

## Interacting controls on innate sources of CO<sub>2</sub> efflux from a calcareous arid zone soil under experimental acidification and wetting



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### ABSTRACT

More than half of global soil carbon is stored as carbonates, primarily in arid and semi-arid zones. Climate change models predict more frequent and severe rainfall events in some parts of the globe, many of which are dominated by calcareous soils. Such events trigger substantial increases in soil CO<sub>2</sub> efflux. We hypothesised that the primary source of CO<sub>2</sub> emissions from calcareous, arid zone soil during a single wetting event is abiotic and that soil acidification and wetting have a positive, potentially interacting, effect. We manipulated soil pH, soil moisture, and controlled soil respiration by gamma irradiating half of an 11 day incubation experiment. All manipulated experimental treatments had a rapid and enormous effect on CO<sub>2</sub> emission. Respiration contributed ca. 5% of total CO<sub>2</sub> efflux; the major source (carbonate buffering) varied depending on the extent of acidification and wetting. Maximum CO<sub>2</sub> efflux occurred when pH was lowest and at intermediate matric potential. CO<sub>2</sub> efflux was lowest at native pH when soil was air dry. Our data suggest that there may be an underestimate of soil-atmosphere carbon fluxes in arid ecosystems with calcareous soils. There is also a clear potential that these soils may become net carbon sources depending on changes in rainfall patterns, rainfall acidity, and future land management. Our findings have major implications for carbon cycling in arid zone soil and further study of carbon dynamics in these terrestrial systems at a landscape level will be required if we are to improve global climate and carbon cycling models.

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### 1. Introduction

The global soil carbon pool is a substantial component of the Earth's carbon cycle. Understanding the dynamics of soil-atmosphere CO<sub>2</sub> exchange is important if we are to make sound predictions about the future impacts of human activity on atmospheric CO<sub>2</sub> concentrations and climate change (Eswaran et al. 2000; Serrano-Ortiz et al. 2010). The size of the terrestrial soil carbon pool to a depth of 1 m has been estimated at about 1500 Pg (Pg = 10<sup>15</sup> g) (Batjes, 1996; Eswaran et al. 2000). Of this pool about 900 Pg is stored as inorganic soil carbonates (Batjes, 1996; Eswaran et al. 2000; Ramnarine et al. 2012; Schlesinger, 1997). Thus the surficial inorganic soil carbonate pool is larger than the

atmospheric pool of 750 Pg C, and is also greater than all CO<sub>2</sub> emissions released from fossil fuels and cement since 1850, which is only about 350 Pg C (Canadell et al. 2007; Lal, 2009). Release of this pool, or even a fraction of it, into the atmosphere could have significant implications for climate change.

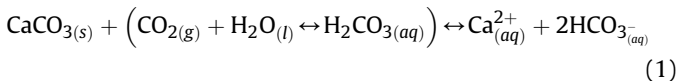
The global soil inorganic carbon (SIC) pool is located primarily in arid and semi-arid regions, occupying a total land area of approximately 4.9 × 10<sup>7</sup> km<sup>2</sup> (Eswaran et al. 2000; Dart et al. 2007). In Australia, these soils occupy about 21% of the total continental landmass or about 1.6 × 10<sup>6</sup> km<sup>2</sup> and much of this area contains calcareous soils associated with karst systems (Dart et al. 2007). In these systems, soil-atmosphere CO<sub>2</sub> flux is driven by factors that affect plant and microbial respiration and also those that influence the dissolution equilibrium of soil carbonates: soil pH, soil moisture, the partial pressure of soil CO<sub>2</sub> (pCO<sub>2</sub>), and temperature (Lal and Kimble, 2000; Stevenson and Verburg, 2006).

In arid and semi-arid environments where agricultural land use is limited but calcareous soils are common, soil moisture is the principle driver of soil CO<sub>2</sub> flux (Inglisma et al. 2009; Zhang et al.

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2010). These ecosystems usually undergo extended periods of aridity that is punctuated by seasonal or sporadic rainfall (Eamus et al. 2013). Rainfall events in these ecosystems are known to trigger sudden and substantial increases in soil CO<sub>2</sub> efflux; initially through gas displacement followed by increased autotrophic and heterotrophic respiration (Inglima et al. 2009; Nobel and Palta, 1989; Tibbett et al. 2011; Zhang et al. 2010), but abiotic sources can also contribute significantly (Ramnarine et al. 2012; Stevenson and Verburg, 2006). The pH of calcareous soils is normally >7 and carbonate dissolution in these soils under wet conditions results in the consumption of CO<sub>2</sub> and the formation of HCO<sub>3</sub><sup>-</sup> and Ca<sup>2+</sup> ions (Eq. (1); for simplicity we show the reaction for calcite only).

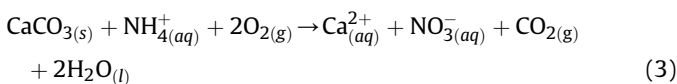


However, secondary carbonate precipitation can occur in these soils at the onset of rainfall (Eq. (2)) provided that previous rainfall events were insufficient to leach dissolved HCO<sub>3</sub><sup>-</sup> and Ca<sup>2+</sup> ions and inter-rainfall drought subdues soil respiration such that soil pCO<sub>2</sub> is sufficiently low (Gandois et al. 2011).



Rainfall events that trigger CO<sub>2</sub> efflux from soils in these landscapes are likely to become more frequent and severe as anthropogenic greenhouse gas emissions continue, particularly in high latitudes, northern mid latitudes, and in tropical/sub-tropical regions (IPCC, 2014; Fischer and Knutti, 2015). This rainfall is also likely to become more acidic as the atmospheric concentration of CO<sub>2</sub> increases (Bogan et al. 2009).

Change in rainfall patterns in some parts of the globe may unlock previously deserted land to agricultural use or forestry, or intensify such land use in previously marginal areas. This change in land use has the potential to acidify soils through addition of nitrogenous fertilisers, proton release and cation extraction by plant roots, and grazing and vegetation harvest (Neumann and Römheld, 2012; Rigueiro-Rodriguez et al. 2012; Sanderman, 2012; Gandois et al. 2011; Tamir et al. 2011; West and McBride, 2005). For example, nitrification of fertiliser applied NH<sub>4</sub><sup>+</sup> could reduce soil pH in microsites across a calcareous landscape to a point where carbonate dissolution results in CO<sub>2</sub> efflux (Gandois et al. 2011):



Currently, calcareous soils in arid regions are seen as a net CO<sub>2</sub> sink (Lal and Kimble, 2000; Serrano-Ortiz et al. 2010), however; it is possible that, as atmospheric CO<sub>2</sub> increases and projected effects of climate change ensue, these soils may, to some degree, become a source of atmospheric CO<sub>2</sub> due to pH reduction and increased soil wetting. Whilst much is known of the chemistry of carbonate dissolution and precipitation under various conditions, the impact of interactions between soil acidification and moisture on CO<sub>2</sub> flux in calcareous soils is not well understood. To identify the innate sources of CO<sub>2</sub> efflux in this soil type and to quantify soil CO<sub>2</sub> efflux from biotic and abiotic sources under different moisture and pH conditions, we tested the hypotheses that the primary source of CO<sub>2</sub> emissions from a calcareous soil from an arid region are of abiotic origin and that soil acidification and wetting have a positive, and potentially interacting, effect on these emissions. To achieve this we used a controlled incubation approach where pH, moisture and soil biology were manipulated experimentally.

## 2. Methods

### 2.1. Site description

The sampling site was located roughly in the centre of Barrow Island (lat. 20° 46' 08" S, long. 115° 24' 05" E, 33 m above sea level). The sampling site covered an area of about 4 m<sup>2</sup> and was devoid of vegetation. The landscape surrounding the site features undulating karstic limestone hills separated by wide, flat valleys with ephemeral creek beds. The vegetation is predominated by hummock grassland composed mostly of spinifex (*Triodia* R.Br. species) interspersed with low acacia shrubs. The soils in the locality of the sampling site may be described as red calcareous leptic rudosols (Isbell, 2002). The sampled soil was devoid of gravel and had a single grained structure with weak aggregation.

The island experiences an arid-subtropical climate. Mean maximum temperature over summer months (December–February) exceeds 32 °C, moderating to around 25 °C during winter (June–August). Annual mean temperature is 25.9 °C and mean annual rainfall is 306 mm. The majority of rainfall is delivered by the three to four summer cyclones that occur most years (BOM, 2013). The current climate trend indicates a moderate increase in annual precipitation on the Island of around 5 mm per decade (BOM, 2015); however, the intensity and frequency of summer cyclones is likely to increase according to IPCC climate model projections (IPCC, 2014).

### 2.2. Soil sampling and characterisation

At the sample site, a core sample of topsoil was taken by driving a brass ring of 7 cm height and 7.3 cm diameter into the soil and carefully lifting out the soil contained within it. The soil within the ring was then transferred to a polyethylene bag and examined to ensure that no soil macro-fauna had been collected with the sample. After transportation to the laboratory, the sample was oven dried to constant weight at 105 °C for determination of soil water content. Soil dry bulk density was calculated as the ratio of dry soil mass to total volume including pores (Cresswell and Hamilton, 2002).

Bulk soil for the incubation experiment was sampled through careful excavation with a trowel to a depth not exceeding 10 cm. Soil was placed into a large polyethylene bag and transferred to a cooler box for transportation to the laboratory where it was stored in a cool room at 4 °C. The humidity within the bulk sample was maintained with a moistened paper towel wick. Prior to subsampling for the experiment, this soil was tumbled within the bag for 10 min to ensure it was well mixed.

Soil pH and electrical conductivity was measured on air dried and sieved material. Electrical conductivity and pH measurements were made on a 1:5 soil:water extract, and pH was also measured on a 1:5 soil:CaCl<sub>2</sub> (0.01 M) extract according to the methods described in Rayment and Lyons (2010). The pH measurements reported here are those taken in the calcium chloride extract. Particle size distribution was determined using the pipette method (Klute, 1986). The texture of the soil was determined by comparing the percentages of sand, silt and clay in the material against a standard texture diagram (McDonald et al. 1990). Soil organic carbon (SOC) and carbonate fractions were measured according to the methods of Walkely and Black (1934) and Horváth et al. (2005) respectively. Total, ammonium and nitrate forms of mineral N were measured simultaneously from extracts of 1:5 soil: 1 M KCl solution using a Flow Injection Analyser (Lechat Instruments, USA). Ammonium nitrogen was measured colorimetrically at 420 nm using the indo-phenol blue reaction (Searle, 1984). Nitrate was reduced to nitrite through a copperised-cadmium column and

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