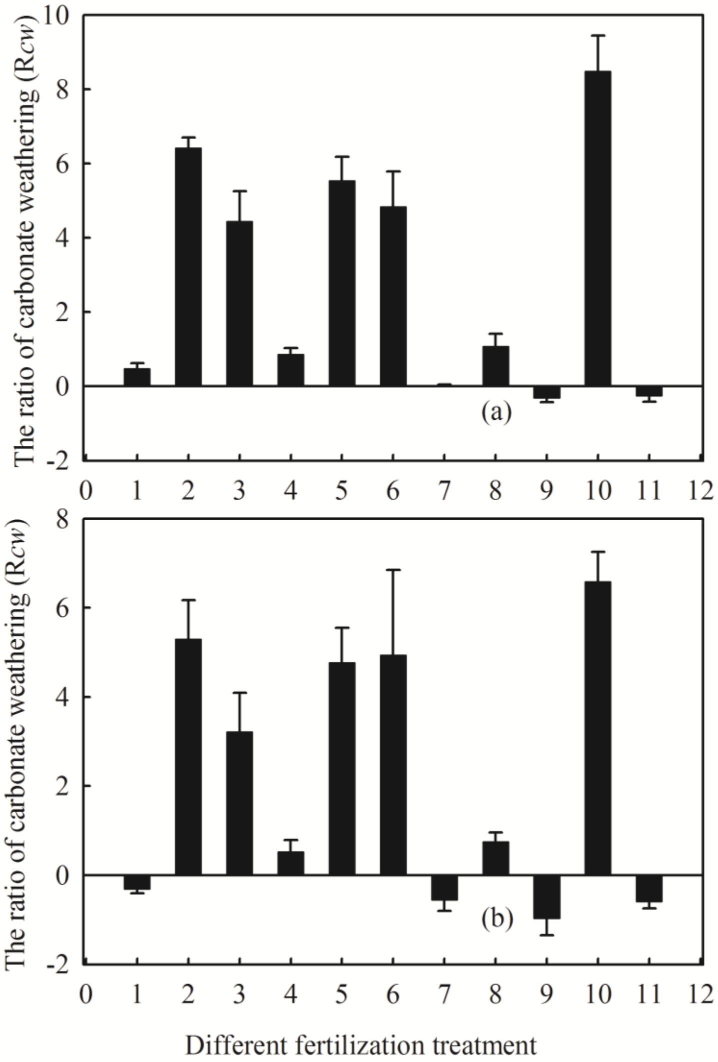
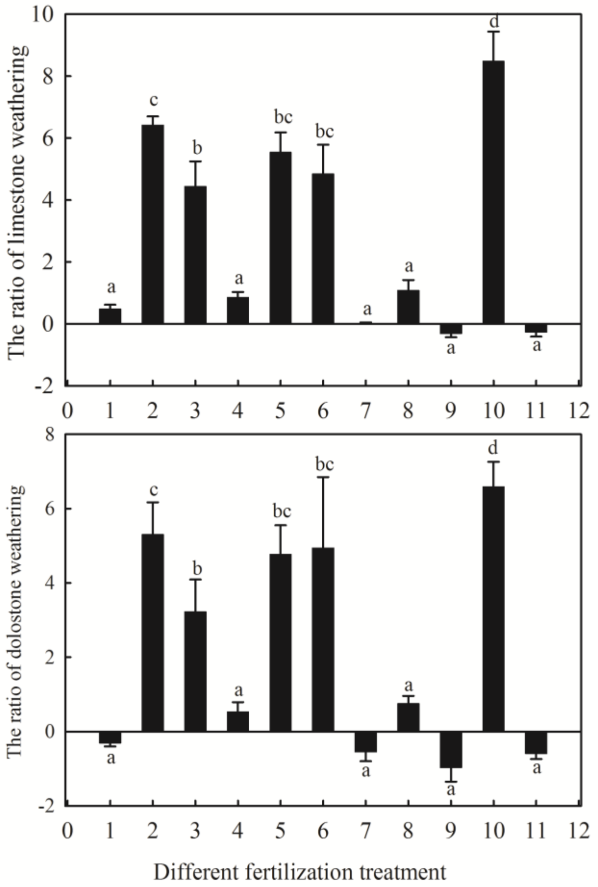


Fig. 2 Sketch ~~map~~ of the soil column with rock tablets

1198 

The ratio of carbonate weathering (R*cw*)
10
8
6
4
2
0
-2
(a)
0 1 2 3 4 5 6 7 8 9 10 11 12
The ratio of carbonate weathering (R*cw*)
8
6
4
2
0
-2
(b)
0 1 2 3 4 5 6 7 8 9 10 11 12
Different fertilization treatment

1199 
The ratio of limestone weathering
10
8
6
4
2
0
-2
0 1 2 3 4 5 6 7 8 9 10 11 12
a
c
b
a
bc
bc
a
a
a
d
a
The ratio of dolostone weathering
8
6
4
2
0
-2
0 1 2 3 4 5 6 7 8 9 10 11 12
a
c
b
a
bc
bc
a
a
a
d
a
Different fertilization treatment

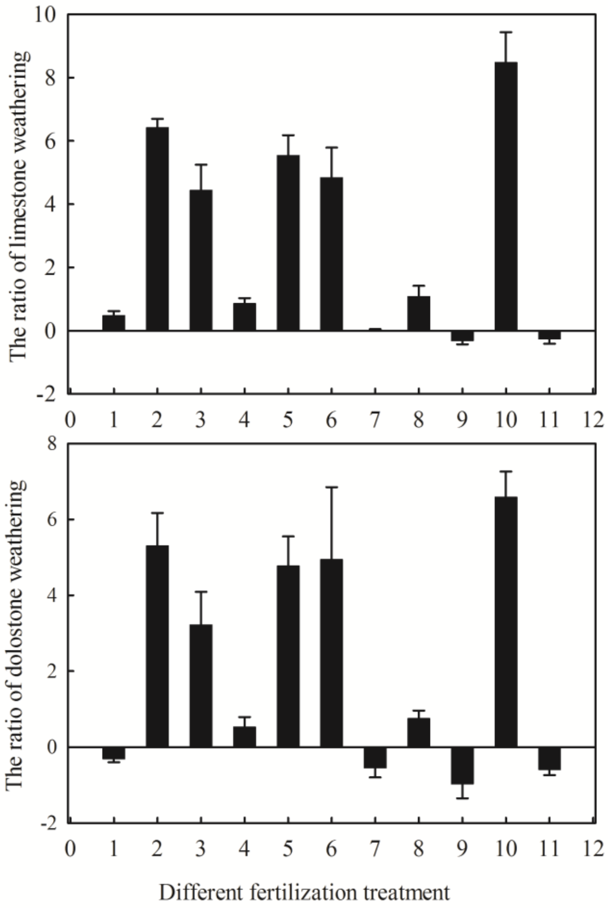


Fig. 3 The ~~ratio of carbonate weathering~~R*w* (‰) of limestone and dolostone –under different ~~fertilization~~ fertilizer treatments

~~(a) limestone; (b) dolostone.~~ Treatment 1~~-~~. Control; 2~~-~~. NH_4NO_3; 3~~-~~. NH_4HCO_3; 4~~-~~. $NaNO_3$; 5~~-~~. NH_4Cl; 6~~-(~~. $(NH_4)_2CO_3$; 7~~-~~. $Ca_3(PO_4)_2$; 8~~-(~~. $(NH_4)_3PO_4$; 9~~-~~. Ca-Mg-P; 10~~-~~. Urea; 11~~-~~. K_2CO_3. R~~c~~*w* = 1000(W*i*-W*f*)/W*i*, where W*i* is the initial weight of the carbonate rock tablet~~s~~, and W*f* is the~~ir~~ final weight. ~~Values in each column followed by different letters are significantly (p <0.05) different based on one-way ANOVA.~~

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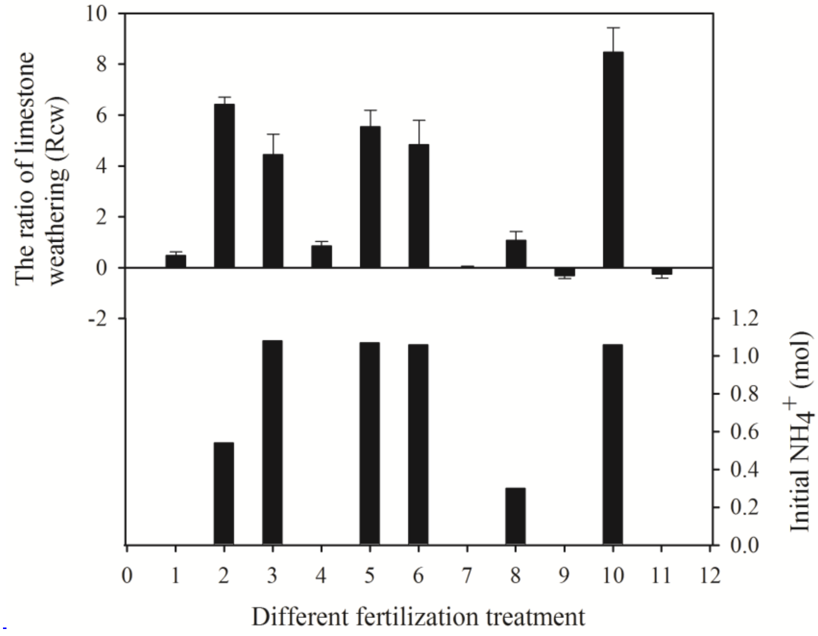

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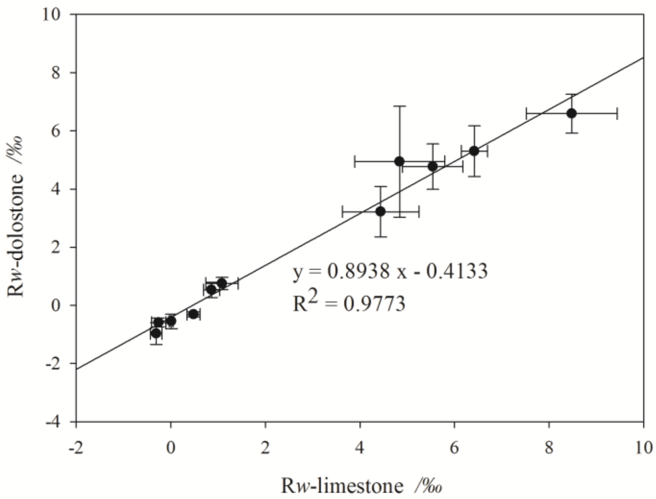
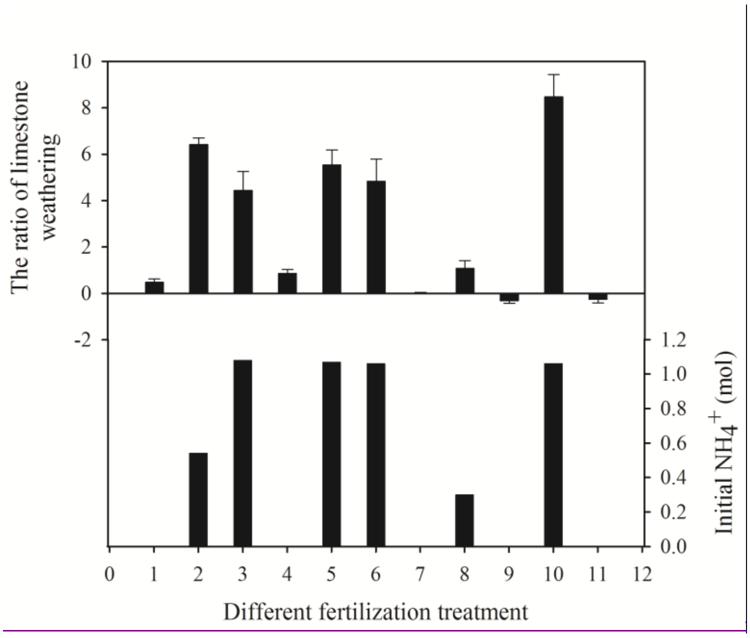


Fig. 4 The linear correlation of R*w* (‰) of limestone and dolostone

R*w* = 1000(W*i*-W*f*)/W*i*, where W*i* is the initial weight of the lim~~s~~estone tablet~~s~~, and W*f* is the~~ir~~ final weight.



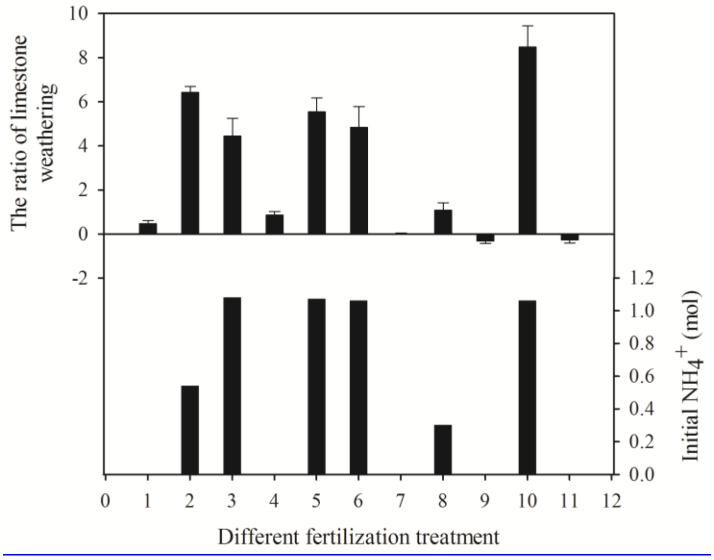


Fig. ~~4~~ 5 The R*w* (‰) of limestone ~~ratio of limestone weathering~~ and the molar amount of produced NH_4^+ under different ~~fertilization~~ fertilizer treatments

Treatment 1~~-~~. Control; 2~~-~~. NH_4NO_3; 3~~-~~. NH_4HCO_3; 4~~-~~. $NaNO_3$; 5~~-~~. NH_4Cl; 6~~-(~~. $(NH_4)_2CO_3$; 7~~-~~. $Ca_3(PO_4)_2$; 8~~-(~~. $(NH_4)_3PO_4$; 9~~-~~. Ca-Mg-P; 10~~-~~. Urea; 11~~-~~. K_2CO_3. R~~e~~*w* =1000(W*i*-W*f*)/W*i*, where W*i* is the initial weight of the lim~~s~~estone tablet~~s~~, and W*f* is the~~ir~~ final weight.

校对报告

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~~所有题录的数据正常~~