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Catalogue of South African mine tailings for geochemical carbon dioxide removal purposes

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ABSTRACT

South Africa has the potential to be a global leader in implementing strategies targeting geochemical carbon dioxide removal (CDR) using mine tailings, utilising methods for alkalinity production and mineral carbonation. This is due to the hundreds of millions of tonnes of geochemically and mineralogically suitable tailings produced annually from diamond, PGM, chrome, nickel, phosphate, copper, talc and magnesite mines, exploiting mafic and ultramafic rocks. This approach may offer an additional route towards emissions offsets and reduction targets for South Africa.

Here, a catalogue of tailings has been developed to highlight overall national CDR potential, drawing attention to site opportunities for pilot schemes. It has been calculated, using bulk geochemistry and shrinking core modelling, that South Africa has an average $1.1-1.7~\rm MtCO_2$ removal capacity per year across all suitable sites, while an additional $11-17~\rm MtCO_2$ could be removed through weathering historic tailings. The total CDR that could be achieved over the $2030-2100~\rm period$, considering all annually produced tailings available to weather, is $79-119~\rm MtCO_2$. The capacity may be higher if mines of limited production information are considered, and pre- $2017~\rm tailings$ production amounts, abandoned stockpiles, and other industrial by-products are included. Diamond mines hold the highest geochemical CDR capacities, along with high tailings producers in Phalaborwa, Nkomati and Mogalakwena metal mines. There are incentives for academia, industry and policymakers to revise the geochemical CDR potential of mine sites in the years to come, and for other countries to catalogue their own suitable tailings and geochemical CDR potential capacities for future Mt-scale opportunities.

1. Introduction

With the increasing need to explore methods of both reducing anthropogenic carbon dioxide (CO_2) emissions and actively removing CO_2 from the atmosphere in order to combat climate change and meet gigatonne (Gt)-scale IPCC reduction targets (IPCC, 2022), attention is turning to methods of CO_2 removal (CDR) strategies. One categorised approach that is now considered a viable option on an industrial scale is geochemical CDR (Kelemen et al., 2020; Canadell et al., 2021; Schenuit et al., 2021; Campbell et al., 2022), encompassing methods that target exploiting natural processes such as weathering and mineral carbonation of suitably Mg- and/or Ca-rich materials, such as mafic and ultramafic rocks, or alkaline industrial wastes. One key aim is to speed up natural processes that remove CO_2 to attain net CDR on human timescales, such as reactions occurring in seconds to tens of years rather than hundreds of thousands of years or longer. The end result is CO_2

conversion to form cation-stabilised soluble bicarbonates, i.e., alkalinity, or precipitated carbonate minerals, effectively storing CO_2 on a permanent (100's of thousands of years) basis. The main natural processes that can be targeted to achieve net CDR include:

Carbonate rock and mineral dissolution: e.g.,
$$H_2O + CO_2 + CaCO_3$$
 (calcite) $\rightarrow Ca^{2+} + 2HCO_3^-$ (1)

Silicate rock and mineral dissolution:

e.g.,
$$2H_2CO_3 + CaSiO_3$$
 (wollastonite) $\rightarrow Ca^{2+} + 2HCO_3^- + SiO_2 + H_2O$ (2)

Ca-carbonate precipitation: e.g.,
$$Ca^{2+} + 2HCO_3^- \rightarrow CO_2 + H_2O + CaCO_3$$
 (calcite) (3)

Mg-carbonate precipitation: e.g.,
$$Mg^{2+} + 2HCO_3^- \rightarrow CO_2 + H_2O + MgCO_3$$
 (magnesite) (4)

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The most common routes to achieve geochemical CDR, either through natural (or passive) means or engineered to occur on more rapid timescales, are through bicarbonate (alkalinity) generation, and carbonate precipitation. Rocks naturally react with weakly acidic rainwater in the atmosphere which, over time, will break down the contained minerals by mineral dissolution. In the case of silicate rocks which contain divalent cations, such as ${\rm Mg}^{2+}$ and ${\rm Ca}^{2+}$, these elements are released from their mineral structures in a reaction with atmospheric CO2 and water, which react to form stable bicarbonate solutions, stabilised by Mg and Ca, known as alkalinity. Through this reaction, CO2 is therefore removed from the atmosphere and stored as alkalinity. The alkalinity products ultimately end up in the ocean, where they can reside for tens to hundreds of thousands of years, or precipitate to form solid carbonate minerals (permanent CO2 storage). The natural process is a slow one, taking hundreds of thousands of years to occur, though deliberate efforts can be made to speed up reactions (e.g., enhanced weathering) so that dissolution and precipitation reactions occur on human-relevant timescales of decades or faster.

Approaches that could be made to achieve geochemical CDR include subsurface in-situ processes for mineral carbonation and geological storage within minerochemically suitable bedrock units, and ex-situ enhanced rock weathering and carbonation of minerochemically suitable loose, excavated or processed materials (Campbell et al., 2022). While regional and site-by-site case studies are vital for a clear understanding of the intricate minerochemical processes that take place under any implemented geochemical CDR scheme or pilot study (e.g., Wilson et al., 2009; Power et al., 2014), there is still a necessity to better catalogue the wider potential of extensive areas that host suitably reactive materials for future policy and regulatory planning, such as through the development of national inventories (Smith et al., 2023). Similar studies have been undertaken for mafic and ultramafic rock types in the United States (Krevor et al., 2009) and Japan (Myers and Nakagaki, 2020), and for suitable rocks and alkaline industrial wastes in the United Kingdom (Renforth, 2012; Alcalde et al., 2018; Kantzas et al., 2022), and Spain (Bullock et al., 2023). The assessment of Krevor et al. (2009) focused on olivine- and serpentine-bearing ultramafic rocks of the United States, deemed suitable for geochemical CDR by means of mineral carbonation. The study highlighted the large capacity and permanence of CDR by reactions of ultramafic rocks, and led to the development of a digital geologic database, noting that any mining operation is subject to several social, economic, and political considerations for any potential development in a given location. The study of Myers and Nakagaki (2020) explored direct gas-solid mineralisation potential of mafic-ultramafic surface exposed bedrock in Japan, noting that acceleration of CDR is best achieved by grinding if the starting material, drawing particular attention to mined surface rocks. Renforth (2012) investigated the Mt potential capacity of enhanced weathering (alkalinity generation) for mafic and ultramafic rocks of the United Kingdom, noting the important role of highly variable mineral dissolution rates achieved under different experimental conditions, particularly reflecting the potential variations in the starting grain size distribution. This was further emphasised by Bullock et al. (2023) for Spanish mafic-ultramafic bedrock and industrial by-products, with consideration for the maximum achievable CDR for a given material and how much could theoretically be achieved within a given time period based on mineral dissolution rates from different reaction conditions and starting grain sizes.

South Africa has also received attention for its high geochemical CDR potential, particularly for mine by-products that are created through the exploitation of the mineralogically and geochemically favourable host rocks in the Bushveld Complex (e.g., Hietkamp, 2005; Doucet, 2011; Vogeli et al., 2011; Meyer et al., 2014), and the extensive kimberlite fields that host diamonds and diamond mine operations (e.g., Mervine et al., 2018; Stubbs et al., 2022; Zeyen et al., 2022a). These studies are warranted based on South Africa's need to reduce its CO₂ emissions. South Africa emitted 435 MtCO₂ from fossil fuel combustion and

industrial purposes in 2020 (Statista, 2021). South Africa's current climate action tracker for meeting reduction targets is classified as "insufficient", although their 2030 emissions reduction target (submitted in September 2021), which follows the Presidential Climate Commission recommendation of a reduction of 350-420 MtCO₂, is close to being 1.5° C compatible (Climate Analytics, 2022). This suggests that additional methods to reduce emissions or remove CO₂ from the atmosphere could be highly beneficial for South Africa.

Due to the favourable geology, the considerable mining expertise and infrastructure, and increasing focus from academia, industrial and governmental bodies, South Africa has the potential to be a global leader in the implementation of geochemical CDR strategies at numerous individual pilot scales or rolled out on a systematic, much larger million tonne (Mt) scale. This is pertinent for the high volumes of mine tailings hosted within the country, which could be targeted for ex-situ geochemical CDR approaches. Mine tailings represent the often silicate-rich material that remains following processing and separation of the economic fraction of the deposit, stored in dams and ponds. As well as the high tonnages of suitably reactive and accessible material, mine tailings are typically fine-grained by design, often finer than 200 µm in mean grain size (Bullock et al., 2022). This means that the materials inherit favourably high reactive surface areas, reducing the need for additional energy-intensive excavation, crushing and grinding.

This study gathers available commodity production and milled material data for the tailings, coupled with geochemical and mineralogical indicators, to provide a baseline assessment of the geochemical CDR capacity of mine operations within South Africa. The database has been developed with the intention of being used for recommendations of suitable sites for further study. With increasing knowledge of sites and materials, the database can be updated and continually revised in the future for more accurate CDR capacity calculations, giving focus to sites that could be targeted for future geochemical CDR pilot schemes. The database may also provide the basis for any future national digital standardisation of tailings information and tailings sample availability for CDR studies or other purposes. Furthermore, the methods applied here may apply to other countries which host similar tailings or other industrial by-products.

2. Methodologies

The geochemical CDR capacity of South African mine sites presented here was compiled by the South African tailings for use in Carbon Capture and Storage (SAT4CCS) consortium (hosted at the University of Pretoria), based on publicly available data on mine operational status, commodity production data, milled or processed site material and inferred representative tailings geochemistry and mineralogy. Individual site capacities were calculated based on the estimated tailings production tonnage, assigned representative geochemistry and modal mineralogy of the tailings and shrinking core modelling.

2.1. Mine site list compilation

The original list of operational mines used in this study is adapted from the publicly available directory of operating mines and quarries, agents and mineral processing plants in South Africa, compiled and maintained by the Department of Mineral Resources and Energy. For this study, the list of operations that comprise CDR calculations is reduced to (1) mine sites that have operated since 2017 in some capacity, (2) sites with publicly available production information, and (3) sites that exploit rock types deemed suitable for geochemical CDR purposes, i.e., mafic and ultramafic host rocks. Targeted tailings include those from kimberlite-hosting diamond mine operations and metal-producing operations from a range of mafic and ultramafic host rocks - platinum group metal (PGM), chrome, nickel, phosphate, copper, talc, manganese and magnesite operations from the Bushveld Complex or geochemically similar lithological units. This is due to their relatively

higher abundance of common Mg- and Ca-rich minerals, such as serpentine (hydrous magnesium-rich phyllosilicate mineral), olivine (Mg-Fe orthosilicate), pyroxene (inosilicate of variable composition), plagioclase (Na-Ca-Al-bearing tectosilicate of the feldspar mineral group), amphibole (inosilicate of variable Fe-Mg-Ca-Na composition), chlorite (sheet silicate of variable composition, commonly containing Mg-Fe) and mica (phyllosilicate of variable composition), compared to other host rocks such as felsic igneous rocks or sedimentary rocks. Sites with post-2017 production data were chosen here due to limited scope and difficulty obtaining pre-2017 data across multiple operators. Additionally, accessing tailings older than five years from the time of writing may be challenging for CDR purposes (e.g., requiring energy-intensive and financially costly excavation techniques), and materials may have cemented, hardened, chemically altered and/or naturally carbonated, limiting their use for implemented geochemical CDR strategies. However, older tailings are not ruled out for geochemical CDR purposes, and their potential use should be assessed on a site-by-site case study basis.

2.2. Tailings production estimates

Tailings calculations made here are based on publicly reported milled tonnages (quantity of material delivered and processed to separate the economic fraction from the uneconomic fraction) for the period 2017-2021. Where milled tonnages are not publicly available, an estimated tonnage is presented based on an average conversion multiplier, calculated using information where both commodity production to tonnage milled are reported for mines targeting a given commodity for a given year (i.e., an average conversion factor for PGM mines based on available accumulated site data). For example, if chrome operations hypothetically produced i Mt of milled material for every j Mt of commodity production (on average across sites where both values are available), the conversion multiplier (k) would be (i/j). For any chrome sites that had production tonnages (x) but no milled material tonnages, the formula for tailings (y) calculation would be y = kx. If no production data is available for a given site, it is excluded from the tailings tonnage and geochemical CDR capacity calculations.

2.3. Geochemical CDR capacity

The inferred geochemical CDR capacity of each site is based on the method outlined in Bullock et al. (2022), which incorporates a maximum theoretical potential of a given material based on the whole rock geochemistry, coupled with shrinking core modelling that considers the starting grain size, modal mineralogy and mineral dissolution rates. The shrinking core model effectively estimates how much dissolution could be theoretically achieved in a calendar year, meaning mineral kinetics are considered alongside the chemical potential of the material. The full methodology is described in Bullock et al. (2022) and summarised here. Calculations require knowledge of the following tailings characteristics: assigned modal mineral composition of the tailings for a given site, assigned chemical composition of each mineral present, an assigned initial grain size, each mineral dissolution (weathering) rate under given conditions, amount of tailings produced annually (averaged across the period 2017-2021) for a given site, and total tailings production 2017-2021 for a given site.

Publicly reported whole rock (tailings) geochemistry and modal mineralogy information are typically not available for the majority of sites assessed here. As a result, a representative dataset has been used, with average site data for a given region designated to all sites within that area for each commodity. For instance, available minerochemical data taken from PGM and Cr-producing Union mine (Western Limb of the Bushveld Complex) has been assigned to all PGM and Cr mine sites in the Western Limb (taken from Amponsah-Dacosta, 2017), Two Rivers minerochemical data has been used as a proxy for Eastern Limb Bushveld Complex sites (from Beukes, 2014), Mogalakwena data for the Northern Limb (Amponsah-Dacosta, 2017), Kimberley data for diamond

mines in the Kimberley area (White et al., 2012), Palabora data for the Palabora copper mine and Foskor phosphate mine in the Phalaborwa area (Yuhara et al., 2005; Giebel et al., 2016), Nkomati data for the Nkomati nickel mine and Strathmore magnesite mine (Amponsah-Dacosta, 2017), Cullinan data for the Cullinan and Voorspoed sites (Hoal et al., 2009; Dongre and Tappe, 2019), and Venetia data for the Venetia mine (Stubbs, 2020; Zeyen et al., 2022a). In reality, mineralogy and geochemistry will vary from site-to-site, and even within a specific site. The method applied here aims to act as a baseline indicator for geochemical CDR potential; further case studies must be undertaken to fully realise a site's potential.

For shrinking core modelling, average dissolution rates achieved under near-neutral conditions (pH ~6-8 solution, temperatures 21-24°C) were used, taken from the compilations of Palandri and (2004)and Bandstra et al. (2008).experimentally-derived dissolution rates were determined under given temperature conditions, atmospheric CO2 concentrations and in a starting media solution of pre-defined composition and acidity. A nominal starting tailings grain size of 75 µm was selected based on some example mean particle diameters of PGM and kimberlite tailings (Humphries et al., 2006; Swami et al., 2007; Amponsah-Dacosta, 2017; Bullock et al., 2022). Volumetrically significant Ca- and Mg-bearing silicate minerals were selected based on their near-ubiquitous (>1%) modal occurrences across targeted mafic and ultramafic rocks, incorporating serpentine, olivine, orthopyroxene, clinopyroxene, amphibole, plagioclase, talc, mica, smectite and chlorite. A theoretical 1 kg of each site tailings material is used in the shrinking core model (after Hangx and Spiers, 2009; Renforth et al., 2015), where the modal mineralogy is converted to grams of mineral in the tailings, with untargeted minerals (e.g., pyrite, magnetite, apatite, quartz) excluded from calculations. The sum for all mineral phases for a given site then gives the overall potential of the material. The shrinking core model used here is:

$$X(t) = \frac{D_0^3 - (D_0 - 2W_r V_m t)^3}{D_0^3}$$
 (5)

Where X is the fractional extent of dissolution, t is dissolution time (s), D_0 is the initial particle diameter (m), W_r is the average dissolution rate (mol(mineral)/m²/s), and V_m is the molar volume of the mineral (m³/mole, converted to one mol Si basis). The maximum CDR capacity of each mineral (in gCO₂/kg material), required for the shrinking core model, is based on a calculated E_{pot} value (alkalinity production) and C_{pot} value (for mineral carbonation). The maximum theoretical potential calculations for each geochemical CDR pathway were made using the following equations:

$$E_{pot} = \frac{M_{CO_2}}{100} \cdot \left(\alpha \frac{CaO}{M_{CaO}} + \beta \frac{MgO}{M_{MgO}} + \varepsilon \frac{Na_2O}{M_{Na_2O}} + \theta \frac{K_2O}{M_{K_2O}} + \rho \frac{MnO}{M_{MnO}} + \gamma \frac{SO_3}{M_{SO_3}} + \delta \frac{P_2O_5}{M_{P_2O_5}} \right) \cdot 10^3 \cdot \eta$$
(6)

$$C_{pot} = \frac{M_{CO_2}}{100} \cdot \left(\alpha * \frac{CaO}{M_{CaO}} + \beta \frac{MgO}{M_{MgO}} + \gamma \frac{SO_3}{M_{SO_3}} + \delta \frac{P_2O_5}{M_{P_2O_5}} \right) \cdot 10^3$$
 (7)

The coefficients α , β , γ , δ , ε , ρ , and θ represent the relative contribution of each major element (wt%) oxide to geochemical CDR (Renforth, 2019), adjusted based on the modal carbonate content of the rock (Bullock et al. 2021a), M_x is the molecular mass of those oxides, and η is molar ratio of CO_2 to cation sequestered during weathering ($\eta=1.5$ to account for buffering in the seawater carbonate system; Renforth and Henderson, 2017; Renforth, 2019; Bullock et al. 2021a). Values are based on typical mineral chemistry using the formulae used by Renforth (2019) and Bullock et al. (2021a, 2022). This permits the geochemical CDR capacity to be calculated for a calendar year as both alkalinity production and carbonate precipitation. Total achievable geochemical CDR capacity for a given year (as alkalinity and carbonates) is then calculated based on the average annual tailings production across the

period 2017-2021. Cumulative CDR potential due to alkalinity generation or carbonation of 1 kg of material i for a time duration t (specific CDR; sCDR), is calculated by:

$$sCDR_i(t) = \sum_{i} f_{i,j} X_{i,j}(t) E^j_{pot}$$
(8)

where $f_{i,j}$ is the mass fraction of mineral j in material i, $X_{i,j}$ is the extent of dissolution calculated using the shrinking core model (Eq. 5) using the kinetics of mineral j and the initial particle size i, and E^j_{pot} is the E_{pot} for mineral j. For total cumulative CDR (tCDR) of material i over a given period of time or by a particular year (y) within this period, including cumulative contribution from tailings replenishment in all the years (k) up to and including year y, this is calculated by:

$$tCDR_i(y) = \sum_{k=1}^{y} sCDR_i(t_k)P_{i,k}$$
(9)

where t_k is time elapsed from year k to year y, and $P_{i,k}$ is the mass of tailing i produced in year k, and k = 1 denoting the first year of weathering (Bullock et al., 2022, 2023). The CDR achieved annually (aCDR) of material i in year y is defined by:

$$aCDR_i(y) = tCDR_i(y) - tCDR_i(y-1)$$
(10)

3. Results

3.1. Tailings production estimates for suitable sites

In total, 118 operations are identified as possible target sites for

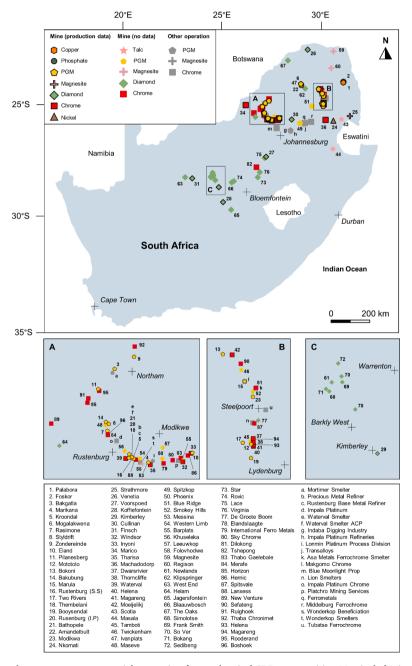


Fig. 1. Map of South African mines that may represent potential target sites for geochemical CDR opportunities. Map includes insets of highly concentrated mining areas, and mines with both available and limited production data and operational status.

Table 1
List of possible geochemical CDR implementation target sites in South Africa, based on the public directory of operating mines and quarries, agents and mineral processing plants. List includes sites with publicly available commodity production data (chosen for further calculations in this study), sites with limited or no available data, and sites that do not specifically mine materials, but may further process materials or offer similar services.

Mine sites with available production data 2017-2021				17-2021	Sites with limite	ed or no	product	ion data availab	le	Sites of related				
	Locat	ion		Main targeted commodity	Lo	ocation			Main targeted commodity				Main targeted commodi	
Site	Lat	Long	Province	•	Site	Lat	Long	Province	•	Site	Lat	Long	Province	
Windsor	-25.71	27.79	North West	Chrome	Rooderand	-25.12	27.02	North West	Chrome	Asa Metals - Ferrochrome Smelter	-24.56	30.15	Mpumalanga	Chrome
Inyoni	-25.65	27.84	North West	Chrome	Boshoek	-25.49	27.10	North West	Chrome	Makgomo Chrome	-24.50	30.06	Mpumalanga	Chrome
Machadodorp	-25.71	30.23	Mpumalanga	Chrome	Dilokong	-24.55	30.15	Mpumalanga	Chrome	Impala Platinum Chrome (Pty) Ltd	-25.54	27.18	North West	Chrome
Dwarsrivier	-24.94	30.13	Mpumalanga	Chrome	Tshepong	-27.87	26.71	Limpopo	Chrome	Platchro Mining Services	-25.70	27.75	North West	Chrome
Marico	-25.03	26.16	North West	Chrome	International Ferro Metals	-25.73	27.62	North West	Chrome	Blue Moonlight Prop	-26.09	27.74	Limpopo	Chrome
Tharisa	-25.74	27.49	North West	Chrome	Sky Chrome	-25.70	27.70	North West	Chrome	Lion Smelters	-24.82	30.12	Mpumalanga	Chrome
Helena	-25.00	30.13	Mpumalanga	Chrome	New Venture	-25.40	26.58	North West	Chrome	Wonderkop Beneficiation	-25.72	27.40	North West	Chrome
Magareng Mine			Mpumalanga	Chrome	Sefateng	-24.37	30.02	Limpopo	Chrome	Wonderkop Smelters	-25.72	27.40	North West	Chrome
Thorncliffe Mine	-24.97	30.13	Mpumalanga	Chrome	Spitsvale	-24.82	30.14	Mpumalanga	Chrome	Ferrometals Ltd	-25.86		Mpumalanga	
Waterval	-25.69	27.27	North West	Chrome	Lanxess	-25.70	27.36	North West	Chrome	Middelburg Ferrochrome	-25.80	29.49	Mpumalanga	Chrome
Moeijelikj	-24.30	29.95	Mpumalanga	Chrome	Helena	-25.00	30.13	Mpumalanga	Chrome	Tubatse Ferrochrome (Pty) Ltd	-24.72	30.23	Mpumalanga	Chrome
Palabora	-23.99	31.13	Limpopo	Copper	Magareng	-24.99	30.13	Mpumalanga	Chrome	Transalloys (Pty) Ltd	-25.89	29.12	Mpumalanga	Manganese
Venetia	-22.45	29.32	Limpopo	Diamond	Ruighoek	-25.18	26.92	North West	Chrome	Mortimer Smelter	-24.97	27.14	Limpopo	PGM
Voorspoed	-27.40	27.20	Free State	Diamond	Thaba Chronimet	-24.75	27.35	Limpopo	Chrome	Precious Metal Refiners	-25.69	27.35	North West	PGM
Cullinan	-25.70	28.50	Gauteng	Diamond	Horizon	-25.22	26.91	North West	Chrome	Waterval Smelter	-25.68	27.33	North West	PGM
Finsch	-28.37	23.44	Northern Cape	Diamond	Hernic	-25.66	27.84	North West	Chrome	Waterval Smelter - Acp	-25.68	27.33	North West	PGM
Koffiefontein	-29.43	25.01	Free State	Diamond	Thabo Gaelebale	-25.68	27.77	North West	Chrome	Rustenburg Base Metals Refinery (Pty) Ltd	-25.68	27.33	North West	PGM
Kimberley	-28.76	24.79	Northern Cape	Diamond	Merafe	-25.49	27.10	North West	Chrome	Impala Platinum Ltd	-25.54	27.18	North West	PGM
Strathmore	-25.54	31.46	Mpumalanga	Magnesite	Voorspoed	-27.40	27.20	Free State	Diamond	Impala Platinum Ltd – Refineries	-26.22	28.45	Gauteng	PGM
Nkomati	-25.75	30.62	Mpumalanga	Nickel	Elandslaagte	-28.45	24.61	Northern Cape	Diamond	Lonmin Platinum Process Division	-25.71	27.52	North West	PGM
Amandelbult	-24.12	29.00	Limpopo	PGM	West End	-28.32	23.08	Northern Cape	Diamond	Indaba Digging Industry Cc	-26.19	28.44	Gauteng	PGM
Modikwa	-24.62	30.12	Limpopo	PGM	Helam	-25.58	26.65	North West	Diamond					
Kroondal	-25.69	27.34	North West	PGM	Newlands	-28.26	24.42	Northern Cape	Diamond					
Mogalakwena	-24.05	28.97	Limpopo	PGM	Klipspringer	-24.26	29.19	Limpopo	Diamond					
Bakgatla	-24.94	27.16	Limpopo	PGM	Sedibeng	-28.12	24.48	Northern Cape	Diamond					
Marikana	-25.72	27.48	North West	PGM	Star	-28.32	26.78	Free State	Diamond					
Rustenburg (Ss)	-25.67	27.32	North West	PGM	So Ver	-28.21	24.50	Northern Cape	Diamond					
Two Rivers	-24.97	30.04	Mpumalanga	PGM	Bokang	-28.30	24.38	Northern Cape	Diamond					
Bakubung	-25.38	27.08	North West	PGM	Virginia	-28.10	26.88	Free State	Diamond					
Marula	-24.50	30.06	Limpopo	PGM	De Groote Boom	-24.80	30.17	Mpumalanga	Diamond					
Rustenburg (Ip)	-25.67	27.32	North West	PGM	Rovic	-28.49	25.56	Free State	Diamond					
Bathopele	-25.68	27.30	North West	PGM	Lace	-27.45	27.13	Free State	Diamond					
Thembelani	-25.65	27.30	North West	PGM	The Oaks	-22.96	28.31	Limpopo	Diamond					
Booysendal	-25.09	30.11	Mpumalanga	PGM	Simolotse	-28.32	24.40	Northern Cape	Diamond					
Zondereinde	-24.84	27.34	Limpopo	PGM	Jagersfontein	-29.77	25.42	Free State	Diamond					
Eland	-25.64	27.88	North West	PGM	Blaauwbosch	-28.55	25.45	Free State	Diamond					
Rasimone	-25.46	27.10	North West	PGM	Frank Smith	-28.25	24.51	Northern Cape	Diamond					
Styldrift	-25.40	27.11	North West	PGM	Folovhodwe	-25.73	27.42	Limpopo	Magnesite					
Mototolo	-24.98	30.11	Mpumalanga	PGM	Magnesite	-22.53	30.63	Limpopo	Magnesite					
Bokoni	-24.29	29.88	Limpopo	PGM	Regison	-23.31	30.52	Limpopo	Magnesite					
Pilanesberg	-25.11	27.01	North West	PGM	Twickenham	-24.42	30.03	Limpopo	PGM					
Foskor	-23.95	31.14	Limpopo	Phosphate	Ivanplats	-24.08	28.97	Limpopo	PGM					
					Phoenix	-25.69	27.60	North West	PGM					
					Blue Ridge	-25.09	29.53	Mpumalanga	PGM					
					Maseve			North West	PGM					
					Spitzkop			Mpumalanga	PGM					
					Western Limb			North West	PGM					
					Barplats			North West	PGM					
					Smokey Hills			Limpopo	PGM					

Table 1 (continued)	ontanuea)						
Mine :	Mine sites with available production data 2017-2021	ta 2017-2021	Sites with	with limited or no production data available	able	Sites of related other (non-mining) activities	S
	Location	Main targeted		Location	Main targeted	Location	Main targeted commodity
Site	Lat Long Province	— Commounty	Site	Lat Long Province	Site	Lat Long Province	ı
			Messina	-25.72 27.48 Limpopo	PGM		
			Leeuwkop	-25.60 27.61 North West	PGM		
			Tamboti	-24.96 30.10 Limpopo	PGM		
			Khuseleka	-25.62 27.26 North West	PGM		
			Scotia	-25.68 31.18 Mpumalanga	Talc		
			Masala	-27.04 30.74 Mpumalanga	Talc		

geochemical CDR opportunities (Fig. 1 and Table 1). Of this total, 21 sites are excluded from tonnage and CDR calculations as they are not dedicated sites for mining activities. These include smelter operations (e.g., Mortimer, Waterval and Lion smelters), refiners (e.g., Rustenburg base metals refinery), beneficiation sites (e.g., Wonderkop beneficiation) and other processing or associated mining service providers. Also excluded are 55 sites that were identified as currently on care and maintenance, abandoned or closed at the time of writing (see Table 1). Other excluded sites are those that have limited or no public information on their production status and tonnages post-2017. It should be stressed that while these sites are excluded from the calculations made here, many smelters and refineries produce by-product wastes such as slags and fly ashes that may be suitable for geochemical CDR purposes. Additionally, sites reported as on care and maintenance, closed, or of unknown status may be actively producing or contain historic tailings of potential use for geochemical CDR purposes. Therefore, sites are not entirely ruled out for targeted geochemical CDR strategies, but updated information is required to consider their total maximum removal potential. Similarly, abandoned asbestos mines across the country may also be considered viable sites for geochemical CDR purposes, but more data is required on their localities and historic stockpile tonnages. Sites in their development stages at the time of writing, such as Ivanplats Platreef and the Frank Smith mine, are also excluded; however, their potential contribution may develop as the sites open or re-open.

In total, 42 mines have been identified as potentially suitable sites with post-2017 operational status and publicly available production information (Table 2). This includes 21 PGM-producing sites, 11 chrome-producing sites, 6 diamond sites, 1 copper site, 1 magnesite site, 1 nickel site, and 1 phosphate site. All identified sites are geologically hosted within the Archean Kaapvaal craton (Fig. 2), which is made up of deformed greenstone belts, gneisses and granitic plutons. The majority of PGM and chrome mines are concentrated in the Paleoproterozoic Bushveld Complex (Fig. 2), comprised of layered suites of mafic and ultramafic rocks. The Bushveld Complex, made up of the Western, Northern and Eastern Limbs, contains exploitable PGM and other precious and base metal-bearing layers, typically hosted within the Merensky Reef, the UG2 Reef and the Platreef. Diamond mines are kimberlite-hosted, recognised as discrete clusters of various crustal deformation events and ages across South Africa, with major mining areas predominantly around the city of Kimberley (Fig. 2). Other diamond-hosting kimberlite cluster districts include areas of the North West, Gauteng and Limpopo provinces.

For several larger producing sites, production information is available in annual reports for the period 2017-2021 (e.g., Mogalakwena, Amandelbult, Venetia, and Rustenburg sites, see Table 2). Other sites may only contain sporadic data for one or some of the years between 2017 and 2021. In this study, it has been calculated that 764 Mt of suitable tailings were produced over the period 2017-2021, dominated by PGM (396 Mt) and diamond (188 Mt) tailings volumes. An average of 137-171 Mt of total material was generated annually for that period. The highest annual amount for an individual site (32 Mt) was calculated for the Palabora copper mine, with notably high annual tailings generation recognised for the Venetia diamond mine (up to 30 Mt), the Mogalakwena PGM mine (up to 14 Mt), Rustenburg PGM mine (up to 14 Mt) and Finsch diamond mine (up to 12 Mt).

3.2. Estimated CDR site capacity

For the 42 mines with available production data, the CDR that could theoretically be achieved in the first calendar year of weathering is 27 kt through alkalinity generation (based on mineral E_{pot} and average tailings production for a given year), and 18 kt by carbonation (mineral C_{pot}) (Table 3 and Fig. 3). Over a weathering period of 2030-2100, the average CDR achieved per year is 1.1-1.7 MtCO₂. If the accumulated five years of tailings production for all sites was targeted for geochemical CDR purposes, the total CDR achieved after one year of implementation

Table 2
Available commodity production data and estimated tailings production tonnages for 42 targeted mines in South Africa. See supplementary material for data sources. Estimated tailings production values presented in italics are values estimated based on an average commodity production to milled material ratio (see text and supplementary material).

	Locat	ion		Main targeted commodity		Commod	dity production	per year		Estir	nated ta	ilings p	roducti	on (Mt) per year	Tailings producti	on 2017-2021 (Mt
Mine site	Lat	Long	Province		2017	2018	2019	2020	2021	2017	2018	2019	2020	2021	Average per year	Total
Windsor	-25.71	27.79	North West	Chrome					0.70 Mt					1.39	1.39	2.79
Inyoni	-25.65	27.84	North West	Chrome					0.72 Mt					1.43	1.43	2.87
Machadodorp	-25.71	30.23	Mpumalanga	Chrome	0.01 Mt	0.002 Mt				0.02	0.004				0.01	0.04
Dwarsrivier	-24.94	30.13	Mpumalanga	Chrome	1.20 Mt	1.89 Mt	1.55 Mt	2.20 Mt		2.04	2.52	2.20	2.40		2.29	11.45
Marico	-25.03	26.16	North West	Chrome					0.10 Mt					0.19	0.19	0.38
Γharisa	-25.74	27.49	North West	Chrome	1.30 Mt	1.4 Mt	1.29 Mt	1.34 Mt	1.51 Mt	4.88	5.03	4.84	4.18	3.91	4.57	27.4
Helena	-25.00	30.13	Mpumalanga	Chrome		1.33 Mt					2.65				2.65	5.29
Magareng Mine	-24.99	30.13	Mpumalanga	Chrome		1.33 Mt					2.65				2.65	5.29
Thorncliffe Mine	-24.97	30.13	Mpumalanga	Chrome		1.35 Mt		1.20 Mt			2.69		2.39		2.54	7.61
Vaterval	-25.69	27.27	North West	Chrome		1.95 Mt	2.26 Mt	2.36 Mt	2.35 Mt		3.63	4.22	4.39	4.39	4.15	20.77
Moeijelikj	-24.30	29.95	Mpumalanga	Chrome	0.24 Mt	0.36 Mt	0.33 Mt	0.29 Mt	0.18 Mt	0.36	0.49	0.33	0.29	0.21	0.34	2.02
Palabora	-23.99	31.13	Limpopo	Copper		0.08 Mt	0.08 Mt	0.02 Mt	0.02 Mt		31.27	31.27	8.21	8.60	19.84	99.19
Venetia			Limpopo	Diamond	4600 k carat	4200 k carat	1900 k carat		5300	25.62			20.60		18.44	110.66
/oorspoed			Free State	Diamond	606 k carat	433 k carat				3.37	2.41				2.89	8.68
Cullinan			Gauteng	Diamond	786 k carat		1700 k carat	1500 k carat	1900	4.38	3.70	4.10	4.20	10.58	5.39	32.35
insch			Northern Cape		2149 k carat		1800 k carat		1200	11.97		3.07	3.00	3.20	4.57	27.41
Coffiefontein			Free State	Diamond	52 k carat	53 k carat	64 k carat	69 k carat	59	0.29	0.30	1.00	1.00	0.33	0.58	3.50
imberley			Northern Cape		939 k carat	914 k carat	700 k carat	1032 k carat	1100	8.02	7.97	6.00	7.41	7.80	7.44	44.64
trathmore			Mpumalanga	Magnesite	303 K curut	JI I K Curut	700 K Carac	1002 K Curut	100,000	0.02	7.57	0.00	/.11	1.00	1.00	2.00
Vkomati			Mpumalanga	Nickel	15,875 t	13,302 t	14,209 t	10,638 t	8,016	7.49	8.04	8.15	6.62	4.70	7.00	42.00
Amandelbult			Limpopo	PGM	858,000 oz	868,800 oz	893,300 oz	608,000 oz	773,000	7.00	7.00	7.10	4.50	5.90	6.30	37.80
Modikwa			Limpopo	PGM	301,228 oz	333,888 oz	306,930 oz	259,360 oz	251,755	2.01	2.43	2.29	1.94	2.05	2.14	12.86
Kroondal			North West	PGM	586,000 oz	624,000 oz	265,000 oz	197,000 oz	227,000	2.52	2.63	4.06	3.00	3.53	3.14	18.87
Mogalakwena			Limpopo	PGM				1,182,000 oz oz	1,215,000						13.76	82.56
			Limpopo	PGM	23,000 oz	1,170,000 02	1,213,000 02	308,536 oz	270,325	13.00	13.60	13.70	13.30	4.82	4.82	9.64
Bakgatla Maniltona			North West		23,000 02		508,000 oz	508,536 02 606,000 oz	765,000	2.04	2.56	6.79	9.06	4.82 10.67	4.82 6.22	9.64 37.35
Marikana			North West	PGM PGM			698,000 oz	562,000 oz	672,374	2.04			10.46		12.22	73.34
Rustenburg (Ss)					200 214	240 405	,		300,172						3.33	19.99
wo Rivers			Mpumalanga	PGM	390,214 oz	348,405 oz	313,406 oz	261,024 oz		3.50	3.46	3.40	3.02	3.28		
Bakubung			North West	PGM	177 (00	000 500	017 000	010 000	420,000	1.50	1.04	1 77	1.64	1.84	1.84	3.67
Marula			Limpopo	PGM	177,600 oz	223,500 oz	217,000 oz	210,000 oz	231,300	1.50	1.84	1.77	1.64	1.80	1.71	10.25
tustenburg (Ip)			North West	PGM	1,246,600 oz		1,390,800 oz	1,2/0,100 oz	1,334,400					10.69	10.52	63.12
Bathopele			North West	PGM		136,000 oz			220,000	1.72		1.72	0.97	3.20	1.87	11.20
hembelani			North West	PGM	100 000	000 075	167.050	010.041	160,000	3.74	3.50	3.42	3.11	1.50	3.05	18.32
looysendal			Mpumalanga	PGM	199,000 oz	229,275 oz	167,359 oz	219,941 oz	375,631	2.50	1.41	2.67	2.40	2.40	2.28	13.66
ondereinde			Limpopo	PGM	280,000 oz	299,295 oz	308,466 oz	255,574 oz	333,246	2.00	3.30	1.93	2.20	2.00	2.29	13.72
land			North West	PGM	150,000 oz	150,000 oz	150,000 oz	84,797 oz	43,664				1.06	1.52	1.29	3.88
Rasimone			North West	PGM	326,000 oz	304,900 oz	298,000 oz	271,000 oz	320,000	1.53	1.73		1.26	1.50	1.50	9.00
Styldrift			North West	PGM	56,400 oz	123,300 oz	196,000 oz	264,500 oz	290,550	2.45	2.30	2.22	2.07	2.30	2.27	13.62
Mototolo			Mpumalanga	PGM	184,800 oz	287,700 oz	242,300 oz	223,600 oz	250,000	0.95	1.55	2.32	2.88	2.90	2.12	12.73
Bokoni			Limpopo	PGM					300,000					1.92	1.92	3.84
Pilanesberg			North West	PGM	133,000 oz	150,744 oz	129,942 oz	128,750 oz	150,000		3.90	3.79		3.50	3.73	14.92
oskor	-23.95	31.14	Limpopo	Phosphate				2.00 Mt	2.05				0.49	0.50	0.49	1.48

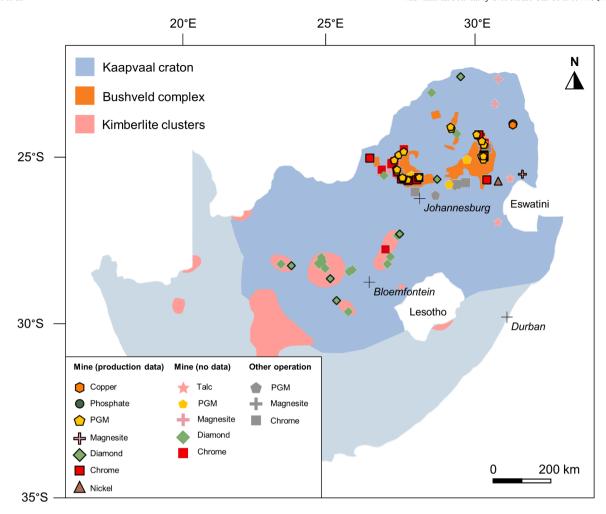


Fig. 2. Simplified geological setting of targeted mine sites and operations, with a high concentration of sites in the Bushveld Complex and at various kimber-lite clusters.

is 11-17 Mt. Finally, the total CDR that may be achieved over 2030-2100, considering all annually produced tailings can be made available to weather in this period (and assuming the average tailings production rate of 2017-2021 continues up to 2100), is 79-119 MtCO₂. Due to the higher modal abundances of more reactive minerals (e.g., serpentine), kimberlite-hosting diamond mines such as Cullinan (497 ktCO2 per year through alkalinity generation, averaged across period 2030-2100), Voorspoed (267 MtCO₂ per year), Kimberley (222 ktCO₂ per year), Finsch (136 ktCO₂ per year) and Venetia (107 ktCO₂ per year) mines hold the highest annual capacities of the selected mines. Despite containing relatively lower abundances of reactive minerals compared to kimberlite-hosting operations, the Palabora copper mine, Nkomati nickel mine and Mogalakwena PGM mine also host high geochemical CDR potential (208 ktCO2, 38 ktCO2 and 33 ktCO2 per year respectively), mainly due to their high production tonnages. The Koffiefontein diamond mine, and Amandelbult and Rustenburg large PGM operations, average removal capacities of ~13-17 ktCO2. Low tailings production tonnage PGM and chrome operations generally show the lowest geochemical CDR potentials (<10 ktCO₂ per year).

4. Discussion

Based on their favourable (Ca-Mg-containing) modal mineralogies, sites targeting diamond, PGM, chrome, nickel, phosphate, copper, talc, manganese and magnesite commodities are considered here to be viable tailings producers for possible geochemical CDR approaches (Fig. 1 and Table 1). The overall annual capacity of South African suitable mine tailings for geochemical CDR at 42 sites (Table 2), averaged across an

operational period of 2030-2100, is calculated here as 1.1 Mt (via carbonation) to 1.7 Mt (via alkalinity production) CO2 per year (based on average annual tailings production for the period 2017-2021; Fig. 3 and Table 3). The achievable CDR in one year based on one year of tailings production may contribute 0.3-0.4% offsets of emissions (based on 2020 emission value of 435 MtCO₂), or 0.3-0.5% contributions toward national reduction targets (350-420 MtCO₂) (Table 4). The achievable CDR in one year based on utilising all 2017-2021 tailings production (CDR capacity ~11-17 Mt) may contribute 2.5-3.8% of emission offsets, or 2.6-4.7% of reduction targets (Table 4). Though contributions are less than 5%, this may be critical for South Africa to improve their climate action classification status, particularly as the country approaches being 1.5°C compliant. Any additional reduction methods (i.e., beyond typical methods already being implemented, such as increasing use of renewables and reducing coal reliance) may be challenging to determine or ascertain leading up to 2030 targets and beyond.

It should be stressed that the shrinking core model utilised here applies a single 'typical' representative grain size for calculations (in this case, 75 μm). However, in reality, tailings show a wide distribution of grain sizes, which may reduce reaction time by $\sim\!30\%$ (Renforth, 2012), depending on the skew of the distribution. Furthermore, some tailings that target metals such as Cr, Cu and Ni may show a higher average grain size of $\sim\!200~\mu m$ (Rodríguez et al., 1998; Das 2015; Hu et al., 2017). Considering an average grain size of 200 μm for the shrinking core model here reduces the overall CDR by, on average, 55% (Table 4), which in turn reduces the average offset based on average annual tailings production to 0.1-0.2%, or 1.1-2.1% for historic tailings. Overall,

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Table 3
Estimated geochemical CDR capacity of chosen targetable sites in South Africa, based on an assigned regional geochemistry, modal mineralogy, and annual average or five year total production estimates for the period 2017-2021.

Location				Main targeted commodity	CDR achieved									
					Annual CDR, averaged ove (ktCO ₂)	•	Total CDR over 2030-21 tailings made available t	*	Additional CDR from wear tailings production over (ktCO ₂	period 2030-2100				
Mine site	Lat	Long	Province		Via alkalinity production	Via carbonation	Via alkalinity production	Via carbonation	Via alkalinity production	Via carbonation				
Windsor	-25.71	27.79	North West	Chrome	1.7	1.1	0.12	0.08	6.7	4.4				
Inyoni	-25.65	27.84	North West	Chrome	1.7	1.1	0.12	0.08	6.9	4.5				
Machadodorp	-25.71	30.23	Mpumalanga	Chrome	0.02	0.01	0.0015	0.001	0.2	0.1				
Dwarsrivier	-24.94	30.13	Mpumalanga	Chrome	4.7	3.1	0.34	0.22	47.4	31.4				
Marico	-25.03	26.16	North West	Chrome	0.2	0.1	0.02	0.01	0.9	0.6				
Tharisa	-25.74	27.49	North West	Chrome	5.5	3.6	0.39	0.26	65.5	43.2				
Helena	-25.00	30.13	Mpumalanga	Chrome	5.5	3.6	0.39	0.26	21.9	14.5				
Magareng	-24.99	30.13	Mpumalanga	Chrome	5.5	3.6	0.39	0.26	21.9	14.5				
Thorncliffe	-24.97	30.13	Mpumalanga	Chrome	5.3	3.5	0.37	0.25	31.5	20.9				
Waterval	-25.69	27.27	North West	Chrome	5.0	3.3	0.35	0.23	49.6	32.7				
Moeijeliki	-24.30	29.95	Mpumalanga	Chrome	0.7	0.5	0.05	0.03	8.4	5.5				
Palabora	-23.99	31.13	Limpopo	Copper	208	138	14.77	9.78	1941	1284				
Venetia	-22.45	29.32	Limpopo	Diamond	107	70	7.59	4.98	1211	794				
Voorspoed	-27.40	27.20	Free State	Diamond	267	178	18.93	12.62	1477	984				
Cullinan	-25.70	28.50	Gauteng	Diamond	497	331	35.31	23.53	5503	3667				
Finsch	-28.37	23.44	Northern Cape	Diamond	136	91	9.68	6.44	1508	1004				
Koffiefontein	-29.43	25.01	Free State	Diamond	17	12	1.23	0.82	193	128				
Kimberlev	-28.76	24.79	Northern Cape	Diamond	222	148	15.75	10.49	2456	1635				
Strathmore	-25.54	31.46	Mpumalanga	Magnesite	5.4	3.6	0.38	0.25	20	14				
Nkomati	-25.75	30.62	Mpumalanga	Nickel	38	25	2.67	1.77	430	285				
Amandelbult	-24.12	29.00	Limpopo	PGM	15	10	1.07	0.71	180	119				
Modikwa	-24.62	30.12	Limpopo	PGM	4.4	2.9	0.32	0.21	53	35				
Kroondal	-25.69	27.34	North West	PGM	3.8	2.5	0.27	0.18	45	30				
Mogalakwena	-24.05	28.97		PGM	33	22	2.34	1.54	394	260				
·	-24.05 -24.94	28.97	Limpopo	PGM	5.8	3.8	0.41	0.27	23	260 15				
Bakgatla Marikana	-24.94 -25.72	27.16	Limpopo North West	PGM	5.8 7.4	3.8 4.9	0.53	0.27	23 89	15 59				
	-25.72 -25.67	27.48	North West	PGM	7.4 15	4.9 9.7	1.04	0.35	175	116				
Rustenburg (Ss)														
Two Rivers	-24.97	30.04	Mpumalanga	PGM	6.9	4.6	0.49	0.32	83	55				
Bakubung	-25.38	27.08	North West	PGM	2.2	1.5	0.16	0.10	8.8	5.8				
Marula	-24.50	30.06	Limpopo	PGM	3.5	2.3	0.25	0.17	42	28				
Rustenburg (Ip)	-25.67	27.32	North West	PGM	13	8.3	0.89	0.59	151	99				
Bathopele	-25.68	27.30	North West	PGM	2.2	1.5	0.16	0.10	27	18				
Thembelani	-25.65	27.30	North West	PGM	3.7	2.4	0.26	0.17	44	29				
Booysendal	-25.09	30.11	Mpumalanga	PGM	4.7	3.1	0.34	0.22	57	37				
Zondereinde	-24.84	27.34	Limpopo	PGM	2.7	1.8	0.19	0.13	33	22				
Eland	-25.64	27.88	North West	PGM	1.5	1.0	0.11	0.07	9.3	6.1				
Rasimone	-25.46	27.10	North West	PGM	1.8	1.2	0.13	0.08	22	14				
Styldrift	-25.40	27.11	North West	PGM	2.7	1.8	0.19	0.13	33	21				
Mototolo	-24.98	30.11	Mpumalanga	PGM	4.4	2.9	0.31	0.21	53	35				
Bokoni	-24.29	29.88	Limpopo	PGM	4.0	2.6	0.28	0.19	16	11				
Pilanesberg	-25.11	27.01	North West	PGM	4.5	2.9	0.32	0.21	36	24				
Foskor	-23.95	31.14	Limpopo	Phosphate	5.1	3.4	0.36	0.24	29	19				

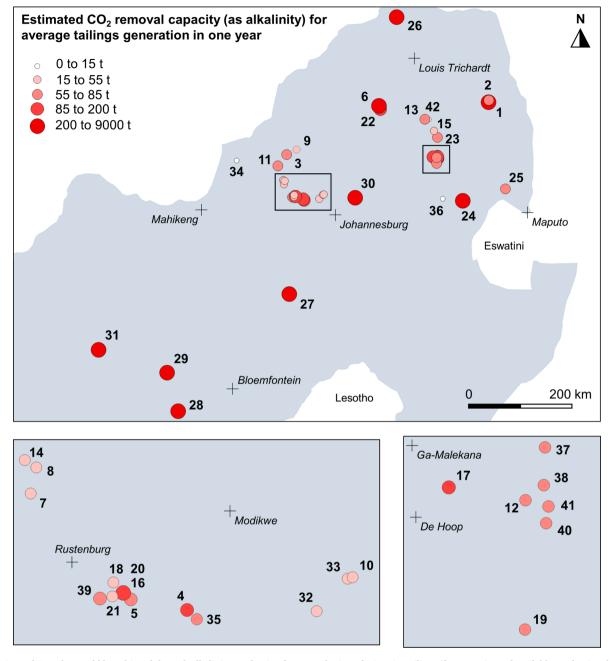


Fig. 3. Estimated CDR that could be achieved through alkalinity production from weathering of mine site tailings (for operations of available production data) over a calendar year, based on geochemical data and shrinking core modelling. Insets for concentrated mines northwest of Johannesburg (bottom left) and north east of Johannesburg (bottom right) shown.

the sCDR calculated here for grains of 75 μm is, on average, 2-3 times higher than grains of 200 μm , for a given W_r of the minerals present. Increasing W_r by up to one magnitude (realistically achievable by means of enhanced weathering to reduce pH of host media, for instance) can also have a positive effect on CDR achieved over a given time. For a grainsize of 75 μm , an increase of W_r by one magnitude for all minerals present results in an increase of sCDR by, on average, 5-10 times, or 7-10 times for grains of 200 μm . Further magnitudinal increases will have greater effects on CDR.

The CDR tonnages presented here are limited to 42 sites of publicly available commodity production information for the period 2017-2021 (out of 118 listed sites), stressing the need for more production (commodity or tailings) information for more closer to reality calculations. The compilation also does not include sites that are not listed in the public directory of operating mines and quarries, agents and mineral

processing plants, which may include newly developed operations. Inferring that there are many more unlisted suitable sites, mine sites with no available information and sites where only limited information exists for the period 2017-2021, it can be assumed that the true geochemical CDR capacity of mine tailings in South Africa is much greater than the values presented here. Furthermore, sites that host accessible pre-2017 historic tailings will have a greater tonnage of materials available for any implemented CDR scheme, coupled with the potential utilisation of any abandoned operations such as historic asbestos mines (almost 6000 recorded abandoned mines; Cornelissen et al., 2019). Finally, it should also be considered that the geochemical CDR calculation and shrinking core modelling utilised in this study does so with published mineral dissolution rates that were achieved at near-neutral conditions (pH ~6–8 solution, temperatures 21–24°C). Although it assumes that all materials are subjected to weathering for

Table 4 Total and select site examples of the calculated total CDR achievable over the 2030-2100 period, from all tailings made available to weather, for a typical grain size of 75 μm and 200 μm .

			00 period, from all tailings weather (MtCO ₂)
		Via alkalinity production	Via carbonation
Across all sites	75 μm	119	79
	200 μm	51	34
Site examples			
Cullinan	75 μm	35.3	23.5
	200 μm	14.6	9.7
Palabora	75 μm	14.8	9.8
	200 μm	6.1	4.1
Venetia	75 μm	7.6	5.0
	200 μm	3.2	2.1
Kimberley	75 μm	15.7	10.5
	200 μm	6.5	4.3
Two Rivers	75 μm	0.5	0.3
	200 μm	0.2	0.2
Nkomati	75 μm	2.7	1.8
	200 μm	1.1	0.8
Marikana	75 μm	0.5	0.3
	200 μm	0.3	0.2
Mogalakwena	75 μm	2.3	1.5
	200 μm	1.2	0.8

one calendar year for alkalinity production and carbonation, which has many practical considerations, it also assumes that no deliberate efforts have been made to speed up mineral dissolution reactions, such as utilising heat, more acidic conditions, microbes or concentrated CO₂ (Chen et al., 2006; Harrison et al., 2013; McCutcheon et al., 2015; McCutcheon et al., 2016; Power et al., 2016; Hamilton et al., 2020) some of which may be harnessed at mine sites. If such enhanced weathering methods were implemented, the increase in mineral dissolution rates (or precipitation rates if methods to precipitate carbonates were targeted), more CDR could theoretically be achieved annually.

This compilation reinforces the suggestion that geochemical CDR potential in South Africa is high. However, several mineral kinetic, energetic, environmental, societal, and engineering constraints mean the theoretical maximum potential is unlikely to be realistically fully achieved, though there is still an incentive to realise the highest possible CDR amount possible. To fully understand the maximum possible capacity, further qualitative and quantitative information on all active and inactive sites is required. Additional capacity could also be sought by including other favourable materials for ex-situ geochemical CDR approaches, such as slags, fly ashes and construction wastes, or through a detailed assessment of possible in-situ implementation sites for subsurface injection of CO2 (Tibane et al., 2021a, b; Dhansay et al., 2022). Regarding ex-situ geochemical CDR opportunities utilising suitable mine tailings, the best opportunities for future strategies identified here for South Africa include the kimberlite-exploiting diamond mines (Cullinan, Voorspoed, Kimberley operations, Finsch and Venetia), Palabora copper mine, Nkomati nickel mine and Mogalakwena PGM mine. Venetia and Voorspoed have been studied for their geochemical CDR potential (e.g., Mervine et al., 2018; Stubbs et al., 2022; Zeyen et al., 2022a), with industry-led projects such as Project CarbonVaultTM continuing to assess pilot study and implementation possibilities (Paulo et al., 2021; Wang et al., 2022; Zeyen et al., 2022b; Zhu et al., 2022). Similarly, PGM mines in the Bushveld Complex are currently subject to CDR investigations from academic projects such as the GGREW Project (Bullock et al., 2021b), and the SAT4CCS Project (Nkosi, 2016; Nkosi, 2018).

Other key opportunities may also exist with newly developed sites, such as the Ivanplats PGM project in the Northern Limb (Mining Weekly, 2022), or sites that may undergo re-development, such as the Blaauwboch and Frank Smith diamond mines (SAHRA, 2019; SSC Diamond, 2022). Sites at the development or re-development stages may be optimal for assessing existing and future tailings resources, and

exploring possible engineered options for geochemical CDR implementation, such as by field spreading of materials (e.g., across croplands; Taylor et al., 2016; Andrews and Taylor, 2019; Beerling et al., 2020; Kelland et al., 2020; Smet et al., 2021; Cipolla et al., 2022) or through reactor system pilot schemes (e.g., within processing or wastewater treatment plants; Darton et al., 2022; Xing et al., 2022). Due to practicality considerations for any on-site CDR implementation, it is likely that most operators would opt to pursue methods that result in CDR through mineral carbonation, as on-site solid material storage is more manageable for mine sites (solid carbonate products could be incorporated into the standard solid waste management, with storage in stockpiles and dams) compared to effluent alkalinity production. Though alkalinity production removes more CO2 than mineral carbonation (precipitation returns some CO₂ to the atmosphere, see Eqs. 3 and 4), both processes result in net CDR for non-carbonate starting materials. Additionally, the ecological and environmental impacts of large discharges of alkalinity to local river systems, groundwater and the ocean are subject to ongoing investigation (e.g., Bach et al., 2019; Burt et al., 2021; Butenschön et al., 2021). There may also be opportunities for sale or re-use of carbonation products, and carbonate products would require minimal post-storage environmental monitoring (Lackner et al., 1995; Sipilä et al., 2008; Power et al., 2013; Boot-Handford et al., 2014).

Though this study emphasises the high potential for geochemical CDR for mine tailings in South Africa, and possible opportunities and benefits open to operators, there is a need to keep such suitable tailings production and CDR capacity information available and up to date for future targeted strategies. This study provides the structural basis for any approach toward cataloguing the country's CDR potential. Still, for more detailed site-by-site information and life cycle analyses, there is a requirement for stronger links and collaboration between policymakers, academia and industry. This includes tailings production tonnages (or estimates based on commodity production and milled volumes) for sites that exploit suitable host rocks, accessible historic stockpile volumes, geochemical data and mineralogical information for each tailings producing site. Such approaches, either as academic studies, pilot scheme assessments or possibly as a newly implemented policy requirement for annual site CDR assessment, could be the next steps for South Africa to become a global leader in geochemical CDR approaches with mine tailings and other industrial wastes. There is also scope for other countries of similar ex-situ resource opportunities to follow this appraisal method, cataloguing materials for future target site opportunities.

5 Conclusions

South Africa hosts and produces hundreds of millions of tonnes of tailings that exhibit suitable bulk geochemistry and mineralogy for exsitu geochemical CDR strategies, particularly in their kimberlitehosted diamond mines, Bushveld-hosted PGM and chrome mines, and other mafic-ultramafic hosted nickel, phosphate, copper, talc and magnesite mines. The average annual CDR capacity of 42 suitable mine sites is 1.1 to 1.7 MtCO₂ for annually produced tailings, averaged across a weathering period of 2030-2100, with additional removal of 11-16 MtCO2 achievable for tailings produced over a period of 2017 to 2021. These tonnages, or accumulated total CDR of up to 119 MtCO2 over the entire period, can contribute to required and targeted national CO2 offsets and reductions, particularly as more novel methods to achieve further reductions become more difficult to find. There are also numerous mine sites of limited public information that produce minerochemically similar tailings appropriate for CDR, as well as historic tailings (in active and inactive sites), and slag and fly ash producers that could be utilised for targeted strategies. Diamond mines at Cullinan, Voorspoed, Kimberley, Finsch and Venetia, the Palabora copper mine, Nkomati nickel mine, and Mogalakwena PGM mine, hold notably high potential, and warrant more detailed investigations. While the presented calculations here are relevant for the years immediately preceding the period 2017 to 2021, there is an incentive to explore collaboration and

policy making opportunities to constantly report tailings production tonnages and/or geochemical CDR capacities of mine sites for future options, including through site-by-site studies and public reporting of commodity production and milled material/tailings production tonnages over the remainder of the century. The methods applied here are also applicable to other countries that host similarly suitable materials for ex-situ geochemical CDR implementation options.

CRediT authorship contribution statement

Liam A. Bullock: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization, Funding acquisition. Zakhele Nkosi: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – review & editing, Visualization, Funding acquisition. Mpho Vele: Validation, Formal analysis, Data curation, Writing – review & editing. Maxwell Amponsah-Dacosta: Conceptualization, Methodology, Validation, Resources, Data curation, Writing – review & editing, Funding acquisition.

Declaration of Competing Interest

The authors have no competing interests to declare.

Data availability

Data will be made available on request.

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