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Carbonatites as rock fertilizers: a review

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Abstract:

Rock fertilizers are geological resources used in agriculture for their nutrient content, but slow weathering rates hinder their effectiveness. Carbonatites are igneous rocks made mostly of carbonate minerals with a relatively high weathering rate and often containing nutrient-bearing accessory minerals (e.g., apatite and biotite). Despite evidence supporting their potential as rock fertilizers, a comprehensive review of such data is missing in the literature. Furthermore, when studies on agricultural uses of carbonatites exist, they typically center on applied research aspects (i.e., “does it work?”) rather than on basic research aspects (i.e., “how does it work?”). Here we evaluate the applicability of carbonatites in agriculture taking into consideration the factors that affect mineral weathering and plant nutrient uptake. While there is sufficient data to conclude that carbonatites can be a source of many plant nutrients, their effectiveness depends on the interactions of many components (e.g., soils, plants, microorganisms). To develop best management practices around carbonatites used in agriculture, it is essential to understand these interactions.

Key words: sustainable agriculture, agrogeology, plant-mineral interactions, microbe-mineral interactions, mineralosphere

1 **Introduction**

2 Sustainability is recognized as one of the largest challenges facing agriculture today. Climate
3 change (Campbell et al. 2016; Altieri and Nicholls 2017), a growing global population (Alexandratos
4 2005), a continued reliance on non-renewable resources (e.g., high-grade phosphate rock resources;
5 Cordell et al. 2009), and a depletion of nutrients in agricultural systems (van Straaten 2007), all contribute
6 to make sustainability of paramount importance. In this context, improved farm management practices are
7 critical to prevent soil degradation, soil erosion (Matson 1997; Tilman et al. 2002), and eutrophication of
8 connected water bodies from highly-soluble nutrient runoff (Diaz and Rosenberg 2008; Savard et al.
9 2010).

10 In an attempt to make agriculture more sustainable, the use of chemically-unprocessed geological
11 resources to improve soil health and fertility has been the focus of increased investigation (Zhang et al.
12 2018). Crop nutrients can originate from many sources with varying degrees of solubility (Figure 1). As
13 such, geological resources used in agriculture can be broadly split into two categories, i.e., rock fertilizers
14 and agrominerals depending on the base material (Figure 1). In this review, these two terms will be used
15 accordingly, as rocks are not usually equivalent to individual minerals (e.g., in terms of their
16 composition). A single rock (e.g., a carbonatite) can be made up of many different minerals (e.g., calcite,
17 apatite, biotite) and should therefore be called a rock-fertilizer. Both rock fertilizers and agrominerals are
18 sought after as local, inexpensive nutrient sources for crops which require less processing and shipping
19 costs than industrially-produced chemical fertilizers. While their low solubility can help mitigate nutrient
20 runoff compared to chemical fertilizers (Bakken et al. 1997a; van Straaten 2006), it also hinders their
21 widespread use because they must be weathered before their nutrients can be accessible to plants (Harley
22 and Gilkes 2000; van Straaten 2006). Carbonatite rocks are particularly appealing as rock fertilizers as
23 they are made up predominantly of carbonate minerals which have higher weathering rates than silicate
24 minerals (Chou et al. 1989). Yet, they have not been well studied in this capacity.

The aim of this review is to assess the agronomic potential of carbonatite rock fertilizers and to determine under which conditions they are effective. A brief overview of agrogeology will first be presented followed by a review of research on the use of carbonatite rocks in agriculture. Next, the factors that affect mineral weathering will be briefly covered to inform appropriate assessment and utilization of carbonatites as nutrient resources. We will then present a working model to explain how carbonatites are predicted to affect agroecosystems. Finally, we will conclude with considerations to guide further research on carbonatites and other similar rock fertilizers.

Agrogeology: the use of geology for agricultural benefit

The use of rocks and minerals to benefit crops is not a new concept. Sedimentary limestones like calcite and dolomite are often used in agricultural contexts to counter soil acidification (Holland et al. 2018). While the main agricultural benefit of limes is to increase soil pH and decrease the availability of harmful metals like aluminum, the soil geochemical changes caused by limes have other indirect effects that can benefit plants. For instance, liming is generally viewed as beneficial to soil bacteria, boosting their activity as soil nutrient-cyclers (Acosta-Martínez and Tabatabai 2000; Fuentes et al. 2006; Holland et al. 2018). Thus, the long-term (7 year) effects of lime to a Kenyon loam soil in Iowa improved the activity of 13 microbial enzymes like glutaminase and alkaline phosphatases but lowered the activity of acid phosphatases (Acosta-Martínez and Tabatabai 2000). Over shorter time-frames (1-3 months), limes were shown to be detrimental to soil stability because they decrease soil particle size through disrupting the aluminum bonds in clay minerals, but over longer time-frames (>6 months), they were shown to be beneficial because of secondary mineral formation (Haynes and Naidu 1998). The soil geochemical alterations brought about by limes have come back into focus with regards to climate change, greenhouse gas emissions from soils, and soil carbon stocks. Liming affects several levels of the carbon cycle depending on the soil and conditions: on one hand it increases the activity of soil microorganisms and thus should encourage the release of more soil-bound CO₂, and on the other hand it stabilizes the soil

materials and increases plant productivity which should promote soil carbon stabilization (Paradelo et al. 2015; Kalkhoran et al. 2019). Furthermore, a reduction of 49% of N₂O emissions from agricultural soils with liming has been recently reported (Hénault et al. 2019); this reduction was attributed to changes in the soil pH which became more conducive to N₂O reduction. While there may be a focus on the ability of geological resources like carbonatites to provide plant nutrients for agricultural benefit, other benefits like those seen from limes, i.e., increased soil microbial activity or reductions in greenhouse gas emissions, are also important to consider.

In the last few decades, a strong emphasis of agrogeology has been placed on the use of rocks and minerals as nutrient sources for plants. The basis of many modern fertilizers has been the reaction of phosphate rock (PR) or potassium-rich minerals (e.g., potash) with various chemicals to convert their contained nutrients into a highly water-soluble form (Zapata and Roy 2004; van Straaten 2007). The gains from these processes have depended strongly on the nutrient content of the rocks, and thus low-value ores with less nutrients have not usually been deemed useful for conversion as the cost to process them was too high. However, with the resurgence of low-input (“organic”) agriculture, these ores are now being reconsidered for use as rock-fertilizers (Zapata and Roy 2004) or soil amendments (Zhang et al. 2018). Much attention has been given to the use and efficacy of PRs as rock fertilizers, and they have been recognized by the United Nations Food and Agriculture Organization as important tools to achieve sustainable agriculture in developing countries (Zapata and Roy 2004). There are a wide variety of PRs, but key examples are the various forms of apatite (e.g., fluoro-apatite: Ca₅(PO₄)₃F or hydroxy-apatite Ca₅(PO₄)₃OH). Because rocks can be composed of one or more minerals, the term PR is often used liberally to mean any rock or mineral that contains P.

However, phosphorus is not the only plant nutrient found within geological resources. Other minerals like nepheline ((Na,K)AlSiO₄) or biotite (K(Mg,Fe)₃AlSi₃O₁₀(F,OH)₂) are also of interest in agrogeology as they are sources of K (Bakken et al. 1997a, 1997b; Manning 2010; Zhang et al. 2018). Regardless of the specific nutrient in question, the limiting factor that determines the effectiveness of a

given rock fertilizer or agromineral is its solubility (Harley and Gilkes 2000). Various methods have been proposed to offset this problem; one example is through the simultaneous application of P-minerals and P-solubilizing microorganisms in quantities appropriate to the specific agroecosystem (Reddy et al. 2002). The complexity of agroecosystems makes the usefulness of a particular method difficult to estimate because plants, microorganisms, and the interactions between them are all known to influence mineral weathering (Calvaruso et al. 2006; van Schöll et al. 2008; Uroz et al. 2009; Burghilea et al. 2015). A path around this complexity is to focus on rocks and minerals which weather rapidly. One group of rocks, the carbonatites, are considered to have relatively high weathering rates compared to other rock types and this makes them appealing targets for agrogeological research. Although carbonatites often contain high percentages of liming materials (e.g., igneous calcite or dolomite), it is unclear whether they are analogous to limes because of their mineralogical diversity.

Carbonatite rocks

Carbonatites are a form of igneous rock that are defined by their mineral composition: they are composed of a majority of carbonate minerals and a minority of silicate minerals (Woolley and Kempe 1989). Worldwide, there are 527 identified carbonatite deposits, of which 477 have been characterized in terms of their mineralogy (Woolley and Kjarsgaard 2008a, 2008b). The majority of described deposits can be found in Africa (32% of known deposits), Asia (30% of known deposits) and North America/Greenland (21% of known deposits; Woolley and Kjarsgaard 2008a). As with other geological resources (e.g., PR), carbonatites are **non-renewable**, and this should be considered with their usage. These deposits are millions or billions of years old (Woolley and Kjarsgaard 2008b), and so their formation occurs on a geological time scale. Therefore, while they may be useful as transitional environmentally-friendly nutrient sources, they should not be seen as a replacement for more renewable options like recaptured phosphorus from wastewater (Baker et al. 2015) or composted municipal solid wastes (Srivastava et al. 2016). Because their efficacy in nutrient delivery is still unclear, these rocks may

100 prove to be more beneficial as soil amendments to positively change soil geochemistry (e.g., similar to
101 lime to improve soil microbial activity) than as stable plant nutrients sources.

102 Carbonatites are highly diverse in their mineralogical composition, and there are numerous
103 minerals with potential agricultural use that have been found with their deposits (Table 1). Three
104 noteworthy examples of such minerals are calcite, apatite, and biotite (Woolley and Kempe 1989),
105 already mentioned for their individual use as agrominerals (Figure 1). However, other minerals
106 associated with carbonatite deposits could be detrimental to agriculture, such as the lead-sulfide mineral
107 galena or the barium-sulphate mineral barite (Table 2). Because of their diverse composition, carbonatites
108 can be named according to their primary carbonate mineral, e.g., calciocarbonatite (Woolley and Kempe
109 1989), or by the type of silicate rock they are associated with, e.g., nephelinite carbonatite (Woolley and
110 Kjarsgaard 2008a). In this review, we will focus on the agriculturally-relevant calciocarbonatites (Ca-
111 rich) and magnesiocarbonatites (Mg-rich), and disregard those which are not agriculturally-relevant, for
112 example the ferrocarbonatites (Fe-rich).

113 Despite their potential as rock fertilizers, carbonatites have only been minimally studied. Indeed,
114 works on rock fertilizers have focused mainly on silicate rocks and have mentioned carbonatites only
115 briefly (e.g., Gough and Herring 1993; van Straaten 2007) or not at all (Zhang et al. 2018). This is likely
116 because their high Ca and Mg contents are thought to limit their ability to break down and release
117 nutrients via the common-ion effect (i.e., high solution concentrations of Ca or Mg limit further
118 dissolution of minerals containing Ca or Mg; van Straaten 2002). Difficulties also arise in identifying
119 whether or not a given study is indeed focused on carbonatites because of the inconsistent or unclear
120 usage of geological terms (e.g., is a phosphate mineral used, or a rock which contains phosphate minerals,
121 or a phosphate rock?). In this review, only studies which refer specifically to carbonatites will be
122 discussed. The simplification of a carbonatite to simply a PR also neglects reference to and integration of
123 the complex mineralogy of these rocks. For instance, a carbonatite is referred to by Arcand et al. (2010)
124 as a phosphate rock "...originating from a carbonatite deposit...", and it is unclear whether the whole

rock or a subset of mineral(s) derived from the more complex carbonatite mineralogy was used. The same carbonatite used by Arcand et al (2010) had been previously found to contain roughly 30 different types of minerals, including apatite (Sage 1987).

Within the limited available research, two carbonatites have received particular attention: the Lillebukt Alkaline Complex carbonatite from Stjernøy, Norway, and the Spanish River Carbonatite (SRC) from Ontario, Canada (Table 3). Both carbonatites are composed mainly of calcite with accessory apatite, biotite, and other minerals, though the former is a silicocarbonatite while the latter is not. The Lillebukt carbonatite deposit was formed by igneous intrusion into hornblende pyroxenite approximately 525 million years ago (Gautneb et al. 2009) whereas the SRC deposit was formed by intrusive igneous activity into quartz monzonite bedrock 1838 million years ago (Sage 1987). Because of the age similarity between the SRC deposit and the nearby Sudbury Impact Crater (dated to 1840 million years ago; Petrus et al. 2015), the deposit may have arisen due to the geological instability caused by the impact.

The agronomic use of carbonatites

Largely, research into the use of agronomic carbonatites has focused on their role as alternative nutrient sources for plants, especially for K. The carbonatite in the Lillebukt Alkaline Complex has prompted several studies on the agronomic potential of this rock fertilizer. Bakken et al. (1997a) compared plant yield and K content of Italian ryegrass (*Lolium multiflorum italicum* var. Torilo) growing in three soils (peat, loamy sand, or silt loam) amended with several rocks and minerals, including the carbonatite from Lillebukt. In all soils, the carbonatite adequately supplied the plants with an amount of K similar to that of KCl; the ryegrass was able to take up 64% of the K provided as carbonatite and 72% of the K provided as KCl. The carbonatite was also significantly more effective in nutrient delivery than the other geological materials: for example, only 20% of the K provided as epidote schists was taken up by the ryegrass (Bakken et al. 1997a). Along with plants given KCl, plants provided with carbonatite had significantly higher dry weight than those plants in the other treatments. The same research group tested

the efficacy of several K-containing rocks as slow release K-fertilizers on barley (*Hordeum vulgare* L.) grown in peat moss (Bakken et al. 1997b); this study included two carbonatite rocks from the Lillebukt complex with different associated minerals (one with nepheline and one with biotite). The authors found that the minerals associated with the carbonatite varieties differed in their ability to provide K, whereby approximately 50% of the K provided as biotite-carbonatite was taken up by the plants and less than 25% was taken up from the nepheline-carbonatite (Bakken et al. 1997b). Plants given the former yielded nearly as much total plant dry matter as those provided with KCl: 112 g pot⁻¹ for biotite carbonatite, and 137 g pot⁻¹ for KCl, whereas those given the latter yielded significantly less (90 g pot⁻¹ for nepheline carbonatite). Despite the differences between the two carbonatite varieties, both of these studies indicate that nutrients within carbonatites can be accessible to plants at rates which are often similar to those of chemical fertilizers, at least in terms of K.

More recently, a study encompassing the nutrient content measurement of the vegetation overlying the Lillebukt deposit and an assessment of the impact of soil Ba on the plants was performed by Myrvang et al. (2016a). Barium is of particular concern to agroecosystems because elevated levels in plants are known to depress photosynthesis and growth (Suwa et al. 2008), and thus have a detrimental impact on yield. Myrvang et al. (2016a) reported that the surveyed herbs, dwarf-shrubs, and grasses growing above the complex had higher levels of K, Mg, P, Ca, Sr, and Ba than would be expected from literature values; however, Ba did not appear to be negatively affecting the plants. Thus, despite its relatively high Ba levels, the carbonatite from the Lillebukt deposit appears to be efficient as a rock fertilizer as the plants growing over it are capable of taking up the nutrients released from it. In addition, the Lillebukt carbonatite may be useful as agricultural lime because of its high Ca and Mg content (Myrvang et al. 2017). While these studies provide invaluable information regarding the efficacy of carbonatite as nutrient sources, it is still unclear how carbonatites affect the majority of crops. Different plants react to the elements within carbonatites in different ways. For example legumes, especially bird vetch (*Vicia cracca*), tend to take up more Ba from carbonatite or Ba-enriched soils than non-legumes

independently of whether soils had been limed (Myrvang et al. 2016a; 2016b), and plants with low Ca affinity (e.g., barley) had reduced Ba uptake when soils were limed (Myrvang et al. 2016b). Thus, the plant type and growing situation (i.e., limed or not) may have important ramifications for the effective agricultural use of carbonatites. The role of microorganisms and their interactions with other agroecosystem components relating to effective carbonatite breakdown remains elusive, although there is evidence that the microbial relationships within the rhizosphere plays a role in the mobilization of nutrients from carbonatites (Myrvang et al. 2016c).

Stemming from the interest of farmers who have used SRC as a commercial liming agent, several recent works have explored the agricultural use of this rock fertilizer. Changes caused by the presence of SRC on the soils, the plants, and the soil microorganisms around the deposit were explored by Jones et al. (2019). Where the glacial till layer is thin enough, SRC appears to have led to the creation of ‘islands’ of more basic soil (~pH 6) in an otherwise acidic (~pH 5) forest soil (Jones et al. 2019). In the previous study, soils in the deposit were separated into different categories of SRC influence based on their chemistry. Those which were highly influenced by SRC were found to have higher Mn content, higher cation-exchange capacities (CEC), and higher Ca levels than those soils with negligible carbonatite influence (Jones et al. 2019). The highly SRC-influenced sites were also lower in Al, S, and P contents than the negligibly-influenced sites. The changes in soil chemistry were found to be associated with changes in microbial abundance and plant community composition. In terms of microorganisms, the abundance of several bacterial operational taxonomic units (OTUs) were decreased in highly SRC-influenced soils compared to soils with low/negligible influence. However, two OTUs from the Gaiellaceae and Micrococcaceae family were of higher abundance (Jones et al. 2019). Fungi were less affected by SRC than bacteria, though there was likely an interaction between fungal abundance and tree species. Specifically, many of the differentially-abundant fungal OTUs were identified as mycorrhizal fungi, and tree species (i.e., mutualistic hosts) were typically dissimilar between the surveyed sites (Jones et al. 2019). The herb and shrub communities in SRC-influenced soils had some similarities to those

located on soils outside the deposit, but were comprised more of ruderal plant species. This study, while not in an agricultural setting, does inform on what the agroecosystem responses might be from carbonatite addition.

In a field study, Arcand et al. (2010) explored the use of phosphate from various rock fertilizers, including SRC, on buckwheat (*Fagopyrum esculentum* Moench). Since buckwheat acidifies its rhizosphere, the authors expected that the crop would acquire P from the rocks, which would then be more available to subsequent crops when the plant residues were used as an organic P source. However, the results did not indicate good P uptake by buckwheat from any of the rock fertilizers, and since the soil was found to have high Ca content and a more basic pH, the authors attributed their findings to the basic soil conditions limiting rock weathering. This study thus emphasizes that the context in which carbonatites are used must be considered for their effectiveness to be realized, and that in some situation they will not be effective. In acidic soils, weathering is promoted and carbonatites should be effective as both nutrient-sources and liming agents (as in Bakken et al. 1997a, 1997b; Myrvang et al. 2017). In basic soils, carbonatites are unlikely to be effective as rock fertilizers as these soils contain high amounts of Ca and Mg which would limit mineral breakdown and nutrient release (as in Arcand et al. 2010). Finally, the nutrient content (e.g., of P or K) of the carbonatite should be kept in mind, as in this study only the highest application of PR (800 kg P ha⁻¹) had any effect. Thus, the usefulness of carbonatite rock fertilizers may be limited if high application rates or nutrient contents are needed.

Aside from these studies, not much is known about agricultural carbonatites. As mentioned earlier, carbonatites have often been considered only as PRs due to their associations with apatite. Such a simplification makes the criteria by which they are considered effective limited to P delivery, and the impacts of other elements are neglected. For instance, carbonatites are often rich in rare-earth elements (Nelson et al. 1988) and these have been shown to promote plant growth (e.g., by acting analogously to Ca⁺², Hu et al. 2004; Tyler 2004). However, recognition of the greater complexity of carbonatites is emerging, and interest in using carbonatite rocks in an agricultural context is demonstrated by several

recent theses (Christie 2019; Jones 2019; VanVolkenburg 2019). These studies include the combined use of carbonatite and cover crops (Christie 2019), the impact of carbonatites on multiple agroecosystem levels (VanVolkenburg 2019), and the responses of different plant types to carbonatite (Jones 2019). Progress is thus being made towards a detailed understanding of how carbonatites can affect and are affected by different agroecosystem components, and what is required for their effective use. However, the interactions between microbes and carbonatites still need to be studied further as these likely play an important role in modulating crop responses to carbonatite. Anecdotal studies on carbonatites conducted by farmers (e.g., a comparison between SRC, wollastonite, and basalt for effects on carrot flavour and nutrition; <https://bluegrassfarm.ca/pass-the-basalt/>) indicate the desire and need for this knowledge.

Mineral weathering, biological action, and nutrient release

While one of the appeals of carbonatites is their high inherent weathering capacity, their effectiveness may still be limited under certain conditions (e.g., basic soils with high Ca and Mg). The weathering of rocks and the minerals within them is the result of the cumulative actions of abiotic and biotic components, and these actions can be numerous even in a highly simplified agroecosystem (Figure 2). At the fundamental level, the relative solubility of a given mineral can be expressed by its solubility product constant (K_{sp}) which is a function of the 3-D arrangement and bonding pattern of the contained elements. The K_{sp} for many minerals has been determined under various abiotic conditions with lower numbers referring to less soluble materials; for example, the log K_{sp} of calcite at standard temperatures and pressures is -8.48 (Chou et al. 1989) whereas the log K_{sp} for quartz under similar conditions is around -13 (Tester et al. 1994). Under acidic pH and abiotic sterile laboratory conditions, carbonate mineral dissolution depends on the movement of the dissolved ions away from the mineral surface, on the ambient temperature, and on the concentration of CO_2 in solution (Chou et al. 1989). In soils, the important abiotic determinants of weathering arise from the soil solution chemistry, and also include

water content and pH (Figure 2A; Goldich 1938; Chou et al. 1989; Harley and Gilkes 2000). These determinants are affected by the size of the particles within the soil, as well as the percentage of organic matter (Figure 2A; Huang et al. 2002). The properties of the mineral, including the composition, the crystal structure, the surface area, and the texture, are also important as they influence the weathering reactions. The amount of silicate in a given material has also been considered indicative of its resistance to weathering - those minerals with lower silicate content are expected to weather more rapidly than those with more silicates (Goldich 1938; Harley and Gilkes 2000). In an agricultural context, both abiotic factors and biotic factors are expected to be drivers of carbonatite weathering.

One of the major biotic components that minerals encounter are microorganisms, and factors that affect microorganisms can influence the extent to which they weather minerals. Directly, bacteria (Figure 2B; Rogers and Bennett 2004; Whitman et al. 2018) and fungi (Figure 2C; Whitman et al. 2018) take up dissolved nutrient ions from weathering minerals, and produce a variety of compounds such as low molecular-weight organic acids and siderophores which will increase mineral dissolution (van Schöll et al. 2008; Ahmed and Holmström 2015). Indirectly, other influences come into play. For example, carbon as a limiting nutrient for microbial growth (Demoling et al. 2007), the presence of a preferred carbon substrate (e.g., glucose for mineral-weathering bacteria; Hameeda et al. 2006; Uroz et al. 2007), the soil pH (e.g., neutral pH for bacteria; Lauber et al. 2009; Rousk et al. 2010), and the presence of mutualistic hosts (e.g., fungi; van Schöll et al. 2008; Burghellea et al. 2015) can all modulate mineral weathering by affecting the activity of microorganisms at the mineral surface. Proper soil nutrition is also a concern for soil fungi, as they respond negatively to the presence or absence of certain nutrients (e.g., high P limiting the growth of mycorrhizal fungi; Zhong et al. 2010).

Microorganisms are expected to play a key role in weathering agricultural carbonatites. In 2015, Uroz et al. proposed to view soil-based minerals as inorganic analogues to plant roots in terms of their effects on microbial population growth; as such, the authors coined the term “mineralosphere”. Just as the rhizosphere is the zone around the root where microorganism communities are shaped by the root’s

influence, the mineralosphere is the zone wherein the mineral determines the microorganism communities. Such a connection between microbial community composition and type of mineral has been recently shown by Whitman et al. (2018) in a microcosm experiment using quartz, ferrihydrite, and kaolinite. This study builds on previous work by Colin et al. (2017), where both the mineral type and the tree species were found to influence the soil microbial communities. Although the number of microbial types on the minerals was comparable to that of the bulk soil, microbial communities around both minerals and trees were composed of different members. Mineralospheric microorganisms have access to nutrients like iron (Rogers and Bennett 2004; Whitman et al. 2018), manganese (Colin et al. 2017), phosphorus (Bennett et al. 2001; Rogers and Bennett 2004) and trace nutrients like silicon (Ferris and Lowson 1997). However, since much of the soil carbon is thought to come from plant roots (Rasse et al. 2005; Philippot et al. 2013), microorganisms in the mineralosphere are not able to access carbon to the same extent as those in the rhizosphere.

Another major biotic component that minerals encounter are plant roots. Like microorganisms, plants can be considered in terms of their direct and indirect actions toward mineral weathering (Figure 2D). Directly, they can increase weathering by exuding organic acids (e.g., isocitric acid from *Arabidopsis* roots; Badri and Vivanco 2009), by taking up newly-liberated ions from the nearby mineral solution (Calvaruso et al. 2006), and through beneficial interactions with soil microorganisms (e.g., mycorrhizal fungi; Burghelea et al. 2015). Indirectly, increased growth benefits from microbial mutualisms (Calvaruso et al. 2006; Philippot et al. 2013; Burghelea et al. 2015), proper plant nutrition (Havlin et al. 1999), growing conditions and pest pressures (Tilman et al. 2002) are all expected to have a positive impact on mineral weathering (Figure 2D).

The intersection between rhizosphere and mineralosphere seems especially important to mineral weathering as microorganisms therein are able to access both plant carbon and mineral nutrients. Indeed, autotrophic carbon from plant roots has been proposed by Banfield et al. (1999) to fuel microbial weathering of nutrient-bearing minerals. Their model is based on the known interaction between minerals

and lichens: as the algal component provides carbon to drive the metabolism of the fungal component, the latter, during its growth, releases organic acids and penetrates the mineral with its hyphae. These two fungal processes promote the release of nutrients which are then used by both fungi and algae for their growth, which in turn further increase weathering. Banfield et al. (1999) postulated that this positive feedback also occurs with plant roots and soil-dwelling microorganisms. However, a plant component is not necessarily needed to provide the autotrophic carbon, as shown in a study by Ferris and Lowson (1997) who found that a microbial ecosystem had developed around mineral weathering in the sedimentary dolomitic limestone of the Niagara escarpment. Cyanobacteria and other photosynthetic microbes were thought to act in producer roles, fixing atmospheric carbon which was then utilized by heterotrophic microorganisms to acquire trace nutrients from the dissolution of the rock. With plants, the combined action of roots and microorganisms to increase mineral weathering can be seen in the two following examples. First, significant increases in the weathering of biotite due to the action of both pine roots (*Pinus sylvestris*) and strains of *Burkholderia glathei* were found by Calvaruso et al. (2006) through direct observation of the minerals via scanning-electron microscopy. Second, a bacterial endophyte (*Enterobacter asburiae* strain 3FII) that colonizes root hairs of *Zea sp.* and *Lolium multiflorum* was noted to not only increase the length of these hairs but also help solubilize rock P through rhizosphere acidification (Shehata et al. 2017). With longer root hairs, the plants had a higher absorptive surface area to take up P solubilized by the bacteria (Shehata et al. 2017). Based on these examples, it is expected that the actions of microorganisms and plants be synergistic and promote the weathering of rock fertilizers and agrominerals in a manner similar to that proposed in the model of Banfield et al. (1999). Additional work is needed to confirm and characterize these interactions, however.

Ultimately, both abiotic and biotic components of mineral weathering need to be considered when evaluating the effectiveness of rock fertilizers such as carbonatites in an agricultural context. While the K_{sp} values estimated in a laboratory can be useful for predicting the efficacy of nutrient delivery to plants, the complexity of agricultural systems often means that these values should not be relied upon

solely to gauge mineral solubility under biotic conditions (Zhang et al. 2018). As such, in studies on rock fertilizers and agrominerals, the differences in the nutrient content between plants grown with and without the geological materials have often been used as an estimation of nutrient release (Bakken et al. 1997a, 1997b; Arcand et al. 2010; Myrvang et al. 2016a).

A model for carbonatite breakdown in a three-component agroecosystem

With the available data, a working model can be proposed to guide predictions on the impacts of carbonatite rock fertilizers on plants, soils, and microorganisms in a three-component agroecosystem (Figure 3). Although the model is simplistic, it is hoped that it can be expanded upon as a more detailed understanding of the interactions between these and other components becomes available.

At the soil level (Figure 3A), dissolution of carbonatites is expected to alter the soil chemistry by increasing pH, CEC, Ca and Mg contents, and the amount of micronutrients like Mn. These expectations are based upon a survey of the SRC deposit (Jones et al. 2019) as well as on data obtained from the Lillebukt carbonatite (Myrvang et al. 2016a, 2016b, 2016c, 2017). However, a number of important questions remain about the impact of carbonatites on soils, such as whether the emission of greenhouse gasses (e.g., CO₂, NO_x, CH₄), the runoff of highly-mobile nutrients, the sequestration of organic matter, and the soil structure (e.g., aggregate stability) are altered. Answers to these questions will greatly influence whether carbonatites can be useful as rock fertilizers. As carbonatites can introduce large quantities of Ca into the soils in a manner similar to that of lime, they may also behave similar to limes in agricultural contexts, e.g., in terms of their effects on soil microbial activity or greenhouse gas emissions. However, this has not yet been assessed. Additionally, since it appears that Mn is introduced with SRC, its use or the use of similar carbonatites may affect Mn cycling in soils systems, e.g., by providing more Mn for use in organic matter breakdown by fungi (Keiluweit et al. 2015) or by increasing long-term production of humic compounds by Mn oxides (Shindo and Huang 1982; Huang et al. 2005). Finally, it is

expected that carbonatites will have negligible effects in soils which are already high in Ca and/or pH due to negligible breakdown of the rocks (e.g., as in Arcand et al. 2010).

At the level of individual plants (Figures 3B and 3C), carbonatites are expected to have a number of impacts which will depend primarily on the plant's traits. Following the work of Wen et al. (2019), plant responses to carbonatites are divided based on their nutrient foraging strategies as exemplified by root thickness. Crops with thin roots (Figure 3B; e.g., wheat) that increase individual root length to explore more soil volume are expected to benefit strongly from carbonatite application. Assuming a random distribution of carbonatite particles throughout the soil, the more soil volume a root system explores, the more particles the roots encounter and the more nutrients are taken up. While each carbonatite particle may not be highly weathered, the incomplete weathering is compensated for by the high number of particles encountered and acted upon by the root system (Figure 3B). While the rhizosphere microbiome is expected to influence nutrient availability and plant growth in this context, more research is needed to elucidate to what extent. For example, rhizosphere microorganisms appeared to play a significant role in the mobilization of Ba from carbonatites (Myrvang et al. 2016c), and likely do the same for other elements.

For crops with thicker root systems (e.g., pea), the situation is more complex, but insights may still be gained through the lens of nutrient-foraging strategies (Figure 3C). Thus, plants with thicker roots are expected to produce more root exudates than those with thin roots; they also generally have stronger partnerships with soil microorganisms (e.g., more root colonization by mycorrhizal fungi; Wen et al. 2019). Pea, specifically, increases exudation patterns and partners with nitrogen-fixing rhizobia and mycorrhizal fungi in situations of low nutrient availability (Lodwig et al. 2003; Carbonnel and Gutjahr 2014; Wen et al. 2019). While fewer carbonatite particles may be encountered by a plant with thick roots, those particles that are encountered will likely be highly weathered due to the combined action of plant and microorganisms (Figure 3C). Partnerships with mycorrhizal fungi may prove to be especially important here as these fungi are considered to extend the reach of plant root systems, solubilize P, and

transfer it to the plants (Friese and Allen 1991; Smith et al. 2001; Kernaghan 2005; Kiers et al. 2011; Zhang et al. 2016). Care will have to be taken when carbonatites are used for thick-rooted plants, as the results will likely depend on the cumulative interactions between the carbonatite, the plants, and the microorganisms. Furthermore, the physiological responses of plants to different nutrient sources are expected to add complexity. For example, the type of nitrogen provided to soybean (*Glycine max* (L.) Merr.) has been shown to influence whether the plants acidifies or alkalizes its rhizosphere based on differential cation/anion uptake (Aguilar and van Diest 1981). Soybean provided with nitrate alkalized its rhizosphere, whereas symbiotic soybean provided with fixed nitrogen acidified its rhizosphere – the latter rhizosphere was much more effective in PR breakdown than the former (Aguilar and van Diest 1981).

The effect of carbonatites on soil microorganisms (Figure 3D) is hard to predict based on existing data. Results obtained from the survey of the SRC deposit indicate that carbonatite influences the abundance of several microorganisms (Jones et al. 2019). While a specific mechanism to explain the differential abundance of microorganisms cannot be presented here, two possibilities which are not mutually exclusive can be put forward: 1) elements in relatively high abundance in carbonatites favour microorganisms able to tolerate them (e.g., Mn and its tolerance by *Arthrobacter spp.*; Ghiorse 1988) or 2) the nutrients in carbonatites select for microorganisms capable of releasing them from the minerals (Uroz et al. 2015). More generally, carbonatites will likely act similarly to lime in promoting bacterial growth through alleviation of acidic soil conditions and the production of a more conducive soil environment (Fuentes et al. 2006). The presence of plants is expected to have a strong impact on microbial responses to carbonatites (Figure 3B and 3C). Soil microbial populations are most often C-limited (Demoling et al. 2007), and plant roots are a large source of soil C (Helal and Sauerbeck 1986). Therefore, when the rhizosphere and the mineralosphere (e.g., around carbonatite particles) intersect, strong benefits would likely be realized by those microorganisms able to take advantage of conditions imposed by both the root and the rock. In other words, a general mutualism between microbes and plants based on cooperative exploitation of a geological resource would be established (Banfield et al.

1999). This would likely lead to a positive feedback loop where nutrients released from the carbonatite dissolution encourage the growth of plants and microorganisms, which then further weather the minerals, resulting in the release of more nutrients. Whether this positive feedback can be effectively harnessed to promote weathering of carbonatites (or other rock fertilizers) for plant benefits or already occurs remains to be seen. The release of CO₂ from carbonate mineral dissolution likely also affects the microbial dynamics, as this carbon has been shown to be available for fixation by autotrophic microorganisms (Guida et al. 2017). The contributions of several microbial genera may prove to be key in effective agricultural usage of carbonatites: For instance *Arthrobacter* *ssp.* with their tolerance to Mn and utilization of diverse carbon/nitrogen sources (Hagedorn and Holt 1975; Hungate et al. 1987; Ghiorse 1988), *Pseudomonas* *ssp.* with their known ability to weather minerals (Grayston et al. 1998; Uroz et al. 2007, 2009), and *Burkholderia* *ssp.* for their weathering effects on several different mineral types (Uroz et al. 2009). Other microbial genera shown to weather minerals include *Collimonas* *ssp.* and *Sphingomonas* *ssp.* (Uroz et al. 2007), *Azotobacter* *ssp.*, *Shewanella* *ssp.*, and *Streptomyces* *ssp.* (Uroz et al. 2009).

Estimating the usefulness of carbonatites

Research on agricultural carbonatites has covered a very small number of the known global deposits. Because of the mineralogical diversity of carbonatites (Table 1), a means of predicting whether a given carbonatite will be agronomically useful would be beneficial. We propose three properties which can be used as a starting point to assess the potential of a carbonatite deposit using available geological information:

- 1) The mineralogy and reactivity of the deposit in terms of nutrients.
- 2) The quantity of the material contained within the deposit.
- 3) The deposit's location relative to the intended sites of use and to ecologically-sensitive area(s)

The mineralogy of a deposit is a key consideration for determining the potential usefulness of a carbonatite and the agricultural contexts in which it might be useful. As examples, two deposits that show promising nutrient composition are the Lillebukt carbonatite deposit in Norway (Myrvang et al. 2017) and the SRC deposit in Canada (Sage 1987; Jones et al. 2019). Both deposits are composed of a calcite carbonatite with constituent biotite and apatite minerals, which would serve as sources of calcium, potassium, and phosphorus, respectively. Conversely, the presence of harmful elements like uranium may discourage exploration, as is found in the deposit located on the Manitou Islands of Lake Nipissing in Canada (Woolley and Kjarsgaard 2008b). The use of carbonatites containing other elements, such as Ba or Mn, should be done cautiously because improper use (e.g., used excessively in hypoxic soils) may lead to phytotoxicity. This may be the case with Ba in the Lillebukt (Myrvang et al. 2016c, 2017) and with Mn in the SRC (Jones et al. 2019) deposits. Excessive uptake of these elements is associated with disruptions to the photosynthetic machinery and carbon fixation and may affect growth and yield (Suwa et al. 2008; Fernando and Lynch 2015). However, Mn is an important plant micronutrient which is integral to the photosynthetic process, and deficiency of this element can be a problem in highly-leached soils (Fageria 2001). It is also worthwhile mentioning that the harmful effects from these two elements have not yet been observed in agricultural use of the Lillebukt and Spanish River carbonatites.

Similarly, the volume of material within the deposit should be considered prior to exploiting a deposit. Carbonatites are non-renewable resources and the amount of carbonatite present is directly proportional to the long-term economic viability of the deposit. The ore grade (amount of mineral in a specific ore) is also a factor, as deposits can be heterogenous in their mineralogy. As an example, the SRC deposit has three zones each with differing amounts of apatite, calcite, biotite, and vermiculite (Sage 1987) which have differing soil chemistry signatures (Jones et al. 2019) and presumably different levels of agricultural usefulness. This can be in terms of carbonatite use as a rock fertilizer, as a liming material, or as a soil amendment.

Finally, the location of the deposit is of prime importance as it directly relates to both the environmental impact of mining operations and the operational cost of mining and transporting the

harvested material. The Elchuru carbonatite deposit in India and the Bull's Run deposit in South Africa (Woolley and Kjarsgaard 2008b) are both near populated areas and share broad mineralogical similarity to the Spanish River and Lillebukt carbonatites, and so these could be viable deposits for agricultural use. As with other industrial processes or agricultural amendments, the development and the use of carbonatites will have to consider health hazards and so environmental and workplace risk assessments will need to be undertaken.

Deposits in ecologically-sensitive areas should not be considered, as the environmental cost of mining needs to be balanced against the agricultural benefits. Furthermore, many areas around the world are protected against resource development like mining. For example, the Allan Lake carbonatite deposit is located in the Algonquin Park area in Ontario, Canada (Ford et al. 1988), and so development would be prohibited following the Provincial Parks and Conservation Reserves Act (<https://www.ontario.ca/laws/statute/06p12#BK16>). Because carbonatites are globally located, preference should be given to those deposits which are local instead of those geographically distant to reduce the shipping costs and need for access roads.

Ultimately, the effectiveness of carbonatites to serve a useful function in agriculture needs to be demonstrated with field trials under different conditions and with different crops. The appeal of carbonatites is that they can serve as locally-sourced, low-cost, and environmentally-friendly nutrient sources for crop plants and so they should be assessed according to how well they meet these criteria. However, given the complexities of mineral weathering (Figure 2), interactions of the carbonatite with plants, soils and soil microorganisms need to be considered in assessments. Also, carbonatites have not yet been assessed in terms of their potential to reduce or contribute to nutrient leaching/runoff (Myrvang et al. 2017).

Conclusions and future directions

Because of their high relative weathering rates compared to those of other rocks and their associations with nutrient-bearing minerals (e.g., apatite and biotite), carbonatites have strong potential as rock fertilizers for crop plant nutrition. However, more research is required as current studies are limited in both substance (few carbonatites assessed) and scope (typically only plant effects on a few species measured). The incorporation of knowledge regarding crop traits (e.g., rhizosphere acidification) with mineral breakdown and carbonatite effectiveness is needed as the effect of carbonatites on plants is strongly context-dependent, e.g., it appears to depend on the soil conditions, microbial communities, plant species, and time span in which they are used. Considerations beyond the effects of carbonatites on plants are thus a necessity to understand how plants-microorganisms-carbonatites-soils interact. This is highlighted by the complexities of mineral weathering, which can be affected by numerous factors even in a simple system. A model is presented to guide the work needed to further characterize which deposits can be considered agriculturally-relevant and the impacts of carbonatites under different agricultural conditions. The environmental impacts of carbonatite use should be incorporated in future research, whether in terms of benefits (e.g., less processing required for use as a fertilizer) or costs (e.g., development of deposit, removal of overlying biota, and the environmental impact of the mining).

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Tables:

Table 1: Non-exhaustive list of nutrient-bearing minerals organized by abundance which have been found associated with carbonatite deposits. Only essential (e), common (c), and moderately common (m) minerals are included here following Heinrich's classification (1980).

Name	Formula	Abundance	Mineral type
<i>ankerite</i>	$\text{CaFe}(\text{CO}_3)_2$	e	carbonate
<i>biotite</i>	$\text{K}(\text{Mg,Fe})_3\text{AlSi}_3\text{O}_{10}(\text{F,OH})_2$	c	silicate
<i>calcite</i>	CaCO_3	e	carbonate
<i>dolomite</i>	$\text{CaMg}(\text{CO}_3)_2$	e	carbonate
<i>aegirine</i>	$\text{NaFeSi}_2\text{O}_6$ (Fe as Fe^{3+})	c	silicate
<i>pyrite</i>	FeS_2	c	sulfide
<i>apatite</i>	$\text{Ca}_5(\text{PO}_4)_3(\text{F,Cl,OH})$	c/e	phosphate
<i>hematite</i>	Fe_2O_3	c/e	oxide/hydroxide
<i>magnetite</i>	Fe_3O_4	c/e	oxide/hydroxide
<i>siderite</i>	FeCO_3	m/e	carbonate
<i>albite</i>	$\text{NaAlSi}_3\text{O}_8$	m	silicate
<i>antigorite</i>	$(\text{Mg,Fe})_3\text{Si}_2\text{O}_5(\text{OH})_4$	m	silicate
<i>augite</i>	$(\text{Ca,Na})(\text{Mg,Fe,Al,Ti})(\text{Si,Al})_2\text{O}_6$	m	silicate
<i>chalcopyrite</i>	CuFeS_2	m	sulfide
<i>chlorite</i>	$(\text{Mg}_5,\text{Fe}_5,\text{Ni}_5,\text{Mn/Al})(\text{AlSi}_3)\text{O}_{10}(\text{OH})_8$	m	silicate
<i>chrysotile</i>	$(\text{Mg,Fe})_2\text{SiO}_4$	m	silicate
<i>crocidolite</i>	$\text{Na}_2(\text{Fe}^{+2}(\text{3}),\text{Fe}^{+3}(\text{2}))\text{Si}_8\text{O}_{22}(\text{OH})_2$	m	silicate
<i>diopside</i>	$\text{MgCaSi}_2\text{O}_6$	m	silicate
<i>microcline</i>	KAlSi_3O_8	m	silicate
<i>nepheline</i>	$\text{Na}_3\text{KAl}_4\text{Si}_4\text{O}_{16}$	m	silicate
<i>olivine</i>	$(\text{Mg}_{+2},\text{Fe}_{+2})_2\text{SiO}_4$	m	silicate
<i>orthoclase</i>	KAlSi_3O_8	m	silicate
<i>phlogopite</i>	$\text{KMg}_3\text{AlSi}_3\text{O}_{10}(\text{F,OH})_2$	m	silicate
<i>pyrrhotite</i>	$\text{Fe}(1-x)\text{S}$, $x=0-0.2$	m	sulfide
<i>sphalerite</i>	Zn/FeS	m	sulfide
<i>sphene</i>	CaSiO_5	m	silicate
<i>vermiculite</i>	$(\text{Mg, Fe}^{+2}, \text{Fe}^{+3})_3(\text{Al,Si})_4\text{O}_{10}(\text{OH})_2 \cdot 4\text{H}_2\text{O}$	m	silicate
<i>vesuvianite</i>	$\text{Ca}_{10}(\text{Mg,Fe})_2\text{Al}_4(\text{SiO}_4)_5(\text{Si}_2\text{O}_7)_2(\text{OH,F})_4$	m	silicate
<i>wollastonite</i>	CaSiO_3	m	silicate

Table 2: Non-exhaustive list of carbonatite-associated minerals organized by abundance which are not useful for agriculture. Only essential (e), common (c), and moderately common (m) minerals are included here following Heinrich' classification (1980).

Name	Formula	Abundance	Mineral type
<i>quartz</i>	SiO ₄	c	silicate
<i>barite</i>	(Ba,Sr)SO ₄	c/e	sulfate
<i>fluorite</i>	CaF ₂	c/e	halide
<i>pyrochlore</i>	(Na,Ca) ₂ Nb ₂ O ₆ (OH,F)	c/e	oxide/hydroxide
<i>bastnaesite</i>	(Ce,La,Y)CO ₃ F	m/e	oxide/hydroxide
<i>baddeleyite</i>	ZrO ₂	m/c	oxide/hydroxide
<i>ilmenite</i>	FeTiO ₃	m/c	oxide/hydroxide
<i>perovskite</i>	CaTiO ₃	m/c	oxide/hydroxide
<i>rutile</i>	TiO ₂	m/c	oxide/hydroxide
<i>betafite</i>	(Ca,U) ₂ (Ti,Nb,Ta) ₂ O ₆ (OH)	m	oxide/hydroxide
<i>columbite</i>	(Fe,Mn)Nb ₂ O ₆	m	oxide/hydroxide
<i>fersmite</i>	(Ca,Ce,Na)(Nb,Ta,Ti) ₂ (O,OH,F) ₆	m	oxide/hydroxide
<i>galena</i>	PbS	m	sulfide
<i>monazite</i>	(Ce,La,Nd,Th,Sm,Gd)PO ₄	m	phosphate
<i>strontianite</i>	SrCO ₃	m	carbonate
<i>zircon</i>	ZrSiO ₄	m	silicate

Table 3: Comparison of the Lillebukt and Spanish River Carbonatites.

Deposit	Lillebukt Carbonatite	Spanish River Carbonatite
Age	525 million years	1838 million years
Surface area	12 km ²	3.25 km ²
Origin	Igneous intrusion	Igneous intrusion/impact
Major minerals	Apatite (4.4%), biotite (37.7%), calcite (46.4%)	Apatite (0-15%), biotite (0-20%), calcite (50-100%)
Nutrient elements	P, K, Ca, Mg	P, K, Ca, Mg, Mn
Agricultural usefulness	Effective liming agent and nutrient-source but concerns regarding Ba content	Effective liming agent and nutrient-source but potential concerns regarding Mn content
Agronomic references	Bakken et al. 1997a, 1997b, Hillersøy 2010, Myrvang et al. 2016, 2017	Arcand et al. 2010, Jones 2016, Jones et al. 2019, Jones 2019, Christie 2019, VanVolkenburg 2019
Geological references	Gautneb et al. 2009	Sage 1987

Note: Major mineral percentages derived from listed geological references.

Figure captions:

Fig. 1: Connections between rocks, minerals, and chemical fertilizers. When whole rocks, which are aggregates of one or more minerals, are used in agriculture they are considered to be rock fertilizers. Minerals are composed of elements arranged in a specific 3D configuration. Like rock fertilizers, they can also be used in agriculture as agrominerals. Both rocks and minerals can be processed to form chemical fertilizers. Typically, nutrients are most available when they are provided as chemical fertilizers, although this is not always the case. For example, nutrient availability from carbonatites can be similar to that from chemical fertilizers. Furthermore, it should be noted that the solubility of rocks is not necessarily less than that of minerals depending on their composition.

Fig. 2: Key direct (black arrows) and indirect (white arrows) factors influencing mineral weathering in a simple agroecosystem composed of soils (A), bacteria (B), fungi (C), and plants (D). Direct factors are those that act at the level of the mineral itself to promote dissolution (e.g., acidic pH). Indirect factors are higher-level effects that impact the activity of components which then increases or decreases the direct effect of these components on mineral dissolution.

Fig. 3: Working model of how carbonatites are predicted to alter agroecosystems. A) The soil changes caused by the addition of a carbonatite amendment centre around geochemical alterations such as increases in Ca, Mg, and micronutrient contents. Plant responses will be a function of plant type, and are predicted to broadly follow one of two paths, depending on the nutrient foraging strategy of the plant. B) Thin-rooted plants are expected to undergo major root architectural changes including increased root branching and root system growth in order to maximize exploited soil volume. C) Thick-rooted plants are not expected to significantly alter their root systems, and instead will interact more with soil microorganisms and release more exudates to maximize nutrient mining of the already occupied soil volume. D) At the intersection between the rhizosphere and the mineralosphere, the changes are harder to anticipate, but it is predicted that carbonatite incorporation into the soil will increase microbial mutualisms with plants via cooperative mineral weathering, and that microbial community structures will be altered because of the presence of elements like Mn.

Figures:

Figure 1:

Figure 2:

Figure 3: