1	Impact of different fertilizers on the carbonate weathering in a typical karst area; Southwest China: a field column experiment	 带格式的:居中	
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15			

17	Abstract: Carbonate weathering, as a significant vector for the movement of carbon
18	both between and within ecosystems, are-is_strongly influenced by anthropogenic
19	perturbations such as agricultural fertilization, since the addition of fertilizers tends
20	to change the chemical characteristics of soil such as the pH-value. Different
21	fertilizers may exert a different impact on carbonate weathering, but their these
22	discrepanciesdifferences are as of yet not still-well-known-so far. In this study, a field
23	column experiment was employed conducted to explore the responses of carbonate
24	weathering to the addition of different fertilizers addition. The eleven different
25	treatments with three replicates including control, NH4NO3, NH4HCO3, NaNO3,
26	NH ₄ Cl, (NH ₄) ₂ CO ₃ , Ca ₃ (PO ₄) ₂ , (NH ₄) ₃ PO ₄ , fused calcium magnesium phosphate
27	fertilizer (Ca Mg P), Urea and K_2CO_3 were established in this column experiment,
28	where limestone and dolostone tablets were buried at the bottom of each to determine
29	the weathering amount and ratio of carbonate in soil. We compared 11 different
30	treatments, including a control treatment, using 3 replicates per treatment. Carbonate
31	weathering was assessed by measuring the weight loss of limestone and dolostone
32	tablets buried at the bottom of thesoil-filled columns. The results showed that the
33	addition of urea, NH4NO3, NH4HCO3, NH4Cl and (NH4)2CO3 distinctly increased
34	carbonate weathering, which was attributed to the nitrification of NH_4^+ , and t The
35	addition of Ca ₃ (PO ₄) ₂ , Ca-Mg-P and K ₂ CO ₃ induced carbonate precipitation due to
36	the common ion effect. Whereas the The addition of (NH ₄) ₃ PO ₄ and NaNO ₃ addition
37	did not <u>significantly</u> impact significantly on carbonate weathering. The results of

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38	NaNO ₃ treatment seem to be raisingraise a new question: the <u>negligible</u> little impact of	
39	nitrate on carbonate weathering may result in the overestimation of the impact of	
40	N-fertilizer on CO_2 consumption by carbonate weathering at the regional/global scale.	
41	if the effects of NO ₃ and NH ₄ are not distinguished. Moreover, in order to avoid	
42	misunderstanding more or less, the statement that nitrogenous fertilizer can aid	
43	carbonate weathering should be replaced by ammonium fertilizer.	
44	Keywords: Carbonate weathering; Column experiment; Nitrogenous fertilizer;	
45	Phosphate fertilizer; Southwest China	
46		
47		
48	1. Introduction	
49	Carbonate weathering plays a significant role in consumption of the elevated	
49 50	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.,	
50	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al.,</u>	
50 51	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al., <u>2000; Liu et al., 2010; 2011)</u>(Kump et al., 2000; Liu et al., 2010; 2011). The rRiverine</u>	
50 51 52	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al.,</u> <u>2000; Liu et al., 2010; 2011)</u> (Kump et al., 2000; Liu et al., 2010; 2011). The rR iverine hydro-chemical composition, such as the ratio of HCO_3^- and to $Ca^{2+} + Mg^{2+}$, is	
50 51 52 53	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al.,</u> <u>2000; Liu et al., 2010; 2011)</u> (Kump et al., 2000; Liu et al., 2010; 2011). The rRiverine 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 <u>natural</u>	
50 51 52 53 54	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al.,</u> <u>2000; Liu et al., 2010; 2011)</u> (Kump et al., 2000; Liu et al., 2010; 2011). The rR iverine 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 <u>natural</u> carbonate weathering at the regional/global scale <u>(Hagedorn and Cartwright, 2009; Li</u>	
50 51 52 53 54 55	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al.,</u> <u>2000; Liu et al., 2010; 2011)</u> (Kump et al., 2000; Liu et al., 2010; 2011). The rRiverine 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 <u>natural</u> carbonate weathering at the regional/global scale <u>(Hagedorn and Cartwright, 2009; Li</u> <u>et al., 2009)(Hagedorn and Cartwright, 2009; Li et al., 2009)(Hagedorn and</u>	
50 51 52 53 54 55 56	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al., 2000; Liu et al., 2010; 2011)</u> . The rR iverine 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 <u>natural</u> carbonate weathering at the regional/global scale <u>(Hagedorn and Cartwright, 2009; Li et al., 2009)(Hagedorn and Cartwright, 2009; Li et al., 2009). However, fluvial alkalinity may also be produced</u>	
50 51 52 53 54 55 56 57	atmospheric CO ₂ <u>(Kump et al., 2000; Liu et al., 2010; Liu et al., 2011)(Kump et al.,</u> <u>2000; Liu et al., 2010; 2011)(Kump et al., 2000; Liu et al., 2010; 2011). The rR</u> iverine 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 <u>natural</u> carbonate weathering at the regional/global scale <u>(Hagedorn and Cartwright, 2009; Li</u> <u>et al., 2009)(Hagedorn and Cartwright, 2009; Li et al., 2009)(Hagedorn and Cartwright, 2009; Li et al., 2009)</u> . However, <u>fluvial alkalinity may also be produced</u> by other processes including the reaction between carbonates and the protons derived	

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60	produced due to the reaction between carbonate and the protons which can originate	
61	from the nitrification processes of N-fertilizer (Barnes and Raymond, 2009; Chao et	域代码已更改
62	al., 2011; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin	
63	et al., 2008; Pierson-wWickmann et al., 2009; Semhi and Suchet, 2000; West and	
64	McBride, 2005)(Barnes and Raymond, 2009; Chao et al., 2011; Gandois et al., 2011;	
65	Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson-wickmann	
66	et al., 2009; Semhi and Suchet, 2000; West and McBride, 2005)(Barnes and Raymond,	
67	2009; Chao et al., 2011; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond,	
68	2006; Perrin et al., 2008; Pierson-wickmann et al., 2009; Semhi and Suchet, 2000;	
69	West and McBride, 2005),; (ii) from the sulfuric acid (Lerman and Wu, 2006; Lerman	带格式的: 字体颜色: 文字 1 域代码已更改
70	et al., 2007; Li et al., 2008; Li et al., 2009)(Lerman and Wu, 2006; Lerman et al.,	
71	2007; Li et al., 2008; Li et al., 2009)(Lerman and Wu, 2006; Lerman et al., 2007; Li et	
72	al., 2008; Li et al., 2009); (iii)-from organic acid secreted by microorganisms (Lian et	域代码已更改
73	al., 2008)(Lian et al., 2008)(Lian et al., 2008);, as well as and (iv) from acidic soil	
74	<u>(Chao et al., 2014; Chao et al., 2017)(Chao et al., 2014)</u> (Chao et al., 2014)Given	域代码已更改
75	the that atmospheric CO ₂ is not the <u>a</u> unique weathering agent, differentiating the	
76	agent of carbonate weathering is more and more significant to enable important for the	
77	accurate budgeting of the net CO_2 consumption by carbonate weathering, especially in	
78	agricultural <u>areas where mineral fertilizers are used</u> area.	带格式的: 字体:(默认) Times New Roman, 小四
79	The world-global average annual increase in mineral fertilizer consumption was	
80	3.3_% from 1961 to 1997, and FAO's study predicts a 1_% increase per year until	
81	2030 (FAO, 2000). For-In_China, the consumption of chemical fertilizer increased	

82	from 12.7 Mt in 1980 to 59.1 Mt in 2013 (Fig. 1). The Increasing-increasing
83	consumption of chemical_mineral_fertilizers is a significant disturbance factor of in
84	carbonate weathering and the carbon cycle. Many Several studies have showed shown
85	that nitrogen fertilizer additions aided increased in the dissolution of lime weathering
86	rates, and also increased the total export of DIC from agricultural watersheds (Barnes
87	and Raymond, 2009; Gandois et al., 2011; Hamilton et al., 2007; Oh and Raymond,
88	2006; Perrin et al., 2008; Pierson-wWickmann et al., 2009; Probst, 1986; Semhi and
89	Suchet, 2000; West and McBride, 2005)(Barnes and Raymond, 2009; Gandois et al.,
90	2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008;
91	Pierson wickmann et al., 2009; Probst, 1986; Semhi and Suchet, 2000; West and
92	McBride, 2005)(Barnes and Raymond, 2009; Gandois et al., 2011; Hamilton et al.,
93	2007; Oh and Raymond, 2006; Perrin et al., 2008; Pierson wickmann et al., 2009;
94	Probst, 1986; Semhi and Suchet, 2000; West and McBride, 2005). According to the
95	estimation from estimates by Probst (1988) and Semhi et al. (2000), the contribution of
96	N-fertilizers to carbonate dissolution represents-was_30_% and 12-26_%, respectively,
97	onin two small agricultural carbonate basins in south-western France, the Girou and
98	the Gers, respectively (subtributary and tributaryies of the Garonne riverRiver,
99	respectively). For-In lager basin level, such as the Garonne river-River basinBasin,
100	which is a larger basin (52,000 km ²), this contribution was estimated at 6 % by Semhi
101	et al. (2000). At national and global scales, Perrin et al. (2008) estimated that the
102	deficit-contribution of CO2-uptake due to N-fertilizer addition (usually in form of
103	NH ₄ NO ₃) represent <u>s</u> up to 5.7-13.4_% and only -1.6-3.8_% of the total CO₂ flux

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naturally consumed bytoof the carbonate dissolution, forin France and across the on a

105 gglobal-scale, respectively.

106 These estimated results - estimates described above wereare usually largely based 107 on calculations that assumeding that a single type of fertilizer (e.g. $(NH_4)_2SO_4$, 108 109 NH₄NO₃, or NH₄Cl) was used throughout the whole basin that was considered. 110 Howeverwere usually based on a hypothesis of individual fertilizer (e.g. (NH4)2SO47 NH4NO3, or NH4Cl) input into an agricultural basin. Nevertheless, in actual 111 112 agricultural practices, at an agricultural basin, different fertilizers are usually added for different crops-in actual agricultural practices. The impact of these fertilizers on 113 114 carbonate weathering and riverine chemical composition may be different. For-In the 115 case of nitrogenous fertilizer, 100_% NO₃⁻ produced after the addition (NH₄)₂SO₄ and NH₄Cl<u>is</u> derive<u>d</u> from the nitrification of NH₄⁺, <u>whilst</u> comparatively, <u>it is</u> only 50_% 116 after the addition of NH4NO3. The dDifferences of in NO3 sources may cause 117 118 the produce an evaluated deviation of $\frac{1}{1000}$ in the impact of N-fertilizer addition on CO₂ consumption by carbonate weathering. Because, since the addition of different 119 120 N-fertilizers (e.g. (NH₄)₂SO₄, NH₄NO₃, NH₄Cl, NaNO₃ or urea) may result in different contributions to carbonate weathering and relative products such as HCO₃, 121 Ca^{2+} and Mg^{2+} . For phosphate fertilizer, the coprecipitation of phosphate ions with 122 123 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 124 weathering to the addition of different fertilizers, such as N-fertilizer (NH₄ and NO₃), 125 126 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 127 agricultural force-impact on natural carbonate weathering, and to accurately evaluate 128

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129 the CO_2 consumption via carbonate weathering in agricultural areas.

125	the CO ₂ consumption via carbonate weathering in agricultural area <u>s</u> .	
130	Moreover, <u>TtThe carbonate-rock-tablet test is used to determine the weathering</u>	
131	rate of carbonate rock/mineral from the laboratory to the field (Chao et al., 2011;	域代码已更改
132	Chao et al., 2014; Dreybrodt et al., 1996; Gams, 1981; Gams, 1985; Jiang and Yuan,	
133	<u>1999; Liu and Dreybrodt, 1997; Plan, 2005; Trudgill, 1975)(Chao et al., 2014; Chao</u>	
134	et al., 2011; Dreybrodt et al., 1996; Gams, 1981; Gams, 1985; Jiang and Yuan, 1999;	
135	Liu and Dreybrod, 1997; Plan, 2005; Trudgill, 1975)(Gams, 1981; Chao et al., 2011;	
136	Trudgill, 1975; Chao et al., 2014; Dreybrodt et al., 1996; Gams, 1985; Jiang and Yuan,	
137	<u>1999; Liu and Dreybrod, 1997; Plan, 2005). In the laboratory, the</u>	
138	carbonate-rock-tablet is employed to study the kinetics of calcite	
139	dissolution/precipitation (Dreybrodt et al., 1996; Liu and Dreybrodt, 1997)(Dreybrodt	域代码已更改
140	et al., 1996; Liu and Dreybrod, 1997)(Dreybrodt et al., 1996; Liu and Dreybrod, 1997)	
141	and determine the rate of carbonate mineral weathering in the soil column (Chao et al.,	域代码已更改
142	2011)(Chao et al., 2011)(Chao et al., 2011). However, in the field, it is also used to	
143	observe the rate of carbonate weathering and estimated CO ₂ consumption-by	
144	carbonate weathering (Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013;	域代码已更改
145	Plan, 2005)(Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013; Plan,	
146	2005)(Chao et al., 2014; Jiang and Yuan, 1999; Jiang et al., 2013; Plan, 2005).	
147	Although-Liu (2011) argued that the carbonate-rock-tablet test may lead to-the	
148	deviations of in estimated CO ₂ consumption by carbonate weathering at the	
149	regional/global scale, in the cases of where there are insufficient representative data	
150	<u>(Liu, 2011)(Liu, 2011)(Liu, 2011), our results show that yet iI is nonetheless</u>	域代码已更改
151	athe preferred optionmethod for the condition controlled contrast comparative or	
152	stimulated experiment (Chao et al., 2011; Chao et al., 2014; Chao et al., 2017) (Chao	域代码已更改
153	et al., 2014; Chao et al., 2011)(Chao et al., 2011; Chao et al., 2014), Where the result	

154	from the carbonate-rock-tablet test is consistent to the major element geochemical
155	data of leachates from soil column(Chao et al., 2011).
156	Thus Therefore, in order to observe their difference between the impacts of
157	different fertilizer addition on carbonate weathering in soil, a A field column
158	experiment that involved embedding carbonate-rock-tablets with eleven different
159	treatments-was carried out in a typical karst area of southwest China, in order to
160	observe the impacts of different fertilizer additions on carbonate weathering in soil.
161	2. Materials and Methods
162	2.1 The study site
163	This study was carried out in a typical karst area, <u>namely the HuaXi-Huaxi</u>
164	district-District of Guiyang cityCity, Guizhou provinceProvince, SW China (26°23'N,
165	106°40'E, 1094 m aslASL). Guiyang, the capital city of Guizhou Province, is located
166	in the central part of The-the Province province, covering an area from 26°11'00" to

elevations ranging from 875 to 1655 m above mean sea levelASL. 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 °C 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

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to produce the vegetables and foods in the suburbs of Guiyang (Liu et al., 2006)(Liu

et al., 2006)(Liu et al., 2006). The consumption of chemical fertilizer increased from 0.8-150 kg/ha Mt-in 1980 to 1.0190 kg/ha Mt in 2013 (GBS, 2014)(GBS, 2014)(GB 178

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2.2 Soil properties 180

2014).

The soil used in this column experiment was sampled from the B horizon (below 181 20 cm in depth) of yellow-brown soil infrom dug from a cabbage-corn or 182 capsicum-corn rotation plantation in Huaxi dDistrict, HThe soil was air-dried, 183 184 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 185 186 characteristics of the soil, in-cluding organic matter (OM), NH₄-N, NO₃-N, available P, available K, available Ca, available Mg, available S and available Fe, and available 187 S were determined according to the Agro Services International (ASI) Method 188 (Hunter, 1980)(Hunter, 1980)(Hunter, 1980), OM was determined using an where the 189 extracting solution used for O.M. containeding 0.2 mol l^{-1} NaOH, 0.01 mol l^{-1} EDTA, 190 2 % methanol, and 0.005 % Superfloc 127-, NH₄-N, NO₃-N, available Ca, and Mg 191 were determined based using on an extraction extracting solution of by $-1 \mod 1^{-1} \text{ KCl}$ 192 solution, whereas available K, P and Fe were determined using an extracted by 193 extracting solution containing 0.25 mol l⁻¹ NaHCO₃, 0.01 mol l⁻¹ EDTA, 0.01 mol l⁻¹ 194 NH₄F, and 0.005_% Superfloc 127-, Finally, and available S was determined using an 195 extracting solution of ed by 0.1 mol 1^{-1} Ca(H₂PO₄)₂ and 0.005 % Superfloc 127. The 196 results are shown in Table 1. 197

2.3 Soil column and different fertilization treatments 198

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199	In order to test the hypothesis that the responses of the impact of different	
200	chemical fertilizers on carbonate weathering may be different, columns ($\emptyset_{=}20$ cm,	
201	H= 15_cm) were constructed from 20cm diameter polyvinylchloride (PVC) pipe (Fig.	
202	2). A hole ($\emptyset_{=2}$ cm) were was established placed at the bottom of each column to	
203	discharge soil water from the of-soil column. A polyethylene net mesh (Ø 0.5 mm)	
204	was placed in the bottom of the columns to prevent the soil loss of the filter material.	
205	A 2 cm thick filter-sand layer, with 2 cm thickness-including gravel, coarse sand and	
206	fine sand, was spread onover the net Two different carbonate rock tablets were	
207	buried in the bottom of each soil column (Fig2). According to Based on the common	
208	kinds of chemical fertilizers and the main objective of this study, eleven fertilization	
209	treatments, each with three replicates, were set up in the field column experiment.	
210	<u>There are-were set up</u> : (1)_control without fertilizer (CK); (2)_43g NH ₄ NO ₃ fertilizer	
211	(CF); (3)_85g NH ₄ HCO ₃ fertilizer (NHC); (4)_91g NaNO ₃ fertilizer (NN); (5)_57g	
212	NH ₄ Cl fertilizer (NCL); (6)_51g (NH ₄) ₂ CO ₃ fertilizer (NC); (7)_52g Ca ₃ (PO ₄) ₂	
213	fertilizer (CP); (8)_15g (NH ₄) ₃ PO ₄ fertilizer (NP); (9)_44g fused calcium-magnesium	
214	phosphate fertilizer (Ca-Mg-P); (10) 32g Urea fertilizer (U); and (11) 10g K ₂ CO ₃	
215	fertilizer (PP). To shorten the experiment time and enhance the effect of fertilization,	
216	the added amount of fertilizers in these treatments motioned above was increased to	
217	30 times than its local practical amount <u>(N fertilizer: 160 kg N-ha</u>⁻¹; P fertilizer: 50 kg	/
218	P-ha ⁻¹ ; K fertilizer: 50 kg K -ha ⁻¹). TheAn aliquot of 6 kg of soil was weighed (bulk	
219	<u>density = 1.3 g/cm^3, mixed perfectthroughly with one of the above fertilizers,</u>	
220	respectively, and filled into its own column. This process was repeated for all three	

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221	replicates of the 11 fertilizer treatments. These soil columns were placed at the field	
222	experiment site in Huaxi District, Guiyang-of Southwestern China for a whole year.	
223	2.4 The rate of carbonate weathering	
224	Two different kinds of carbonate rock tablets (2 cm $\times 1$ cm $\times 0.5$ cm in size) were	
225	established placed in the bottom of each soil column to explore examine the rate of	
226	carbonate weathering in the soil. The two different kinds of carbonate rock collected	
227	from the karst area of Huaxi dDistrict were: (1) limestone with 60-65_% micrite,	
228	30-35_% microcrystalline calcite, and 2-3_% pyrite; and (2) dolostone with 98-99_%	
229	power-fine_crystalline dolomite, 3-5% microcrystalline calcite, 1_% pyrite, and little	
230	<u>trace quantities</u> organic matter. All <u>of the</u> tablets were <u>baked heated</u> at 80 $^{\circ}$ C for 4	
231	hours, then weighed in a 1/10000 electronic balance in the laboratory, tied to a labeled	
232	by tying a label with fishing line, and then buried at the bottom of each soil column.	
233	After a whole year, <u>-</u> Tthey tablets were taken outremoved carefully, rinsed, baked	
234	and weighed after a whole year.	
235	The amount of weathering earbonate weathering (Aew), the ratio of earbonate	
236	weathering(Rew) and the rate of earbonate-weathering (Raew) for limestone and	带格式的:字体:倾斜
237	dolomite were calculated according to the weight difference of the tablets using the	
238	following formulas:	
239	$\mathbf{A}\boldsymbol{\epsilon}\boldsymbol{w} = (\mathbf{W}\boldsymbol{i} - \mathbf{W}\boldsymbol{f}) \tag{1}$	
240	$\mathbf{R}\boldsymbol{\boldsymbol{\varepsilon}}\boldsymbol{\boldsymbol{w}} = (\mathbf{W}\boldsymbol{i} - \mathbf{W}\boldsymbol{f})/ \mathbf{W}\boldsymbol{i} \tag{2}$	
241	$R_{aew} = (Wi-Wf)/(S^*T) $ (3)	带格式的: 字体: 倾斜
242	where W_i is the initial weight of the carbonaterocktablets, W_f is the initial weights,	

243	S is the surface area of carbonate weathering rock tablets, and T is the length of the			
244	experiment <u>al</u> period.			
245	2.5 Statistical analysis			
246	Statistical analysis was performed using IBM SPSS 20.0 (Statistical Graphics	 带格式的: 厘米	缩进: 首行缩进	0.74
247	Crorp, Princeton, USA). All results of carbonate weathering were reported as the			
248	means ± standard deviations (SD)means standard errors (SE) for the three			
249	replicationes. as Statistical analysis was performed using IBM SPSS 20.0 (Statistical			
250	Graphics Crop, Princeton, USA).	带格式的:	字体:加粗	
251				
252	3. Results			
253	3.1 The wWeathering rate of carbonate under different fertilized treatments	 带格式的: 米	缩进: 首行缩进	0厘
254	weathering of under different treatments The amount (ARew), and the ratio			
255	(Raew_) and the rate (Raew) of limestone and dolostone earbonate weathering weare	带格式的:	字体:倾斜	
256	listed -in Table 2. were listed The results showed that in Table 2, and the Rew were			
257	plotted in Fig. 3. The results in Table 2 and Fig. 3 <u>The results showed Tthe Acw</u> , \mathbf{R}_{cw}	带格式的:	字体:倾斜	
258	and Racw of carbonate weatheringlimestone and dolomite weathering under urea,	带格式的:	字体:倾斜	
259	NH ₄ NO ₃ , <u>NH₄Cl, (NH₄)₂CO₃NH₄HCO₃, NH₄Cl-and <u>NH₄HCO₃ (NH₄)₂CO₃-treatments</u></u>			
260	were 8.48 ± 0.96 , 6.42 ± 0.28 , 5.54 ± 0.64 , 4.44 ± 0.81 and 4.48 ± 0.95 ‰ (mean \pm SD,			
261	$p \le 0.05$ positive, and much biggersignificantly greater than that under the control			
262	treatment 0.48 \pm 0.14 ‰ (see Fig. 3,-). In addition, the observed Rw of as observed in			
263	<u>dolomitestone were (6.59 ± 0.67, 5.30 ± 0.87, 4.77 ± 0.78, 4.94 ± 1.91 and 3.22 ±</u>			
264	$0.87 \ $ ^{mathcal{ma}			

265	-0.31 ± 0.09 % in the control treatment). This suggesting manifested clearly	
266	demonstrates that the addition of these five fertilizers can aid and increased the rate of	
267	the chemical weathering of carbonate weathering	
268		
269	The remaining st-treatments hadmade no significant differences in the Rw and #格式的: 字体: 倾斜 一种	
	The remaining st-treatments had made no significant differences in the Kw and 带格式的: 缩进: 首行	丁邹
270	Reaw of limestone and dolomitestone in comparison with to the control treatment (Fig. 带格式的: 字体: 倾斜	화
270	Real of Innestone and doto-innestone in comparison with to the control treatment (Fig. 带格式的: 字体: 倾斜	
274		
271	<u>3). In In the</u> (NH ₄) ₃ PO ₄ treatment, the Acw Rw, and Racw were only $1.08 \pm$ ##ALM: \neq A: [M]	
272	<u>0.34 ‰, 0.0028g</u> and <u>-0.00070.75 \pm 0.21‰g</u> for limestone and dolomite, <u>respectively</u> ,	
	帯格式的: 上标	
273	while the Raw were $4.00 \pm 1.15 \text{ g m}^{-2} \text{ a}^{-1} \pm 1.08\%$ and $1.00 \pm 1.01 \pm 0.75 \text{ g m}^{-2} \text{ a}^{-1}\%$ for #A AD: $\pm 4\%$	
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274	limestone and dolomite, respectively,— <u>. These values are less</u> than those under the 带格式的: 上标	
	带格式的: 字体:倾斜	<u>.</u>
275	other four NH ₄ -fertilizers, as mentioned above. The <u>Acw, Rew Rw</u> and Raew in the	
276	NaNO ₃ treatments failed to show a remarkable <u>notable</u> differences with the control	
277	treatment, implying exhibiting little effect of the NaNO3 fertilizer addition on	
278	carbonate weathering (Fig. 3)	
279	However, eExcept for the Rew of limestone approaching zero in the Ca ₃ (PO ₄) ₂	
280	treatment approaching zero, all the values of the Acw, Rew and Raca w of two 带格式的: 字体: 倾斜 带格式的: 字体: 倾斜	
281	different carbonate in Ca-Mg-P-and, K_2CO_3 and $Ca_3(PO_4)_2$ treatments showed a	
282	negative values. <u>. This</u> indicating indicates that the addition of Ca-Mg-P-, K ₂ CO ₃ and	
283	$Ca_3(PO_4)_2$ fertilizers <u>can leadled</u> to <u>the</u> -precipitation at the surface of <u>the</u> carbonate	
284	mineral, which can be explained by common ion effect.	
285	<u>3.2 The cComparison of limestone of dolomite</u>	- 1/-
286	In Fig. 4, we plotted the Rw of In order to compare The result of limestone with vs. 带格式的: 缩进: 首行 厘米 带格式的: 字体: 倾斜	

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287	dolomitestone tablets in a, we plotted Fig. 4, a linear correelation diagram, in order to		
288	compare the weathering responses of -with the Rw of limestone vs. dolostone		带格式的: 字体:倾斜
289	tabletslimestone with dolostone. The results shows that the Rw of limestone and		
290	dolostone exhibits a high positive correlaction $(R_1^2=0.9773_{\overline{1}}; see Fig. 4)$,		带格式的: 上标
291	suggestindicating that the weathering of ANOVA was use the limestone and		
292	dolostone are similar under different treatments were similar. Thus, we will explain		
293	the results with in terms of carbonates, rather than instead of by way of the individual		
294	dolostone and limestonewas used to determine the differences of weathering rate		
295	between limestone and dolostone.		
296 297	4. Discussion		
298	4.1 The carbonate rock tablet test: the validation of this experiment		
299	The carbonate-rock-tablet test is used to determine the weathering rate of	/	带格式的: 字体:加粗
300	carbonate rock/mineral-from laboratory to field (Gams, 1981; Chao et al., 2011;		带格式的: 字体:加粗 域代码已更改
301	Trudgill, 1975; Chao et al., 2014; Dreybrodt et al., 1996; Gams, 1985; Jiang and		
302	Yuan, 1999; Liu and Dreybrod, 1997; Plan, 2005), In laboratory, the	/	带格式的: 字体:加粗
303	carbonate-rock-tablet is employed to study the kinetics of caleite		
303 304	carbonate-rock-tablet is employed to study the kinetics of calcite dissolution/precipitation (Dreybrodt et al., 1996; Liu and Dreybrod, 1997), and		带格式的: 字体:加粗 域代码已更改
			 带格式的: 字体: 加粗 域代码已更改 带格式的: 字体: 加粗 带格式的: 字体: 加粗
304	dissolution/precipitation (Dreybrodt-et-al., 1996; Liu-and-Dreybrod, 1997) and		域代码已更改 带格式的: 字体:加粗
304 305	dissolution/precipitation <u>(Dreybrodt-et-al., 1996; Liu-and-Dreybrod, 1997)</u> and determine the rate of carbonate mineral weathering in soil column <u>(Chao et al.,</u>		域代码已更改 带格式的: 字体: 加粗 带格式的: 字体: 加粗 域代码已更改 带格式的: 字体: 加粗 带格式的: 字体: 加粗
304 305 306	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		域代码已更改 带格式的:字体:加粗 带格式的:字体:加粗 域代码已更改 带格式的:字体:加粗

310	estimated-CO ₂ -consumption by carbonate weathering at the regional/global scale			
		1	带格式的: 字体:加粗	
311	in the case of insufficient representative data (Liu, 2011), our results show that it	$\langle -$	域代码已更改	
			带格式的: 字体:加粗	
312	is a preferred option for the condition controlled contrast or stimulated			
		1	带格式的: 字体:加粗	
313	experiment (Chao et al., 2011; Chao et al., 2014), where the result from the		域代码已更改	
			带格式的:字体:加粗	
314	carbonate-rock-tablet test-is consistent to the major element geochemical data of			
		(
315	leachates from soil column(Chao et al., 2011),		域代码已更改	
			带格式的: 字体:加粗	
316	In this study, every procedure to establish soil column with carbonate rock	U	巾桁八的 , 丁件, 加祖	
317	tablets in the bottom of each was strictly same, including the size of column, the			
517	tublets in the bottom of each was strictly sund, metading the size of column, the			
318	preparation and column filling of soil sample, the setting and test of carbonate			
319	preparation and column mining of son sample, the setting and test of carbonate			
319	rock tablets, etc. Moreover, three replicates of each treatment were designed. We			
320	consider the experiment design can meet the objective of this study and the			
321	results of carbonate-rock-tablet test are therefore valid and credible.			
321		_	带格式的: 字体: 加粗	
321 322	results of carbonate-rock-tablet test are therefore valid and credible. <u>4.1 The kK</u> inetics of carbonate dissolution/precipitation: controlling factors		带格式的: 字体:加粗	
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322	4.1 The kK inetics of carbonate dissolution/precipitation: controlling factors	(带格式的: 字体:加粗	
322	4.1 The kK inetics of carbonate dissolution/precipitation: controlling factors	(带格式的: 字体:加粗	
322 323	<u>4.1 The kKjnetics of carbonate dissolution/precipitation: controlling factors</u> <u>Experimental studies of carbonate dissolution kinetics have shown metal</u>	(
322 323	4.1 The kKinetics 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	(带格式的: 字体:加粗 域代码已更改	
322 323 324	<u>4.1 The kKjnetics of carbonate dissolution/precipitation: controlling factors</u> <u>Experimental studies of carbonate dissolution kinetics have shown metal</u>	(
322 323 324 325	4.1 The kK inetics 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.,	(
322 323 324 325 326	4.1 The kK inetics 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):	(域代码已更改	
322 323 324 325	4.1 The kK inetics 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.,	(
322 323 324 325 326 327	4.1 The kK inetics 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 ⁻ (4)		域代码已更改 带格式的: 非上标/下标	
322 323 324 325 326	4.1 The kK inetics 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):		域代码已更改	
 322 323 324 325 326 327 328 	4.1 The kK inetics 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 ⁻ (4) MeCO ₃ + H ₂ CO ₃ \leftrightarrow Me ²⁺ + 2HCO ₃ ⁻ (5)		域代码已更改 带格式的: 非上标/下标	
322 323 324 325 326 327	4.1 The kK inetics 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 ⁻ (4)		域代码已更改 带格式的: 非上标/下标	
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 322 323 324 325 326 327 328 329 	4.1 The kK inetics of carbonate dissolution/precipitation: controlling factorsExperimental studies of carbonate dissolution kinetics have shown metalcarbonate weathering usually depends upon three parallel reactions occurring at thecarbonate interface (Chou et al., 1989; Plummer et al., 1978; Pokrovsky et al.,2009)(Chou et al., 1989; Plummer et al., 1978; Pokrovsky et al.,MeCO ₃ + H ⁺ \leftrightarrow Me ²⁺ + HCO ₃ ⁻ (4)MeCO ₃ + H ⁺ \leftrightarrow Me ²⁺ + 2HCO ₃ ⁻ (5)MeCO ₃ \leftrightarrow Me ²⁺ + CO ₃ ²⁻ (6)		域代码已更改 带格式的:非上标/下标 带格式的:非上标/下标	

332	with calcite- and dolomite-containing bedrock, H ₂ CO ₃ formed in the soil zone usually	
333	reacts with carbonate minerals, resulting in dissolved Ca, Mg, and HCO3 ⁻ as described	
334	in Eq. (5) (Andrews and Schlesinger, 2001; Shin et al., 2014)(Andrews and 2001; Shin et al., 2014)(
335	Schlesinger, 2001; Shin et al., 2014). Although it has been proven that the reaction of	
336	carbonate dissolution is mainly controlled by the amount of rainfall (Amiotte Suchet 域代码已更改	
337	et al., 2003; Egli and Fitze, 2001; Kiefer, 1994)(Amiotte Suchet et al., 2003; Egli and	
338	Fitze, 2001; Kiefer, 1994), we consider that the effect of rainfall is equal in each soil	
339	column, and hence is disregarded unconsidered as a controlling factor in this study.	
340	The-Eq. (4) suggests that the proton from other origins, such as the nitrification	
341	processes of NH_4^+ , as mentioned in the iIntroduction section, can play the role of	
342	weathering agent in agricultural areas. In this study, the urea, NH ₄ NO ₃ , NH ₄ HCO ₃ ,	
343	<u>NH₄Cl, and (NH₄)₂CO₃ amendments increased (10 to 17-fold) the natural weathering</u>	
344	rate of from 2.00 g m ⁻² a ⁻¹ from for limestone tablets in the control treatment (\pm Table 2).	
345	Thus, these increases are strongly relativeted to the effect of the proton released from	
346	the nitrification of NH_4^+ . OnIn the contraryst, the carbonate precipitation will occur as	
347	due to the backward reaction of the Eq. (5) in the following cases: (1) the degassing of	
348	<u>dissolved CO_{25}; (2) soil evapotranspiration; or (3) the common ion effect: the increase</u>	
349	of Ca ²⁺ , Mg ²⁺ or CO ₃ ²⁻ in a weathering-system with equilibrium between water and	
350	<u>calcite (Calmels et al., 2014; Dreybrodt, 1988)(Calmels et al., 2014; Dreybrodt,</u> 域代码已更改	
351	<u>1988).</u>	
352	4.2 The mMain reactions and effects in different treatments	
353	The main reactions and effects of every treatment in this study weare listed in 带格式的: 缩进: 首行缩进: 厘米	0.74

354	Table 3.	
355	(1) The nNitrification in NH ₄ -fertilizer: NH ₄ NO ₃ , NH ₄ HCO ₃ , NH ₄ Cl, (NH ₄) ₂ CO ₃	
356	and urea	
357	In urea (CO(NH ₂) ₂) treatment, the enzyme urease rapidly hydrolyzes the urea-N \leftarrow	带格式的: 缩进: 首行缩进: 0.74 厘米
358	(CO(NH ₂) ₂) to NH ₄ ⁺ ions (Eq. (7)) when urea is applied to the soil (Soares et al.,	域代码已更改
359	<u>2012)(Soares et al., 2012).</u>	
360	$\underline{\text{CO}(\text{NH}_2)_2 + 3\text{H}_2\text{O}} \to 2\text{NH}_4^+ + 2\text{OH}^- + \text{CO}_2 $ (7)	
361	Although the study from f Singh et al. showed that a part of NH_4^+ may be lost as	 带格式的: 缩进:首行缩进: 0.74 厘米
362	ammonia (NH ₃) and subsequently as nitrous oxide (N ₂ O) (Singh et al., 2013)(Singh et al., 2013)	域代码已更改
363	al., 2013), yet the restremaining ammonium (NH_4^+) is mainly oxidized during	
364	nitrification in soil by autotrophic bacteria-(, likesuch as Nitrosomonas,) during	
365	<u>nitrification</u> , resulting in nitrite NO_2^- and H ⁺ ions. Nitrite is, <u>-in turn</u> , oxidized by	
366	another bacterium, such as Nitrobacter, resulting in nitrate (NO ₃) (Eq. (8)) (Perrin et	域代码已更改
367	<u>al., 2008)(Perrin et al., 2008).</u>	
368	$\underline{\mathrm{NH}_4}^+ + 2\mathrm{O}_2 \longrightarrow \mathrm{NO}_3^- + \mathrm{H}_2\mathrm{O} + 2\mathrm{H}^+ \tag{8}$	
369	<u>The protons (H⁺) produced by nitrification can be neutralized in two ways:</u> \bullet	 带格式的: 缩进: 首行缩进: 0.74 厘米
370	(i) either by exchange process with base cations in the soil exchange complex	 带格式的: 缩进: 悬挂缩进: 0.06 字符, 左 2.02 字符
371	(Eq. (9)) Soil – Ca + 2H ⁺ \rightarrow Soil – 2H ⁺ + Ca ²⁺ (9)	
372	or (ii)-or via carbonate mineral dissolution (Eq.(10))	带格式的: 缩进:首行缩进: 0.74 厘米
373	$\underline{Ca_{(1-x)}Mg_{x}CO_{3}} + H^{+} \rightarrow (1-x)Ca^{2+} + xMg^{2+} + HCO_{3}^{-} $ (10)	
374	Consequently, afterby combining Eq. (8) and Eq. (10)-are combined, carbonate	
375	weathering by protons produced by nitrification iscan supposedly be expressed as to	
376	becomes (Eq. 11) (See details in Perrin et al., 2008 and Gandois et al., 2011).	

377	$\underline{2Ca_{(1-x)}Mg_{x}CO_{3} + NH_{4}^{+} + 2O_{2} \rightarrow 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{3}^{-} + H_{2}O + 2HCO_{3}^{-}}$	
378	<u>(11)</u>	
379	As discussed above, provided that the loss as ammonia (NH ₃) and nitrous oxide ^{\bullet}	带格式的: 缩进: 首行缩进: 0.74 厘米
380	(N_2O) after hydrolyzation is unconsidered disregarded in this study, the final equation	
381	of carbonate weathering in NH ₄ NO ₃ , NH ₄ HCO ₃ , NH ₄ Cl, (NH ₄) ₂ CO ₃ and urea	
382	treatments will be as followeds-as, respectively:	
383	$\underline{2Ca_{(1-x)}Mg_{x}CO_{3} + NH_{4}NO_{3} + 2O_{2} \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + 2NO_{3}^{-} + H_{2}O + 2MG_{3}^{-} + M_{2}O_{3} + M_$	
384	<u>2HCO₃ (12)</u>	
385	$\underline{2Ca_{(1-x)}Mg_{x}CO_{3} + NH_{4}HCO_{3} + 2O_{2} \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + NO_{3}^{-} + H_{2}O + 2(1-x) Ca^{2+} + 2(1-x) Ca^{2+} + 2(1-x) Ca^{2+} + NO_{3}^{-} + H_{2}O + 2(1-x) Ca^{2+} + 2(1-x) Ca^{2+} + 2(1-x) Ca^{2+} + NO_{3}^{-} + H_{2}O + 2(1-x) Ca^{2+} + 2(1-x) Ca^{2+} + NO_{3}^{-} + H_{2}O + 2(1-x) Ca^{2+} + 2(1-$	
386	<u>3HCO₃ (13)</u>	
387	$\underline{2Ca_{(1-x)}Mg_{x}CO_{3} + NH_{4}Cl + 2O_{2} \rightarrow 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{3}^{-} + Cl^{-} + H_{2}O + 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{3}^{-} + Cl^{-} + H_{2}O + 2(1-x)Ca^{2+} + 2(1-x)Ca^{2+} + 2(1-x)Ca^{2+} + NO_{3}^{-} + Cl^{-} + H_{2}O + 2(1-x)Ca^{2+} + 2(1-x)Ca^{2+} + NO_{3}^{-} + Cl^{-} + H_{2}O + 2(1-x)Ca^{2+} + 2(1-x)Ca^{2+} + 2(1-x)Ca^{2+} + NO_{3}^{-} + Cl^{-} + H_{2}O + 2(1-x)Ca^{2+} + 2(1-x)Ca^$	
388	<u>2HCO₃ (14)</u>	
389	$\underline{3Ca_{(1-x)}Mg_{x}CO_{3} + (NH_{4})_{2}CO_{3} + 4O_{2} \rightarrow 3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{3}^{-} + 2H_{2}O + 2NO_{3}^{-} + 2NO_{3}^$	
390	<u>4HCO₃ (15)</u>	
391	$\underline{3Ca_{(1-x)}Mg_{x}CO_{3} + CO(NH_{2})_{2} + 4O_{2} \rightarrow 3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{3}^{-} + 4HCO_{3}^{-}}$	
392	<u>(16)</u>	
393	(2) No effect of NO ₃ -fertilizer treatment: NaNO ₃ treatment	
394	In the NaNO ₃ treatment, the reaction occurs according to as-Eq. (17), indicating	带格式的: 缩进: 首行缩进: 0.74 厘米
395	that the addition of NO3-fertilizer does not significantly influence carbonate	
396	weathering.	
397	$\underline{Ca_{(1-x)}Mg_{x}CO_{3} + NaNO_{3} + CO_{2} + H_{2}O \rightarrow (1-x)Ca^{2+} + xMg^{2+} + Na^{+} + NO_{3}^{-} + M_{3}^{-} + M_{3}^{-$	
398	<u>2HCO₃ (17)</u>	
399	(3) The cCommon ion effect: K ₂ CO ₃ treatment	
400	In the K_2CO_3 treatment, CO_3^2 and HCO_3^2 will be produced after the addition of	带格式的: 缩进:首行缩进: 0.74 厘米
401	<u>K_2CO_3 according to Eq. (18) after adding K_2CO_3, hence resulting in carbonate</u>	带格式的: 下标 带格式的: 下标
	18	

402	precipitation as described in Eq. (19), due to the common ion effect.					
403	$\underline{K_2CO_3 + H_2O} \rightarrow 2K^+ + HCO_3^- + OH^- $ (18)	 带格式的: 米	缩进:	左侧: 0	.74 /国	Ē
404 405	$(1-x) Ca^{2+} + xMg^{2+} + 2HCO_3^{-} \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O $ (19) (4) Complex effects: Nitrification versus H inhibition effect of PO ₄ in (NH ₄) ₃ PO ₄					
406	treatments					
407	<u>For In the $(NH_4)_3PO_4$ treatment, the reaction of carbonate weathering will occur</u>	 带格式的: 厘米	缩进:	首行缩进	: 0.	74
408	according to Eq. (11) due to the nitrification of NH_4^+ ionized from the $(NH_4)_3PO_4$					
409	fertilizer-will occur the nitrification. Whilst tThe PO_4^{3-} anion will exert an inhibition					
410	to calcite dissolution:, as calcium orthophosphate (Ca-P) precipitation is producesd on					
411	the surface of calcite after the addition of PO_4^{3-} in soil (reaction: <u>Ca + PO_4</u> \rightarrow Ca-P),	带格式的:	字体:	小四		
412	resulting in inhibitingion of the calcite dissolution-of calcite.					
413	(5) Complex effects: Common ion effect versus Linhibition effect of PO ₄ in					
414	Ca ₃ (PO ₄₎₂ and Ca-Mg-P treatments					
415	In the Ca ₃ (PO ₄) ₂ and Ca-Mg-P treatments, on the one hand, the Ca _(1-x) Mg _x CO _{3 is}	 带格式的: 厘米	缩进:	首行缩进	: 0.	74
416	produceds when the concentrations of Ca^{2+} (or/and Mg^{2+}) increases as according to					
417	following Eq. (19). On the other hand, the inhibition effect of phosphate will cause					
418	that-calcium phosphate precipitation to be producesd on the surface of carbonate					
419	minerals after the addition of P in soil (reaction: $Ca + PO_4 \rightarrow Ca-P$), correspondingly	带格式的:	下标			
420	resulting in inhibitingion the carbonate precipitation.					
421	<u>4.3 The dD</u>ifference between NH_4^+ and NO_3^- in impacts on carbonate weathering					
422	and the implication on the estimation of CO ₂ consumption					
423	In order to further compare the difference S between NH_4^+ and NO_3^- effects on	带格式的: 厘米	缩进:	首行缩进	: 0.	74
424	carbonate weathering, the initial molar amount of fertilizer-derived NH ₄ per unit in					

425	every treatment were calculated, and are listed in Table 4. The results show that the	
426	amount of NH_4^+ hydrolyzed from urea is 1.06 mole, while NH_4^+ ionized from	
427	<u>NH₄NO₃, NH₄HCO₃, NH₄Cl, (NH₄)₂CO₃ and (NH₄)₃PO₄ is 0.54 mole, 1.08 mole,</u>	
428	1.07 mole, 1.06 mole, and 0.03 mole, respectively (Table 4). The Rw of limestone	
429	tablets and the initial amount of NH_4^+ per treatment are plotted in Fig. 45. A distinct	
430	relationship between them is observed, in that: the Rw values in NH ₄ NO ₃ , NH ₄ HCO ₃ ,	
431	<u>NH₄Cl, (NH₄)₂CO₃ and urea treatments are bigg larger than in the control treatment,</u>	
432	where the initial amount of NH_4^+ displaysyields similar results (Fig. 45). This	
433	suggests that carbonate weathering in NH ₄ NO ₃ , NH ₄ HCO ₃ , NH ₄ Cl, (NH ₄) ₂ CO ₃ and	
434	urea treatments are mainly attributed to the dissolution reaction described as Eq. (11).	
435	This process of carbonate weathering by protons released from nitrification has been	
436	proven by many studies, from the laboratory to the field (Barnes and Raymond, 2009;	域代码已更改
437	Bertrand et al., 2007; Biasi et al., 2008; Chao et al., 2011; Errin et al., 2006; Gandois	
438	et al., 2011; Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Semhi	
439	and Suchet, 2000; West and McBride, 2005)(Barnes and Raymond, 2009; Bertrand et	
440	al., 2007; Biasi et al., 2008; Chao et al., 2011; Errin et al., 2006; Gandois et al., 2011;	
441	Hamilton et al., 2007; Oh and Raymond, 2006; Perrin et al., 2008; Semhi and Suchet,	
442	2000; West and McBride, 2005). We have noted that the Rw values in NH ₄ HCO ₃ and	
443	(NH ₄) ₂ CO ₃ treatments are lowerless than even half of those in urea treatment in	
444	despite-of adding the same amount of fertilizer-derived NH ₄ (about approximately	
445	<u>1.07 mole). This is probably because the two fertilizers, NH_4HCO_3 and $(NH_4)_2CO_3$.</u>	
446	are easier to decompose and produce the-NH ₃ and CO ₂ gases as followingaccording to	
447	Eq. (20) and (21), resulting in the amounts of fertilizer-derived NH ₄ of that are lower	
448	than 1.07 moles.	
449	$\underline{\text{NH}_{4}\text{HCO}_{3}} \rightarrow \underline{\text{NH}_{3}} \uparrow + \underline{\text{H}_{2}\text{O}} + \underline{\text{CO}_{2}} \uparrow \tag{20}$	带格式的: 右,缩进:首行缩进: 0.74 厘米

$(NH_4)_2CO_3 \rightarrow 2NH_3 \uparrow + H_2O + CO_2 \uparrow $ (21)	
The Aw and Rw in the $(NH_4)_3PO_4$ treatment, unlike in other NH_4 -fertlizer	带格式的: 缩进: 首行缩进: 0.74 厘米
treatments, had not do not show a significant increase comparinged 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.,	域代码已更改
<u>2012</u>). 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;	域代码已更改
Wang et al., 2012)(Chien et al., 2011; Wang et al., 2012).	
However, in Fig. 3, there is no significant different between the Rw without	
significant difference with control treatment in the NaNO3 treatment compared to the	
control treatment, indicatinges that the addition of NO3-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 ₄ NO ₃ , NH ₄ HCO ₃ , NH ₄ Cl, (NH ₄) ₂ CO ₃	
and urea), but it fails to impact on carbonate weathering, which is raisinges 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	
estimationes 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	域代码已更改
et al., 2009). AtIn agricultural areas, the relationship between (Ca+Mg)/HCO ₃ ² and 21	
	The Aw and Rw in the (NH ₄);PO ₄ treatment, unlike in other NH ₄ -fertlizer- treatments, had note only owing to the low amount of added NH ₆ ⁺ in (NH ₄);PO 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);PO 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). 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, indicatinges 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 ₄ NO ₃ , NH ₄ HCO ₃ , NH ₄ Cl, (NH ₄) ₂ CO ₃ and urea), but it fails to impact on carbonate weathering, which it-raisinges 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, is an important reaction—for in understanding the kinetics process of Carbonate, is an important reaction—for in understanding the kinetics process of Carbonate, is an important reaction—for in understanding the kinetics process of Carbonate, is an important reaction—for in understanding the kinetics process of Carbonate, is an important reaction—for in understanding the kinetics process of Carbonate, is an important reaction—for in understanding the kinetics process of Carbonate, is an important reaction—for in understanding the kinetics process of Carbonate, is an

475	NO_3 is usually employed to estimate the contribution of N-fertilizer to riverine Ca^{2+} ,	
476	Mg ²⁺ , and alkalinity (Etchanchu and Probst, 1988; Jiang, 2013; Jiang et al., 2009;	ţ,
477	Perrin et al., 2008; Semhi and Suchet, 2000)(Etchanchu and Probst, 1988; Jiang, 2013;	
478	Jiang et al., 2009; Perrin et al., 2008; Semhi and Suchet, 2000). In these studies, the	
479	nitrification described as in Eq. (8) is usually considered as the unique origin of NO_3^{-} .	
480	According to the results of the NaNO ₃ treatment in this study, the contribution of	
481	protons from nitrification to carbonate weathering may be overestimated, if	
482	anthropogenic NO ₃ ⁻ is neglected, since the anthropogenic NO ₃ ⁻ does not release the	
483	proton described asin Eq. (8). For NH ₄ NO ₃ fertilizer, the (Eq. (12)) shows that the two	
484	moles of $Ca^{2+}+Mg^{2+}$, NO_3^- , and HCO_3^- will be produced when one mole NH_4NO_3	
485	reacts with 2 moles of carbonate, where only half of the NO ₃ originates from	
486	nitrification described as Eq. (8). This will result in a double overestimation onf the	
487	contribution of the nitrification to carbonate weathering, and thus thereby mislead the	
487 488	contribution of the nitrification to carbonate weathering, and thus thereby mislead the estimation of CO ₂ consumption therein.	
488	estimation of CO ₂ consumption-therein.	
488 489	estimation of CO ₂ consumption therein. At regional scales, Hif different fertilizers are added simultaneously added to an	
488 489 490	estimation of CO ₂ consumption-therein. At regional scales, Iif different fertilizers are-added simultaneously added to an agricultural area, the estimation of CO ₂ consumption by carbonate weathering might	
488 489 490 491	estimation of CO ₂ consumption-therein. <u>At regional scales</u> , <u>Fif different fertilizers are-added simultaneously added to an</u> <u>agricultural area</u> , the estimation of CO ₂ consumption by carbonate weathering might <u>became more complicated</u> , since the mole ratios of Ca+Mg, HCO ₃ ⁻ -, and/or NO ₃ ⁻	
488 489 490 491 492	estimation of CO ₂ consumption therein. <u>At regional scales</u> , <u>Fif different fertilizers are added simultaneously added to an</u> 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	
488 489 490 491 492 493	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 isare different (see Table 3). Thus, the related anthropogenic inputs (e.g. Ca+Mg, NH ₄ , NO ₃ ⁻ , HCO ₃ ⁻ , etc.) need to be	
488 489 490 491 492 493 494	estimation of CO ₂ consumption-therein. <u>At regional scales, Fif different fertilizers are-added simultaneously added to an</u> 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 isare 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	
488 489 490 491 492 493 494 495	estimation of CO ₂ consumption-therein. At regional scales, Hif 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 isare 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.	<u>0</u>
488 489 490 491 492 493 494 495 496	 estimation of CO₂ consumption therein. At regional scales, Fif different fertilizers are-added simultaneously added to an agricultural area, the estimation of CO₂ consumption by carbonate weathering might became more complicated, since the mole ratios of Ca+Mg, HCO₃, and/or NO₃ between different fertilization treatments isare different (see Table 3). Thus, the related anthropogenic inputs (e.g. Ca+Mg, NH₄, NO₃⁻, HCO₃⁻, etc.) need to be investigated to more accurately estimate the impact of fertilization on carbonate weathering and its CO₂ consumption. 4.4 The comparison with other studied results_ 	<u>0</u> 件 胆

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500	Southwestern China (Zhang, 2011)(Zhang, 2011), and with the results of 0.05-5.06 %	域代码已更改
501	(for Rw) and 1.08-136.90 g m ⁻² a ⁻¹ (for Raw) from the north slope of the Hochschwab	
502	mMassif in Australia (Plan, 2005)(Plan, 2005), as determined -using the limestone	域代码已更改
503	tablet method. But the Raw of 2.00 g m ⁻² a ⁻¹ is lower than the results (of 7.0-63.5)	
504	g m ⁻² a ⁻¹ for Raw) from Jinfo Mountiain in Chongqing-of, China (Zhang,	域代码已更改
505	2011)(Zhang, 2011). These differences in carbonate weathering are mainly attributed	
506	to the different types of carbonate rock tablets, climate, micro-environments of soil,	
507	etc. The Raw of limestone in the N-fertilizers treatments is are 20.57-34.71 g m ⁻² a ⁻¹ ,	
508	similar to the weathering rate of carbonate in Oorchard (32.97 g m ⁻² a ⁻¹) at Nongla,	
509	Manshan, Guangxi-of-, China, whereich usually involves in fertilization activities.	
510	At larger scales, such as -like watersheds, the weathering rate is usually	
511	estimated by using the riverine hydro-chemical method, which is inconsistent with the	
512	results from the carbonate-rock-tablet test. The estimation of Zeng, et al. (2014)	
513	estimateviews that the carbon sink intensity calculated by the carbonaterocktablet	
514	test is only one sixth of that estimated by using the riverine hydro-chemical method,	
515	due to its own limits in methodology (Zeng et al., 2014)(Zeng et al., 2014). The	域代码已更改
516	results from Semhi, et al. (2000) shows theat weathering rates of carbonate rock by	
517	using riverine hydro-chemical method are- about approximately 77.5 g m ⁻² a^{-1} and	
518	50.4 g m ⁻² a ⁻¹ in the upstream and downstream, respectively, of the Garonne river,	
519	France, respectively, which are about approximately 25-35 and 2-3-times greater than	
520	that in the control treatment (2.00 g m ⁻² a ⁻¹ for natural weathering rate-) and 2-3 times	
521	greater than in the N-fertilizer treatment (20.57-34.71 g m ⁻² a ⁻¹ for anthropic	
522	weathering rate) in this study. The global natural weathering rate of carbonate	
523	reported by Amiotte Suchet, et al. (2003) is 47.8 g m ⁻² a ⁻¹ , which is much higher than	
524	that we observed. Thus, we conclude that it is difficult to compare between the results	

525	from the carbonate-rock-tablet test and the riverine hydro-chemical method. The	
526	carbonate-rock-tablet test is suitable for the research on the condition	
527	eontrolled comparative contrast or stimulated experiments, while the riverine	
528	hydro-chemical method is appropriate for the regional investigations and estimations.	
529	According to the estimation from Yue et al. (2015), $\frac{1}{2}$ the enhanced HCO ₃ flux due to	
530	<u>nitrification of NH_4^+ at Houzhai catchment of Guizhou pProvince would be 3.72×10^5</u>	
531	kg C/year and account for 18.7 % of this flux in the entire catchment(Yue et al.,	域代码已更改
532	2015)(Yue et al., 2015). This is similar to estimates from other small agricultural	
533	carbonate basins (12-26 %) in Southwest France (Perrin et al., 2008; Semhi and	域代码已更改
534	Suchet, 2000)(Perrin et al., 2008; Semhi and Suchet, 2000).	
535	5. Conclusions	
536	The impact of the addition of different fertilizers (NH ₄ NO ₃ , NH ₄ HCO ₃ , NaNO ₃ ,	
537	<u>NH₄Cl, (NH₄)₂CO₃, Ca₃(PO₄)₂, (NH₄)₃PO₄, Ca-Mg-P, Uurea, and K₂CO₃) on</u>	
538	carbonate weathering was studied in a field column experiment withusing carbonate	
539	-rocktablets at its bottom of each. The amount of weathering amount-and the ratio of	
540	weathering of carbonate rock tablets showed that the addition of urea, NH ₄ NO ₃ ,	
541	<u>NH₄HCO₃, NH₄Cl, and (NH₄)₂CO₃ distinctly increased carbonate weathering, which</u>	
542	was attributed to the nitrification of NH_{4^+} , and while the addition of $Ca_3(PO_4)_{2_4}$	
543	<u>Ca-Mg-P and K₂CO₃ induced carbonate precipitation due to the common ion effect.</u>	
544	<u>While tThe addition of $(NH_4)_3PO_4$ and $NaNO_3$ addition did not impact significantly</u>	
545	on carbonate weathering, where the former can be attributed to the low added amount	
546	of (NH4) ₃ PO ₄ ,- and may be related to the inhibition of phosphate, and while the	
547	latter seemed to be-raisinge a new question. The littleminor impact of nitrate on	

548	carbonate weathering may result in the overestimation of the impact of N-fertilizer on
549	CO ₂ consumption by carbonate weathering at the regional/global scale, if the effects
550	of NO ₃ and NH ₄ are not distinguished. Thus, the related anthropogenic inputs (e.g.
551	Ca+ Mg, NH ₄ , NO ₃ , HCO ₃ , etc.) need to be investigated to more accurately estimate
552	the impact of fertilization on carbonate weathering and its consumption of CO2
553	consumption. 4.2 The kinetics and controlled factors of carbonate weathering
554	Experimental studies of carbonate dissolution kinetics have shown metal
555	carbonate weathering usually depends upon three parallel reactions occurring at the
556	carbonate interface (Chou et al., 1989; Plummer et al., 1978; Pokrovsky et al., 2009):
557	$\frac{\text{MeCO}_3 + \text{H}^+ \rightarrow \text{Me}^{2+} + \text{HCO}_3}{(4)}$
558	$\frac{\text{MeCO}_3 + \text{H}_2\text{CO}_3 \rightarrow \text{Me}^{2+} + 2\text{HCO}_3}{(5)}$
559	$MeCO_{3} \longrightarrow Me^{2+} + CO_{3}^{2-} $ (6)
560	where Me=Ca, Mg. As Eq. (5) describes, atmospheric/soil CO_2 is usually regard as
561	the natural weathering agent of carbonate, whereas many studies have exposed that
562	carbonate weathering can occur due to the reaction (Eq. (4)) between carbonate and
563	the other proton contributors, as mentioned in introduction section: s which can
564	originate from the nitrification processes of N-fertilizer H ₄ \pm (Semhi and Suchet, 2000;
565	West and McBride, 2005; Oh and Raymond, 2006; Hamilton et al., 2007; Perrin et al.,
566	2008; Barnes and Raymond, 2009; Pierson wickmann et al., 2009; Chao et al., 2011;
567	Gandois et al., 2011), from the sulfuric acid acid, (Lerman and Wu, 2006; Lerman et
568	al., 2007; Li et al., 2008; Li et al., 2009), from organic acid secreted by
569	microorganisms (Lian et al., 2008), as well and as from acidic soil (Chao et al.,

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570	2014)<u>the role of</u>
570	2014)<u>the role of</u>.

571	In field, carbonate dissolution is mainly controlled by the amount of rainfall (Amiotte		带格式的: 米	缩进:	首行缩进:	0 厘
572	Suchet et al., 2003; Egli and Fitze, 2001; Kiefer, 1994), as well as impacted of soil		_ ·			
572						
573	CO ₂ (Andrews and Schlesinger, 2001). We consider that the effect of rainfall on each					
574	soil column is same. In this study, the urea, NH4NO3, NH4HCO3, NH4Cl and					
575	(NH ₄) ₂ CO ₃ -amendment increased (10 to 17 fold) the natural weathering rate of 2.00					
576	g m ⁻² -a ⁻¹ -from limestone tablets in control treatment (table 2). These increases may be,					
577	in the one hand, attributed to the effect of the proton released from the nitrification of					
578	NH4 ⁺ . On the other hand, it may be, in theory, related to enhanced microbiogenic CO ₂					
579	due to nitrogenous nutrients stimulation (Eq(5)), because fertilizer application can					
580	increase soil CO ₂ -flux (Sainju et al., 2008; Bhattacharyya et al., 2013), the increased					
581	CO2 can enhance carbonate dissolution rate at near neutral or alkali pH (Andrews and					
582	Schlesinger, 2001).					
583	According to the added amount of different fertilization treatment, the molar amount					
584	of added nitrogen nutrient in NaNO3 treatment is 1.07mol, much bigger than in					
585	NH4NO3, equivalent to NH4HCO3 and NH4Cl treatment. However, the Acw and Rcw,					
586	and Racw of NaNO3 treatment is far less (Fig. 3 and table 2), inhibiting that the	1	带格式的:	字体:	倾斜	
587	increases of carbonate weathering rate in urea, NH4NO3, NH4HCO3, NH4Cl and					
588	$(NH_4)_2CO_3$ -amendment have no distinct relationship with enhanced microbiogenic					
589	CO ₂ -due to nitrogenous fertilizer amendment.					
590	4.3 The effect of nitrification of NH4-fertilizer		带格式的: 符,定义网 调整西文与	格后不	调整右缩进	0字 ,不 不
591	In urea (CO(NH ₂) ₂) treatment, the enzyme urease rapidly hydrolyzes the urea N		调整西文与调整中文和	/ - <u>、</u> 之 数字之	间的空格	
592	(CO(NH ₂) ₂) to NH ₄ ⁺ ions (Eq. (7)) when urea is applied to the soil (Soares et al., 26					

593	2012).
594	$\frac{\text{CO}(\text{NH}_2)_2 + 3\text{H}_2\text{O} \rightarrow 2\text{NH}_4^+ + 2\text{OH}^- + \text{CO}_2}{(7)}$
595	Table 3 shows that the amount of NH4 ⁺ hydrolyzed from urea is 1.06 mol, while
596	NH4 ⁺ -ionized from NH4NO3, NH4HCO3, NH4Cl, (NH4)2CO3-and (NH4)3PO4-is 0.54
597	mol, 1.08 mol, 1.07 mol, 1.06 mol and 0.03 mol, respectively (Table 3). Although the
598	study from Singh et al showed that a part of NH_4^+ may be lost as ammonia (NH_3) and
599	subsequently as nitrous oxide (N_2O) (Singh et al., 2013), yet the rest ammonium
600	$(\mathbf{NH_4}^+)$ is mainly oxidized in soil by autotrophic bacteria (like Nitrosomonas) during
601	nitrification, resulting in nitrite NO_2^- and H^+ ions. Nitrite is, in turn, oxidized by
602	another bacterium, such as Nitrobacter, resulting in nitrate (NO37) (Eq. (8)) (Perrin et
603	al., 2008).
604	$\mathbf{NH_4}^+ + \mathbf{2O_2} \longrightarrow \mathbf{NO_3}^- + \mathbf{H_2O} + \mathbf{2H}^+ - \mathbf{(8)}$
604 605	$NH_4^+ + 2O_2 \rightarrow NO_3^- + H_2O + 2H^+$ (8) The protons (H ⁺) produced by nitrification can be neutralized in two ways:
605	The protons (H^+) produced by nitrification can be neutralized in two ways:
605 606	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
605 606 607	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⁺ (9)
605 606 607 608	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 ⁺ – Soil – 2H ⁺ + Ca ²⁺ – (9) (ii) or via carbonate mineral dissolution (Eq.(10))
605 606 607 608 609	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⁺ (9) (ii) or via carbonate mineral dissolution (Eq.(10)) Ca _(1-x) Mg _x CO ₃ + H ⁺ \rightarrow (1-x) Ca ²⁺ + xMg ²⁺ + HCO ₃ ⁻ (10)
605 606 607 608 609 610	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 ⁺ –> Soil – 2H ⁺ + Ca ²⁺ – (9) (ii) or via carbonate mineral dissolution (Eq.(10)) $Ca_{(1-x)}Mg_xCO_3 + H^+ \rightarrow (1-x)Ca^{2+} + xMg^{2+} + HCO_3^-$ (10) — Consequently, after Eq. (8) and Eq. (10) are combined, carbonate weathering by
605 606 607 608 609 610 611	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⁺ – (9) (ii) or via carbonate mineral dissolution (Eq.(10)) Ca _(1-x) Mg _x CO ₃ + H ⁺ \rightarrow (1-x) Ca ²⁺ + xMg ²⁺ + HCO ₃ ⁻ – (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
605 606 607 608 609 610 611 612	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⁺ – (9) (ii) or via carbonate mineral dissolution (Eq.(10)) Ca _(1-x) Mg _x CO ₃ + H ⁺ \rightarrow (1-x) Ca ²⁺ + xMg ²⁺ + HCO ₃ ⁻ – (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).

616	Fig. 4. A distinct relationship between them is observed: the Acw and Rcw in NH_4NO_3 ,
617	NH_4HCO_3 , NH_4Cl , $(NH_4)_2CO_3$ and urea treatments are bigger than in control
618	treatment, where the initial concentration of NH_4^+ -displays similar results (Fig. 4).
619	This suggests that carbonate weathering in NH4NO3, NH4HCO3, NH4Cl, (NH4)2CO3
620	and urea treatments are mainly attributed to the dissolution reaction described as Eq.
621	(11). This process of carbonate weathering by protons from nitrification has been
622	proven by many studies, from laboratory to field (Semhi and Suchet, 2000; Bertrand
623	et al., 2007; Oh and Raymond, 2006; Errin et al., 2006; Hamilton et al., 2007; Biasi et
624	al., 2008; Perrin et al., 2008; Barnes and Raymond, 2009; Chao et al., 2011; West and
625	McBride, 2005; Gandois et al., 2011). According to the estimation from Yue et al.
626	(2015), The enhanced HCO ₃ ⁻ flux due to nitrification of NH_4^+ at Houzhai catchment
627	of Guizhou province would be 3.72×10^5 -kg C/year and account for 18.7% of this
628	flux in the entire catchment(Yue et al., 2015). This is similar to estimates from other
629	small agricultural carbonate basins (12-26%) in Southwest France (Semhi and Suchet,
630	2000; Perrin et al., 2008).
631	As discussed above, provided that the loss as ammonia (NH ₃) and nitrous oxide
632	(N_2O) after hydrolyzation is unconsidered in this study, the final equation of
633	carbonate weathering in NH4NO3, NH4HCO3, NH4Cl, (NH4)2CO3 and urea treatments
634	will be followed as, respectively:
635	$\frac{2Ca_{(1-x)}Mg_{x}CO_{3} + NH_{4}NO_{3} + 2O_{2} \rightarrow 2(1-x) Ca^{2+} + 2xMg^{2+} + 2NO_{3}^{-} + H_{2}O_{3} + H_{2}O_{3}}{2}$
636	2HCO ₃ (12)
637	$\frac{2Ca_{(1-x)}Mg_{*}CO_{3} + NH_{4}HCO_{3} + 2O_{2} \rightarrow 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{3} + H_{2}O + CO_{3}}{2}$
638	3HCO₃ (13)
	28

The Acw and Rcw in (NH4)3PO4 treatment, unlike in other NH4 fertlizer 645 646 treatments, had not a significant increase comparing with control treatment, which is 647 not only owing to the low amount of added NH4⁺ in (NH4)3PO4 treatment (0.3 mol, see Table 3) but also relative to the inhibition of phosphate. After the addition of 648 649 (NH4)₃PO₄-in soil, calcium orthophosphate (Ca-P) precipitation will form on calcite surface which is initiated with the aggregation of clusters leading to the nucleation 650 and subsequent growth of Ca-P phases, at various pH values and ionic strengths 651 652 relevant to soil solution conditions (Chien et al., 2011; Wang et al., 2012).

653 **4.4 Little/no effect of NO₃-fertilizer on carbonate weathering and its implication**

654 to the evaluation of CO₂ consumption by carbonate weathering

In Fig. 3, the Acw and Rcw without significant difference with control treatment
in NaNO₃ treatment indicates that the addition of NO₃-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 HCO₃⁻and Me²⁺-in
the river as an indicator is usually used to make estimations of CO₂ 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)/HCO₃⁻and NO₃⁻

664	is usually employed to estimate the contribution of N-fertilizer to riverine Ca ²⁺ , Mg ²⁺
665	and alkalinity (Etchanchu and Probst, 1988; Jiang, 2013; Jiang et al., 2009; Perrin et
666	al., 2008; Semhi and Suchet, 2000). In these studies, the nitrification described as Eq.
667	(8) is usually considered as the unique origin of NO3. According to the result of
668	NaNO3- treatment in this study, the contribution of protons from nitrification to
669	carbonate weathering may be overestimated if anthropogenic NO3-is neglected, since
670	the anthropogenic NO ₃ ⁻ does not release the proton described as Eq. (8). For NH_4NO_3
671	fertilizer, the (Eq. (12)) show that the two moles of Ca ²⁺ +Mg ²⁺ , NO ₃ ⁻ and HCO ₃ ⁻ will
672	be produced when one mole NH_4NO_3 react with 2 moles of carbonate, where only
673	half of NO_3^- originate from nitrification described as Eq. (8). This will result in
674	doubled overestimation on the true contribution of the nitrification to CO2
675	consumption by carbonate weathering.
676	At regional scales, If different fertilizers are added to an agricultural area, the
677	estimation of CO ₂ consumption by carbonate weathering might became more
678	complicated, since the mole ratio of Ca+Mg, HCO3 ⁻ and/or NO3 ⁻ between different
679	fertilization treatment is different (see Eq. (8) (12)). Thus, the related anthropogenic
680	inputs (e.g. Ca+Mg, NH ₄ , NO ₃ ⁻ , HCO ₃ ⁻ , etc.) need to be investigated to more
681	accurately estimate the impact of fertilization on carbonate weathering and its CO2
682	consumption. Moreover, the statement that nitrogenous fertilizer can aid carbonate
683	weathering may result in misunderstanding more or less, it should not be nitrogenous
684	fertilizer but, rather, ammonium fertilizer.
685	5. Conclusion
686	The impact of the addition of different fertilizer (NH4NO3, NH4HCO3, NaNO3,
687	NH ₄ Cl, (NH ₄) ₂ CO ₃ , Ca ₃ (PO ₄) ₂ , (NH ₄) ₃ PO ₄ , Ca Mg P, Urea and K ₂ CO ₃) on carbonate
688	weathering was studied in a field column experiment with carbonate rock tablets at its 30

689	bottom of each. The weathering amount and ratio of carbonate rock tablets showed
690	that the addition of urea, NH_4NO_3 , NH_4HCO_3 , NH_4CI and $(NH_4)_2CO_3$ distinctly
691	increased carbonate weathering, which was attributed to the nitrification of $\mathbf{NH_4}^+$, and
692	the addition of $Ca_3(PO_4)_2$, Ca Mg P and K_2CO_3 -induced carbonate precipitation due
693	to common ion effect. While the $(NH_4)_3PO_4$ and $NaNO_3$ -addition did not impact
694	significantly on carbonate weathering, where the former can be attributed to low
695	added amount of (NH4) ₃ PO ₄ , may be related to the inhibition of phosphate, and the
696	latter seemed to be raising a new question. The little impact of nitrate on carbonate
697	weathering may result in the overestimation of impact of N-fertilizer on CO2
698	consumption by carbonate weathering at the regional/global scale if the effect of NO3
699	and NH4 are not distinguished. Thus, the related anthropogenic inputs (e.g. Ca+ Mg,
700	NH ₄ , NO ₃ ⁻ , HCO ₃ ⁻ , etc.) need to be investigated to more accurately estimate the
701	impact of fertilization on carbonate weathering and its CO2 consumption. Moreover,
702	in order to avoid misunderstanding more or less, the statement that nitrogenous
703	fertilizer can aid carbonate weathering should be replaced by ammonium fertilizer.
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1153			
1154	Table 1 Chemica	al composition of	soil
	Parameter	Unit	Values
	pH	-	6.94
	Content of particles (<0.01mm)	<u>%</u>	<u>74</u>
	Content of particles (<0.001mm)	<u>%</u>	<u>45</u>
	Organic matter	%	0.99
	$\mathbf{NH_{4}^{+}}$ -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

带格式表格

1157

Table 2 Carbonate weathering under different fertilizer treatments

	Limestone		Dolostone	_		
Treatment	<u>Acw Rw / ‰</u> g	Raw /	<u>Acw-Rw / ‰g</u>	Raw /		带格式表格
Treatment		<u>g m⁻² a⁻¹ R<i>cw</i> /‰</u>		<u>g m⁻² a⁻¹ Rew</u>		
				/%		
Control	0. 0014<u>48</u>_±	<u>2.00 ±0.58a0.48</u>	-0. 0011<u>31</u>±	<u>-1.57 ±</u>		【 带格式的: 字体: Times New Roman】
	<u>0.14a</u>		<u>0.09a</u>	<u>0.86a 0.31</u>		【 带格式的: 字体: Times New Roman 】
NH ₄ NO ₃	<u>0.01746.42 _±</u>	<u>24.86 ± 2.01</u>	<u>0.01445.30±</u>	<u>20.57 ±</u>	\frown	(带格式的: 字体: Times New Roman
	<u>0.28e</u>	6.42	<u>0.87e</u>	<u>1.15b5.30</u>		带格式的: 字体: Times New Roman
NH ₄ HCO ₃	<u>0.01474.44 .</u> ±	<u>21.00 ± 3.45</u>	0.0096 <u>3.22±</u>	<u>13.71 ±</u>		(带格式的: 字体: Times New Roman)
	<u>0.81b</u>	4.44	0.87 b	<u>3.88b3.22</u>		(带格式的: 字体: Times New Roman
NaNO ₃	0.0031 0.86 <u>±</u>	4.43 ±1.73 a0.86	0. 0022 53 <u>+</u>	<u>3.14 ±</u>		(带格式的: 字体: Times New Roman
	0.17 a		0.26 a	1.73 a0.53		(带格式的: 字体: Times New Roman) (带格式的: 字体: Times New Roman)
NH₄Cl	0.0149<u>5.54</u>_±	21.29 ±2.45 b	0.0131 4.77 ±	18.71 ±0.86 b		带格式的: 子体: Times New Roman 带格式的: 字体: Times New Roman
	0.64 be	5.54	0.78 be	4.77		带格式的: 字体: Times New Roman 带格式的: 字体: Times New Roman
$(NH_4)_2CO_3$	0.0144 4.48 .±	20.57 ±4.46 b	0.0186 4.94 <u>±</u>	26.57 ±		带格式的: 字体: Times New Roman 带格式的: 字体: Times New Roman
(),2 5	0.95 bc	4.84	1.91 be	7.62 b4.94	7/////	带格式的: 字体: Times New Roman
$Ca_3(PO_4)_2$	0.00030.01 ±	$0.43 \pm 0.86a0.01$	-0. 0013 55 <u>±</u>	-1.86 ±		带格式的: 字体: Times New Roman
5(+)2	0.04 a		0.25 a	1.29 a 0.55	<u> </u>	带格式的: 字体: Times New Roman
$(NH_4)_3PO_4$	0.0028 <u>1.08</u> _±	4.00 ±1.15 a1.08	0.0007 0.75 ±	1.00 ±		
· • · · · ·	0.34 a		0.21 a	1.01 a0.75	<u>אוווור</u>	
Ca-Mg-P	-0.0013-0.31 ,±	-1.86 ±0.43 a 0.31	-0. 0022 97 ,±	-3.14 ±	71/////	【 带格式的: 字体: Times New Roman】
8	0.12 a		0.38 a	0.72 a-0.97	<u>ווווור</u>	【 带格式的: 字体: Times New Roman 】
Urea	0.0243 <u>8.48</u> _±	<u>34.71 ±4.32</u> e	0.01856.59 ±	26.43 ±	<u>ווווור</u>	(带格式的: 字体: Times New Roman
	0.96 d	8.48	0.67 d	2.73 c6.59		(带格式的: 字体: Times New Roman
K ₂ CO ₃	-0. 0008 26 <u>.</u> ±	$-1.14 \pm 0.58a - 0.26$	-0. 0018 59 .±	-2.57 ±		(带格式的: 字体: Times New Roman)
H ₂ 003	<u>0.15a</u>	<u></u>	0.15 a	<u>0.43a-0.59</u>	ווווווור	带格式的: 字体: Times New Roman
A cw the amount	-	ering: Rew - the ratio		ring; Raew - the rate of		(帯格式的: 字体: Times New Roman)
		6.		Raew = (Wi-Wf)/(S*T),		(带格式的: 字体: Times New Roman) (带格式的: 字体: Times New Roman)
	-	-	-	l weight. S is the surface	18111111	带格式的: 字体: Times New Roman 带格式的: 字体: Times New Roman
	-		-	-		带格式的: 字体: Times New Roman 带格式的: 字体: Times New Roman
tablets), and T is the experiment period. Values are reported as means \pm standard deviations, n = 3. Values in each column followed by different letters are significantly (p < 0.05) different based on #格式的: 字体: Times New Roman						
one-way ANOVA		mer ent tetters are sig t	meaning (p <0.05) (merent based on		带格式的: 字体: Times New Roman
one-way Mixe V/Y	<u> </u>					带格式的: 字体: Times New Roman
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1169		he main reaction and effects in fertilized treatments, and the potential nitrogenous	一件	時格式的: 左 则: 3.17 厘	侧: 3.17 厘米 米,顶端: 2.5	,右 4 厘
1170	transform	》 一 闭	长, 底端: 2 〒米 高度・	米,顶端: 2.5 54 厘米,宽度 29.7 厘米	: 21	
	Treatment	Main reactions and effects	$\sim >$	<u></u> 時格式的:下		
				時格式的: 上		
	1.Control	$\underline{Ce_{(1-x)}Mg_{x}CO_{2}} + \underline{H_{2}O} \xrightarrow{\rightarrow} (1-x)Ce^{2+} + xMg^{2+} + 2HCO_{2}^{2-}$		萨格式表格		<u>_</u>
	<u>2.NH₄NO₃</u>	$\frac{2Ce_{4+x_2}Mg_{x_2}CO_{2}+NH_{4}NO_{2}+2O_{2}-2(1-x)Ce^{2+}+2xMg^{2+}+2NO_{2}-H_{2}O+2HCO_{2}}{2}$		持格式的: 居	中	
	<u>3. H₄HCO₃</u>	$\frac{2C_{a_{1}+b_{2}}Mg_{2}CO_{2}+NH_{2}HCO_{2}+2O_{2}}{2} \rightarrow 2(1-x)Ca^{2+}+2xMg^{2+}+NO_{2}+H_{2}O+3HCO_{2}=$	4	持格式的: 居	中	
	4. NaNO ₃	$\underline{Ca_{(1-x)}Mg_xCQ_2 + NaNQ_2 + CQ_2 + H_2Q \rightarrow (1-x)Ca^{2+} + xMg^{2+} + Na^+ + NQ_2^- + 2HCQ_2^-}$		持格式的: 居		
	5. NH ₄ Cl	$\frac{2Ca_{1+x}Mg_{x}CO_{2} + NH_{2}CI + 2O_{2} \rightarrow 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{2}^{-} + CI^{-} + H_{2}O + 2HCO_{2}^{-}}{2}$	\sim	持格式的 :居		
	<u>6. (NH</u> 4 <u>2</u> 2 <u>CO</u> 3	$\frac{3Ca_{1+x_2}Mg_{a_2}CO_{2} + (NH_{a_1})_{2}CO_{2} + 4O_{2}}{\rightarrow} 3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{2}^{-} + 2H_{2}O + 4HCO_{2}^{-}$		持格式的 :居		
		(1) Common ion effect: The Ca _{11-x2} Mg _x CO ₃ produces when the concentrations of Ca ²⁺ and Mg ²⁺ increase	<u>ses</u> ा	時格式的: 居	Ψ 	
	$7. Ca_3(PO_4)_2$	$\frac{(1-x)\operatorname{Ca}^{2+} + x\operatorname{Mg}^{2+} + 2\operatorname{HCO}_2}{\longrightarrow} \operatorname{Ca}_{(1-x)}\operatorname{Mg}_x\operatorname{CO}_2 + \operatorname{CO}_2 + \operatorname{H}_2\operatorname{O}} $	州		中	,
		(2) Inhibition of phosphate to calcite precipitation: calcium phosphate precipitation produces on the sur	face o	±		
		calcite after the addition of PO ₄ ³⁻ in soil, resulting in inhibiting the precipitation of calcite				
		$\frac{(1) 2Ca_{1+2}Mg_{x}CO_{2} + NH_{4}^{+} + 2O_{2} \rightarrow 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{2}^{-} + H_{2}O + 2HCO_{2}^{-}}{2}$				
	8. (NH ₄) ₃ PO ₄	(2) Inhibition of phosphate to calcite dissolution: calcium orthophosphate (Ca P) precipitation produces	<u>s on</u> '''	节格式的: 居	円	!
		surface of calcite after the addition of PO ₄ ³ -in soil, resulting in inhibiting the dissolution of calcite				
		(1) Common ion effect: The $Ca_{1+x_2}Mg_xCO_3$ produces when the concentrations of Ca^{2+} and Mg^{2+} interest	ses 竹 オ	脊格式的 : 缩 <	进: 首行缩进:	0 厘
	9. Ca Mg P	$\frac{(1-x)\operatorname{Ca}^{2+} + x\operatorname{Mg}^{2+} + 2\operatorname{HCO}_2}{(1-x)\operatorname{Ca}_2 + 2\operatorname{HCO}_2} \xrightarrow{\sim} \operatorname{Ca}_{(1-x)}\operatorname{Mg}_2\operatorname{CO}_2 + \operatorname{CO}_2 + \operatorname{H}_2\operatorname{O}_2} \xrightarrow{\leftarrow} \operatorname{Ca}_{(1-x)}\operatorname{Mg}_2\operatorname{CO}_2 + \operatorname{Co}_2 + \operatorname{H}_2\operatorname{O}_2$	梢		中,缩进:首行	缩进:
		(2) Inhibition of phosphate to calcite precipitation: calcium phosphate precipitation produces on the sur	fac	0 厘米		
		calcite after the addition of P in soil, resulting in inhibiting the precipitation of calcite				
	<u>10. Urea</u>	$\frac{3C_{8,1}}{2C_{8,1}}Mg_{2}CO_{3} + CO(NH_{2})_{2} + 4O_{2} \rightarrow 3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{3}^{-} + 4HCO_{2}^{-}$	Ħ	节格式的: 居	甲	
		<u>Common ion effect: The Ca_(1-x)Mg_xCO₃ produces when the concentration of HCO₃ increases</u>	(-+			
	<u>11. K₂CO3</u>	$(\underline{i}) (\underline{l} \cdot \underline{x}) Ca^{2+} + \underline{x}Mg^{2+} + \underline{2HCO}_2 \xrightarrow{\rightarrow} Ca_{(\underline{l} \cdot \underline{x})}Mg_{\underline{x}}CO_2 + \underline{CO}_2 + \underline{H}_2O$	1	节格式的: 居	中 	<u></u>
		$(\underline{ii}) \underline{K_2CO_2 + H_2O \rightarrow 2K^+ + HCO_2} + OH^-$				
1171		Cable 3: The main reaction and effects in these 11 fertilizeder treatments				
	Treatment	Main reactions and effects				
	<u>1. Control</u>	$\underline{\operatorname{Ca}_{(1-x)}\operatorname{Mg}_{x}\operatorname{CO}_{3} + \operatorname{CO}_{2} + \operatorname{H}_{2}\operatorname{O} \rightarrow (1-x)\operatorname{Ca}^{2+} + x\operatorname{Mg}^{2+} + 2\operatorname{HCO}_{3}^{2-}$				
	<u>2. NH₄NO₃</u>	$\underline{2Ca_{(1-x)}Mg_xCO_3 + NH_4NO_3 + 2O_2} \rightarrow \underline{2(1-x)Ca^{2+} + 2xMg^{2+} + 2NO_3 + H_2O + 2HCO_3}$				
	<u>3. NH₄HCO₃</u>	$\underline{\mathrm{NH}_4\mathrm{HCO}_3} \mathrm{NH}_3\uparrow + \mathrm{H}_2\mathrm{O} + \mathrm{CO}_2\uparrow$				
		$\underline{2Ca_{(1-x)}Mg_xCO_3 + NH_4HCO_3 + 2O_2} \rightarrow \underline{2(1-x)Ca^{2+} + 2xMg^{2+} + NO_3 + H_2O + 3HCO_3}$				
	<u>4. NaNO₃</u>	$\underline{Ca_{(1-x)}Mg_{3}CO_{3} + NaNO_{3} + CO_{2} + H_{2}O \rightarrow (1-x)Ca^{2+} + xMg^{2+} + Na^{+} + NO_{3} + 2HCO_{3}}{}^{2}$				
	<u>5. NH₄Cl</u>	$\underline{2Ca_{(1-x)}Mg_xCO_3 + NH_4Cl + 2O_2} \rightarrow \underline{2(1-x)Ca^{2+} + 2xMg^{2+} + NO_3 + Cl^+ + H_2O + 2HCO_2}$				
	<u>6. (NH₄)₂CO₃</u>	$(\mathrm{NH}_4)_2\mathrm{CO}_3 \rightarrow 2\mathrm{NH}_3 \uparrow + \mathrm{H}_2\mathrm{O} + \mathrm{CO}_2 \uparrow$				
		$\frac{3Ca_{(1-x)}Mg_{x}CO_{3} + (NH_{4})_{2}CO_{3} + 4O_{2} \rightarrow 3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{3} + 2H_{2}O + 4HCO_{3}}{3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{3} + 2H_{2}O + 4HCO_{3}}$	_	1. 1 A. 15 1 A. 10	11 I.M	
	<u>7. $Ca_3(PO_4)_2$</u>	$(1) (1-x) Ca^{2+} + xMg^{2+} + 2HCO_3 \rightarrow Ca_{(1-x)}Mg_3CO_3 + CO_2 + H_2O$	骨長	5格式的: 缩 悬挂缩进: 2	进:左侧: 0 厚 字符,首行缩进	‼米, : −2
		$(2) Ca + PO_{\underline{4}} \rightarrow Ca - P$	ļ	Z符		
	<u>8. (NH₄)₃PO₄</u>	$(1) 2Ca_{(1-x)}Mg_{x}CO_{3} + NH_{4}^{+} + 2O_{2} \rightarrow 2(1-x)Ca^{2+} + 2xMg^{2+} + NO_{3}^{-} + H_{2}O + 2HCO_{3}^{-}$				
		$(2) Ca + PO_{\underline{A}} \rightarrow Ca - P$				
	<u>9. Ca-Mg-P</u>	$(1) (1-x) Ca^{2+} + xMg^{2+} + 2HCO_3 \rightarrow Ca_{(1-x)}Mg_xCO_3 + CO_2 + H_2O$				
		$(2) Ca + PO_{\underline{4}} \rightarrow Ca - P$				
	<u>10. Urea</u>	$\underline{3Ca_{(1-x)}Mg_{x}CO_{3} + CO(NH_{2})_{2} + 4O_{2}} \rightarrow 3(1-x)Ca^{2+} + 3xMg^{2+} + 2NO_{3} + 4HCO_{3}$				
	<u>11. K₂CO₃</u>	<u>(i) (1-x) $Ca^{2+} + xMg^{2+} + 2HCO_3 \rightarrow Ca_{(1-x)}Mg_3CO_3 + CO_2 + H_2O_3$</u>				

	$(\underline{ii}) \underline{K_2CO_3} + \underline{H_2O} \rightarrow 2K^+ + \underline{HCO_3} + \underline{OH^-}$		带格式的:	缩进:	首行缩进:	0.5
1172	Note: (1) Common ion effect: The $Ca_{(1-x)}Mg_xCO_3$ produceds when the concentrations of Ca^{2+} , $-Mg^{2+}$ and/or		带格式的:	症进.	苦仁症进,	0 2
1173	<u>HCO₃</u> increases (for T treatment 7, 9 and 11): (1-x) Ca ²⁺ + xMg ²⁺ + 2HCO ₃ \rightarrow Ca _(1-x) Mg ₃ CO ₃ + CO ₂ + H ₂ O;		符 符	细灯.	自1] 湘灯,	0 手
1174	(2) Inhibition of phosphate to calcite dissolution/precipitation by phosphate: calcium orthophosphate (Ca-P)		带格式的:	下标		
1175	precipitation produceds on the surface of calcite after the addition of PO_4^{3-} in soil, resulting in the inhibiting on of	$\langle \rangle$	带格式的:	上标		
1176	the dissolution/precipitation of calcite (for \pm treatment 7, 8 and 9): Ca + PO ₄ \rightarrow Ca-P		带格式的:	左		
I						

1177 <u>Table 4: – The amount of fertilizer-derived NH_4^+ at the initial phase of the experiment and the</u>

	Molecular	Amount of	Molar	Amount of	The maximum	
Treatment	mass	added	<u>amount</u>	fertilizer-derived	of N products	
	<u>g/mol</u>	<u>fertilizer /g</u>	/mole	$\underline{NH_4}^+/mole$	/mole	
<u>NH₄NO₃</u>	<u>80</u>	<u>43</u>	<u>0.54</u>	<u>0.54</u>	1.08	
<u>NH₄HCO₃</u>	<u>79</u>	<u>85</u>	<u>1.08</u>	<u>1.08</u>	1.08	
<u>NaNO₃</u>	<u>85</u>	<u>91</u>	<u>1.07</u>	<u>0.00</u>	1.07	
<u>NH₄C1</u>	<u>53.5</u>	<u>57</u>	<u>1.07</u>	<u>1.07</u>	<u>1.07</u>	
<u>(NH₄)₂CO₃</u>	<u>96</u>	<u>51</u>	<u>0.53</u>	<u>1.06</u>	<u>1.06</u>	
<u>Ca₃(PO₄)₂</u>	<u>310</u>	<u>52</u>	<u>0.17</u>	<u>0.00</u>	0.00	
<u>(NH₄)₃PO₄</u>	<u>149</u>	<u>15</u>	<u>0.10</u>	<u>0.30</u>	<u>0.30</u>	
<u>Ca-Mg-P</u>	<u>wnd</u>	<u>44</u>	<u>wnd</u>	<u>0.00</u>	0.00	
<u>Urea</u>	<u>60</u>	<u>32</u>	<u>0.53</u>	<u>1.06</u>	1.06	
<u>K₂CO₃</u>	<u>138</u>	<u>10</u>	<u>0.07</u>	<u>0.00</u>	0.00	
wnd=withoutno data; The amount of added fertilizer (g) divided by its molecular mass (g/mol)						

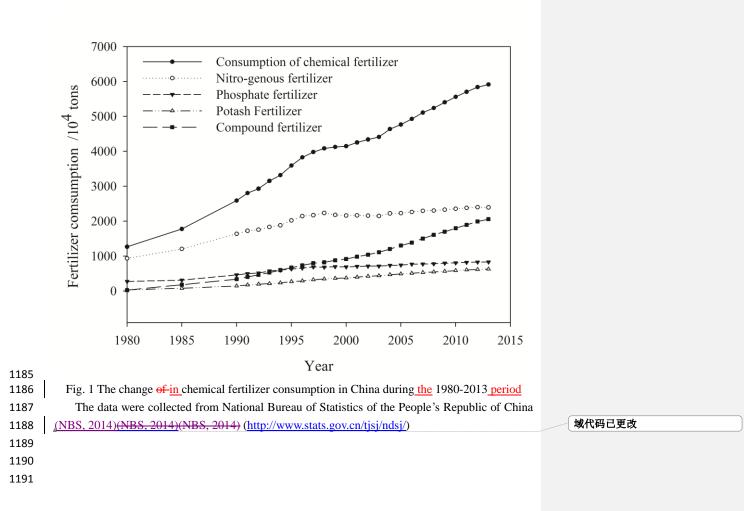
1180 wais the molar amount of fertilizer (mole); Gadf-gram amount of added fertilizers (g); Maafof
 1181 added fertilizers (mol). The amounts of fertilizer-derived NH₄⁺ is calculated by their own

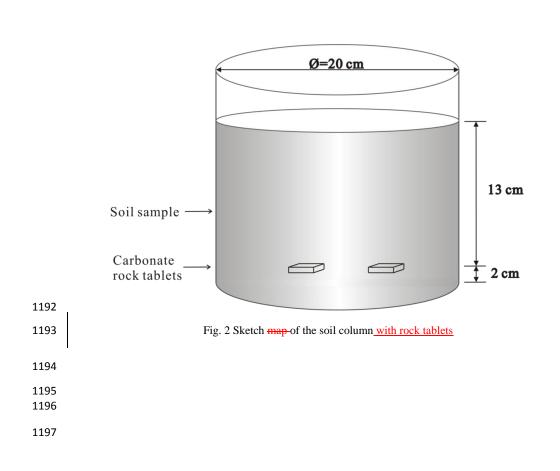
1181 added fertilizers (mol). The amounts of fertilizer-derived NH₄⁺ is calculated by their own
1182 ionization or hydrolysis processes. The maximum of N products is estimated by their main

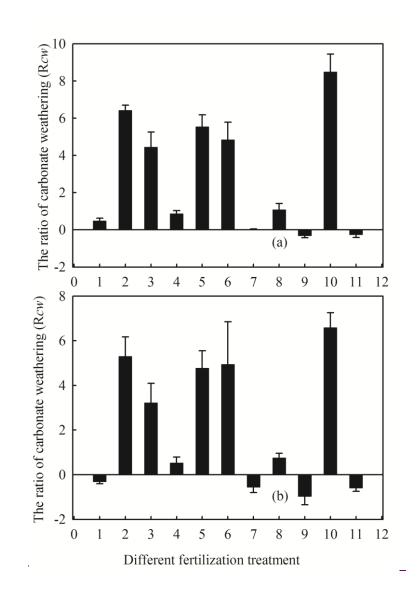
1183 <u>reactions in **t**</u><u>Table 3.</u>

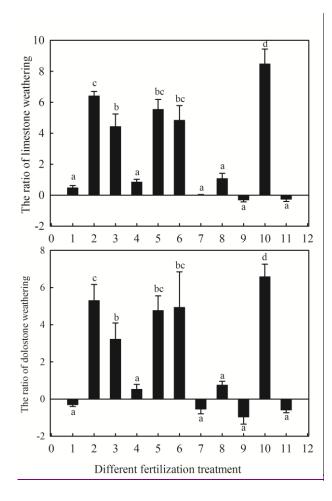
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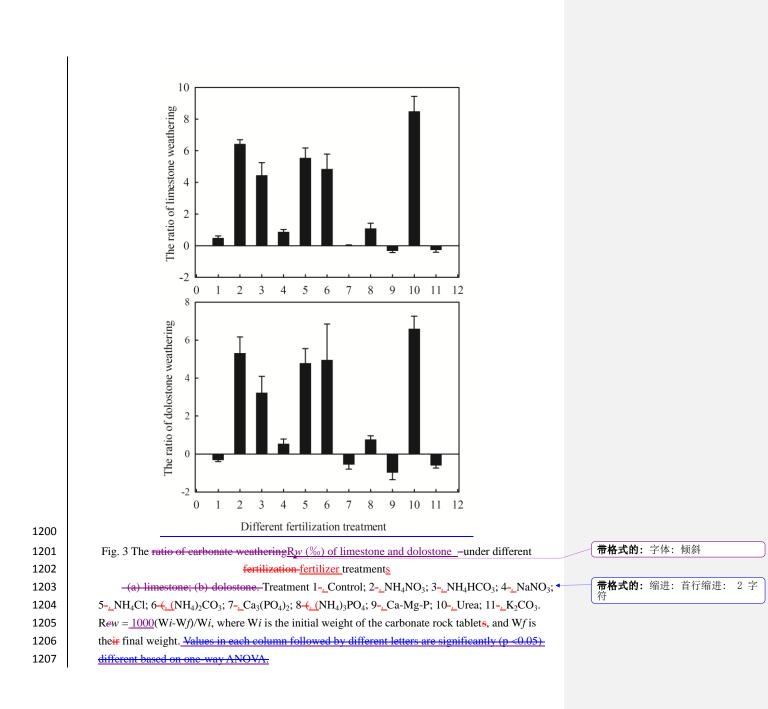
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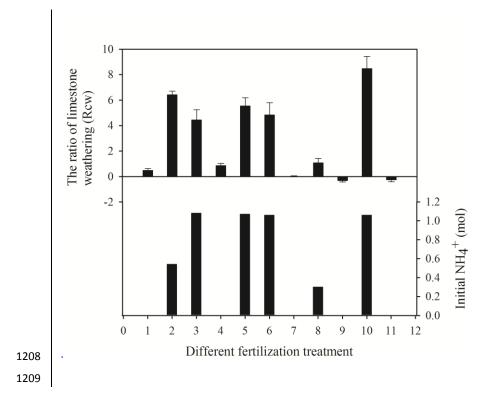




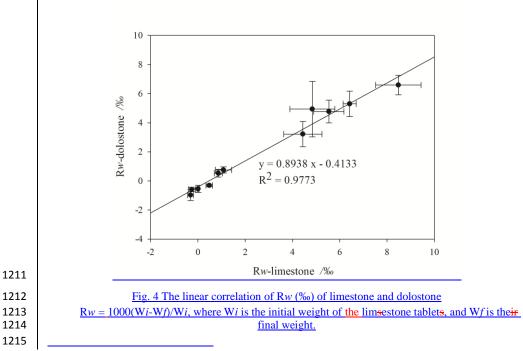


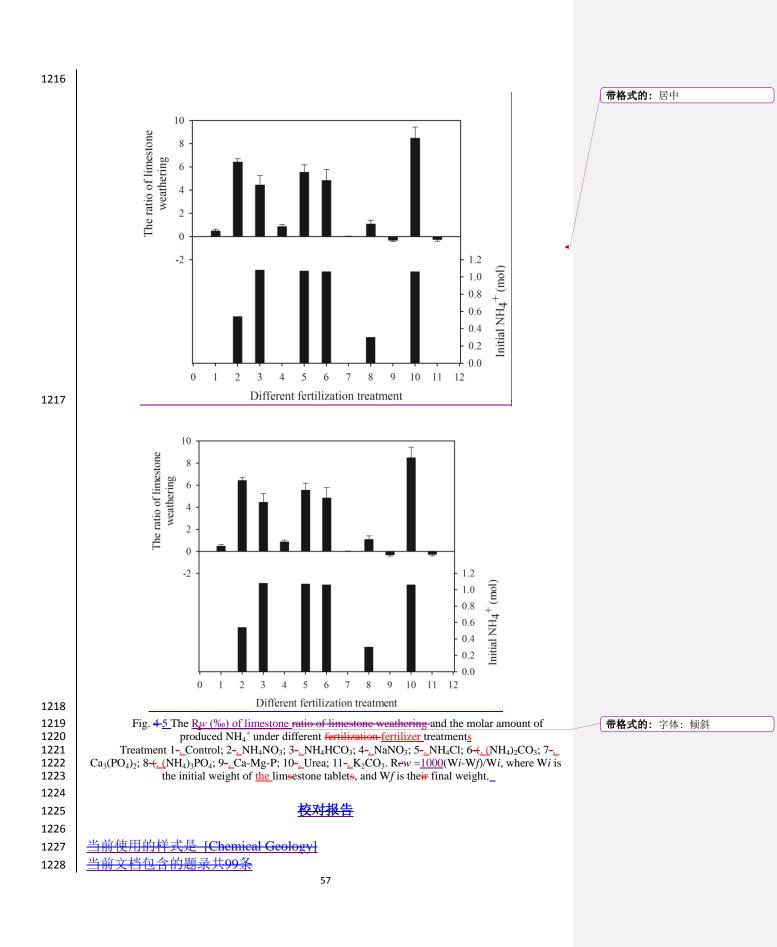






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<u>有0条题录存在必填字段内容缺失的问题</u> 所有题录的数据正常