

Assessment of carbon dioxide sequestration potential of ultramafic rocks in the greenstone belts of southern India

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The growing universal concern about anthropogenically induced climate change is resulting in the development of strategies that can reduce or at least slow down the build-up of greenhouse gases in the atmosphere. Disposing of the excess carbon dioxide (CO₂) by capturing it from industrial sources, separating it from flue gases and storing into potential geological reservoirs is emerging as a new technology for mitigating the detrimental effects of emissions and is referred to as carbon sequestration. An effective geochemical trapping system for storing the CO₂ underground is mineral carbonation. It is based on the weathering/alteration processes occurring in nature, wherein CO₂ reacts with Ca, Mg and/or Fe-bearing silicate-rich rocks such as ultramafics and mafics to form the respective carbonates. Under controlled experimental conditions with optimized reaction kinetics, mineral carbonation has considerable potential for the safe disposal of CO₂ in the form of environmentally benign carbonates. India has about twenty-five major greenstone belts with maximum thickness up to

10 km and lithologies containing Ca, Fe and Mg silicate-rich minerals such as olivine (Mg, FeSiO₄), serpentine (Mg₃Si₂O₅(OH)₄), pyroxene (Mg, FeSiO₃), etc. In this article, the mineral carbonation potential of ultramafic rocks of the greenstone belts of the southern Indian Peninsula is considered. The alkaline silicates exist in abundance in the greenstone belts and may act as possible sinks to sequester CO₂ in the form of magnesium, iron or calcium carbonates. The distribution of ultramafic rocks in southern India is noted and the approximate amounts of CO₂ that can be sequestered in the two greenstone belts of the southern Indian Peninsula, namely Kolar and Chitradurga is estimated. The areal extent of the Kolar belt is about 320 sq. km with an average width of ~6 km, while that of Chitradurga belt is 6000 sq. km and maximum depth determined is up to 10 km. Estimates show that an ultramafic portion of 1 km³ in the Kolar belt can store ~2.94 million tonnes (mt) of CO₂ and that in the Chitradurga belt ~4.7 mt, which accounts for about 0.6% of annual CO₂ production in India.

Keywords: Carbon dioxide, mafic/ultramafic rocks, mineral carbonation, sequestration.

THE burning of fossil fuels and other anthropogenic activities have increased the atmospheric carbon dioxide (CO₂) concentrations^{1,2} from 315 ppm in the year 1958 to about 378 ppm at the end of 2004. Perturbation in the global carbon cycle over the past century has exerted a discernable influence on the global climate change, leading to warmer temperatures, increased ice melts, especially in the polar regions and rise in sea levels³. It is now evident that the rise in CO₂ concentration within the atmosphere is one of the main causes for the apparent rise in the average global temperature. India, being a fast developing country with high growth rate of industrial development, is experiencing a dramatic rise in fossil-fuel CO₂ emissions and has become the world's fourth largest CO₂ emitter in the world⁴. The rising CO₂ levels resulting in climate change may affect the prevailing weather, river

basins, rainfall, coastal areas, ecosystem and forestry in India. With the Indian population being more dependent on climate-sensitive factors like agriculture and forestry for sustenance, India has strong reasons to be concerned about the impact of increased CO₂ emission. This problem can be partly resolved by disposing of the excess CO₂ by capturing it from the point sources, separating it from flue gases and storing into potential reservoirs other than the atmosphere, which is referred to as carbon sequestration^{5,6}. The potential reservoirs include terrestrial biosphere, oceans and geological formations. Figure 1 shows the various geological sequestration options for storing the CO₂ underground⁷. The geological formations like deep saline aquifers, depleted oil and gas fields, unmineable coal seams, oil-bearing shales, mafic/ultramafic rocks and especially the continental flood basalts seem to be promising options for the long-term sequestration of CO₂ compared to other reservoirs⁸. To a certain extent, oceans can also contribute in CO₂ storage, but certainties about the environmental impact on marine life are controversial because of the changes in ocean water pH.

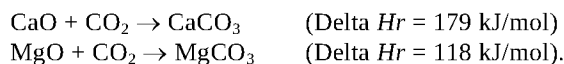
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period of time, CO_2 would dissolve in the interstitial solu-

Saline aquifers: Deep saline aquifers (depth ≥ 800 m) provide pressures high enough to keep the supercritical

Depleted oil and gas field: The depleted fields are reasonably good stratigraphic traps for fluids and gases and can be utilized as efficient CO₂ sequestration sites¹³. Injection of CO₂ into depleted oil fields is a commercially proven process used for enhanced oil recovery¹⁴.





Finally, the potential of the technology to store appreciable amounts of the CO_2 resulting from fossil fuel combustion is large enough because serpentine, olivine, and pyroxene-rich rocks occur in large amounts in nature^{18,19}. The utilization and remediation of by-products of carbonation, like silica, can be achieved for use in glass making, and magnesite deposits have their own importance in steel and cement industries. Experimentally, in a carbonation reaction, CO_2 can be made to react with the silicate minerals under controlled conditions or transporting CO_2 from an anthropogenic source such as a coal-fired thermal plant, separating it from the flue gas mixture and pumping the same into mafic/ultramafic rock formations. For industrial applications, the process is largely to be completed in hours compared to the natural weathering reactions, which take considerable time. Thus optimization of reaction kinetics is of prime importance in mineral carbonation^{20,21}. Olivines are slow to react and serpentines react poorly, unless pretreated to remove chemically bound water. At a high temperature of $\sim 600^\circ\text{C}$ and pressure of about <0.5 kbar, the reaction has favourable conditions for kickstarting the carbonation pathways²¹. Results indicate that under suitable conditions of pressure and temperature, CO_2 mineral trapping capacity after 100,000 years reaches about ~ 90 kg/cubic m in a sandstone shale system²².

Ultramafic abundance in greenstone belts of the southern Indian Peninsula

Southern India is represented by the well-studied greenstone belts of Dharwar Craton along with the Nellore schist belt and the partly preserved Khammam schist belt^{23,24}. Greenstone belts with mafic and ultramafic lithologies occurring in this cratonic block are illustrated in Figure 2 (ref. 24) and Table 1 (ref. 25). The Dharwar cratonic block is divided into the eastern and western blocks^{24,26}. Eastern Dharwar has an areal extent of 375,000 sq. km with nine major greenstone belts and the maximum thickness determined²⁷ is 6–8 km. The Kolar schist belt is one of the best-studied belts, extending for a length of 80 km in the north–south direction and having an average width²⁴ of 6 km. The Kolar belt is divided into the eastern and western parts with respect to a central ridge, which is made up of fine-grained metavolcanic units. The belt consists of two suites of tholeiitic and komatiitic lavas subjected to amphibolite facies metamorphism. Other belts of similar type in the eastern Dharwar Craton are Pennar–Hagari, Hutti–Maski, Manglur, Hungund–Kushtagi and Raichur–Deodurg belts²⁴. The stratigraphic succession for the Kolar belt²⁸ is shown in Table 2. The pattern of lithology is consistent with vertical slabs of igneous rocks extending to great depths²⁷.

The western Dharwar cratonic block has an areal extent of 225,000 sq. km and about 20 greenstone belts are present²⁷ with the maximum thickness of 12–15 km. In addition to the older Sargur Group, lithostratigraphically the entire succession has been divided into two main groups, the Bababudan Group occupying a basal position and the Chitradurga Group occupying an upper position with the gneissic complex forming the basement^{24,26} (Table 3). The western Dharwar has three basins, of which Shimoga basin is the largest and extends for about 250 km from the south of Bababudan to Belgam in the north. Important belts present here are Bababudan, Kudremukh and Shimoga²⁴. The Sandur Basin covers an area of 960 sq. km and is characterized by well-developed mafic felsic magmatism and sedimentary rocks²⁴. The Chitradurga Basin is elongated in the form of a narrow belt of 450 km length with an areal extent of 6000 sq. km and a maximum thickness^{24,27} up to 10 km. The eastern margin of this belt is marked by prominent tectonized zone. The belt also has a western arm near Kibbanahalli from which high Mg basalts (komatiites) with spinifex texture have been

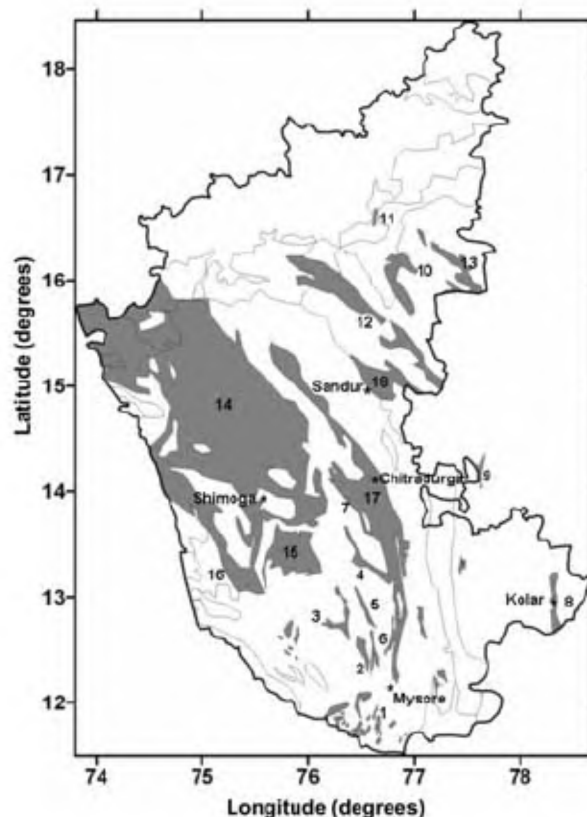


Figure 2. Greenstone belts of Southern India (modified after Radhakrishnan and Vaidyanadhan²⁴). 1, Sargur; 2, Krishnarajpet; 3, Holenarasipur; 4, J. C. Pura; 5, Nuggihalli; 6, Kalyadi; 7, Gattihosahalli; 8, Kolar; 9, Pennar–Hagari; 10, Hutti–Maski; 11, Manglur; 12, Hungund–Kushtagi; 13, Raichur–Deodurg; 14, Shimoga; 15, Bababudan; 16, Kudremukh; 17, Chitradurga, and 18, Sandur.

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Table 1. Greenstone belts of southern India (compiled after Swami Nath and Ramakrishnan²⁵)

Greenstone belts	Ultramafic/mafic lithology	Locality
Sargur	Serpentinized ultramafites with metapyroxinite Portions of peridotite Serpentinized dunite, peridotite, metapyroxinite Bands of serpentinite, steatite, talc-chlorite and pyroxinite Layered gabbro-anorthosite	Gopalpura Mavinahalli Sindhuvalli–Talur, Dodkanya Hampapura–Jayapura, Nanjangud Hullahalli
Nuggihalli	Prominent mafic–ultramafic complexes Dunites, gabbros and ultramafites Metaperidotite Metapyroxinite Prominent dykes of meta-anorthosite Magnetite gabbro and titanomagnetite	Tagadur Byrapur, Tagadur, Ponnasamudra, Bhaktarahalli, Gobbalihalli East of Belagumba Gobbalihalli North of Jambur Ranganabetta, Belagumba, Nuggihalli and Bhaktarahalli
Krishnarajpet	Ultramafites and ultrabasic rocks Serpentinites associated with dunites	Nagamangala, Hasan East of Bellibetta, Vallekotte Koppal, Sindhughatta, Aichanahalli Settinayakan and Katharighatta
Bababudan	Bababudan Group Kalsapura Formation Basic rocks as associated volcanics, amygdular amphibolites Allampur Formation Metapyroxinite and metagabbro Santaveri Formation Major volcanic piles, meta-andesites Rhyolitic to rhyodacitic volcanics, silicic volcanics Chitradurga Group As ultramafic intrusives, serpentinites with carbonate rocks	Southern part of belt Allampur Lingadahalli and Santaveri Sakunagiri Hill, Galipuje Lakkavalli, Kundur–Kavalapura
Holenarsipur	Sargur Group Ultramafic–mafic complexes with serpentinite after dunite peridotite, metapyroxinite Peridotite, hornblende Epidotes Amphibolites of ultramafic–mafic complexes Bababudan Group Modulgudda Formation Metabasalts and amphibolites Pillow lavas Metabasites of green schists consisting of epidotes Mallappanabetta Formation Metaultramafites	Yenneholeranganabetta and Bantratalal Sarvanur, Dodda Kadanur, Tirumalapur Bantratalal and Mangalapura, Near Ranganahalli, Bantratala Maddibetta hills Modulgudda Hills, Chigaranahalli Hill and Mallappanabetta arm, west of Tavanandi, Hariharapur Modulgudda Hills and NE of Kamasamudra NW of Dumgere and Dandiganahalli
Sigegudda	Metagabbro Mafic–ultramafic sequence	Along Hasan Belur Road Along Ishwarahalli Medarahalli
Western Ghats	Hornblende and ultramafics Calc silicate rocks Ultramafite with talc, chlorite, tremolite and calcite Amygdular metavolcanics with amphibolites, metagabbro Olivine dolerite Gabbro with pyroxenite Serpentinite	Sringeri Sampagimane Sampagimane, Anashebail, Nellikere, Navara, Nuralvattu, Daregudde, Poradi Malige, Hadangaja, Manipura, Kekrodi, Pambaddettu, Aladangadi Kudremukh, Ballarayandurga, Malvadi, South of Bilegal, Malige, Bakalgudde Kakkarguda Kempadi, Gundi, Padangani and Neralakatte Between Bella and Kandur

(Contd.)

Table 1. (Contd.)

Greenstone belts	Ultramafic/mafic lithology	Locality
Chitradurga	Sargur Group	
	Ultramafites and amphibolites	Ghattihosahalli belt
	Ultramafites with magnetite–chlorite rocks	Javanahalli belt
	Bababudan Group	
	Metabasalts	Kibbanahalli and Mayakonda–Madadkere belt, Halekal, Yadiyur, Karighatta
	Chitradurga Group	
	Ingaldhal Formation	
Shimoga	Basic volcanic rocks	Main Chitradurga arc
	Hiriyur Formation	
	Basic volcanic rocks, mainly andesitic to basaltic	Bellari, Mallappanahalli, Maradihalli
	Sargur Group	
	Ultramafites represented by tremolite–actinolite–talc–chlorite schists, metapyroxinite	Tarikere Valley
	Metaultramafites	Lokikere Enclave
	Peninsular Gneiss	
	Numerous enclaves of ultramafic and mafic rocks with subordinate meta sediments	Tarikere–Channagiri, Honnali
	Chitradurga Group	
	Jhandimatti Formation	
	Serpentine, ultramafic schists	Gajanur, Ubrani, Shivani and Sulekere areas
	Medur Formation	
Kolar	Thin pile of basic and intermediate volcanics	Medur, near Shikaripura
	Ranibennur Formation	
	Silicic volcanics	Amballigolla–Choradi area, Basavapatna–Daginkatte near Kumudavati River section, NE of Shikaripura
	Ultramafics, Calc-silicates	Near Sakarasanahalli, Betrayaswami Konda
	Actinolite quartzite	Near Kudarasanahalli
	Mafic amphibolite	Near Kamsamudra, Bodgurki, Vareadpur and Harohalli
	Yerrakonda Formation	
	Metapyroxinite, metagabbro	Yerrakonda Hill
	Metabasalts, Kolar Gold Field Volcanics (KGF), magnesia hypersthene basalts, basaltic andesites, meta gabbro	Badmakanahalli, Madamangala and Dodduru–Karapanahalli
	Basalts with pyroxenes	Byatarayanahalli,
	Basaltic andesites	KGF, Kamsamudra and Kempinkote mines

Table 2. Stratigraphy of Kolar belt (modified after Srinivasan and Sreenivas²⁸)

6. Bisattam and Patna granites
5. Migmatitic gneisses (mainly along the margins of the belts)
4. Banded ferruginous quartzites
3. Champion gneiss
2. Kolar amphibolite series (pillow lavas grading to ultrabasic flow)
----- Unconformity -----
1. Metamorphosed and granitized orthoquartzites, shales and limestones of Sakarsanite series (Sargur Group)

reported^{29–31}. The mafic–ultramafic supracrustal rocks underlying the basal unconformity marked by a thin but persistent oligomict–quartz–pebble–conglomerate horizon are placed under the Sargur Group^{30,31}. In addition, other important linear belts are well exposed at Sargur, Holenarsipur, Nuggihalli, Krishnarajpet, Sasivala, Ghattihosahalli and Belavadi^{26,31}.

Quantification of ultramafic rocks for CO₂ sequestration

The quantification of ultramafic rocks for their carbon storage capacity has been done taking into consideration

the weight percentage of MgO^{18,32}. Detailed information about the geology and structure of ultramafics and data on their areal distribution, approximate thickness, chemical composition and mineralogy are utilized to calculate the volume of ultramafic rocks in Kolar and Chitradurga schist belts and multiplied by wt% of MgO to assess the quantity of ultramafic rocks needed to sequester CO₂ in these two belts. The experimental results^{18,32} show that 1 tonne of serpentine (38–45 wt% MgO) can dispose of 1½ tonnes of CO₂ and thus about a tonne of wt% MgO can sequester about a tonne of CO₂. With an emission rate of 333 million metric tonnes for the year 2002 in the Indian context, the quantity of CO₂ which can be sequestered in

Table 3. Stratigraphy of Dharwar (modified after Swami Nath and Ramkrishnan³⁰)

Dharwar Supergroup	
Chitradurga Group	Hiriyur Formation Greywacke–argillite suite with volcanics, pyroclastics cherts and polymict conglomerates Ingaldhal volcanics Basic volcanics and pyroclastics acid volcanics, cherts and phyllites Vanivilas Formation Iron and manganese formations, limestones, dolomites, phyllites and quartzites Talya and Dodguni conglomerates
Bababudan Group	Mulaingiri Formation Ironstones with chloritic and gabbroic schists Santaveri Formation Basic and acid volcanics and pyroclastics Cross-bedded quartzites Allampur Formation Metapyroxinite and meta gabbro Cross-bedded quartzites Kalasapura Formation Alternations of amygdular basalt Cross-bedded quartzites and phyllites Oligomict conglomerate and cross-bedded quartzite Unconformity
Peninsular Gneiss	
Sargur Group	Ultramafic–mafic complexes and anorthosites Ironstone, amphibolites and pyroclastics Marbles and calc-silicate rocks Metapelites with kyanite, staurolite, garnet, sillimanite graphite and corundum Fuchsite (\pm sillimanite, kyanite) quartzite locally with barite beds and chromite layers

ultramafics of Kolar and Chitradurga is estimated using the following equation:

$$T = 1 * p * a * t * d * (1 - \phi), \quad (1)$$

where T is the amount of CO_2 that can be sequestered, p the % MgO in ultramafics, a the area, t the thickness, d the average density and ϕ the average porosity of ultramafics.

For the Kolar schist belt (KSB), considering an effective sequestration of about 20% at a depth of 1 km:

Volume of belt = $320 * 1 = 320 \text{ km}^3$.

Effective volume of belt for sequestration = 20% of $320 = 64 \text{ km}^3$.

% Komatiites = 10% of $64 = 6.4 \text{ km}^3$, out of which 90% is ultramafic.

Volume of ultramafic komatiites = 90% of $6.4 \text{ km}^3 = 5.8 * 10^6 \text{ m}^3$.

Average density³³ = 3000 kg/m^3 .

Mass of komatiites = volume * density = $3000 * 5.8 = 17.4 * 10^9 \text{ t}$.

Take average % MgO in komatiites = 17.5% (CaO , $\text{FeO} \leq 1\text{--}2\%$ have not been considered in the calculation).

Total MgO in komatiites of KSB = 17.5% of $17.4 * 10^9 = 3 \text{ million tonnes (mt)}$.

Since 1 t of MgO can dispose of approximately 1 t of CO_2 (ref. 32), with an average porosity³⁴ of 2% in ultra-

mafic rocks, 3 mt of MgO in KSB can sequester $1 * 3 * (1 - 0.02) = 2.94 \text{ mt}$ of CO_2 in the form of magnesium carbonate in the entire belt.

Similarly, for Chitradurga schist belt (CSB), considering an effective sequestration of 20% in 1 km^3 :

Volume of belt = 6000 km^3 .

Effective volume of belt for sequestration = 20% of $6000 = 1200 \text{ km}^3$.

% Komatiites = 1% of $1200 = 12 \text{ km}^3$, out of which 50% is ultramafic komatiites.

% Ultramafic komatiites = 50% of $12 \text{ km}^3 = 6 * 10^6 \text{ km}^3$.

Average density³³ = 3000 kg/km^3 .

Mass of komatiites = volume * density = $3000 * 6 * 10^6 = 18 * 10^9 \text{ t}$.

% MgO in komatiites = 27% (CaO , $\text{FeO} \leq 1\text{--}2\%$ have not been considered in the calculation).

Total MgO in komatiites of CSB = 27% of $18 * 10^9 \text{ t} = 4.8 \text{ mt}$.

Since 1 t of MgO can dispose of approximately 1 t of CO_2 (ref. 32), with an average porosity³⁴ of 2% in ultramafic rocks, 4.8 mt of MgO in the CSB can sequester $1 * 4.8 * (1 - 0.02) = \sim 4.7 \text{ mt}$ of CO_2 in the form of magnesium carbonate in the entire belt.

The results obtained are presented in Table 4. The estimates, however, can be further improved by including the

Table 4. Sequestration potential of ultramafic rocks in greenstone belts

Greenstone belt	Kolar	Chitradurga
Areal extent* (sq. km)	320	6000
Thickness*	Unknown (~4 km and beyond)*	Up to 10 km
% Komatiites of volcanics*	10	~1
% Mafic to ultramafic*	90	Probably < 50
Max MgO content of komatiites* (%)	14–21	27
Considering an effective sequestration volume of 20% up to a depth of 1 km, the carbon sequestering capacity (for 1 km thickness; million metric tonnes)	2.94	4.7

Average density of ultramafics = 3.0 g/cubic cm (ref. 33). Average porosity of ultramafics = 2% (ref. 34).

*Data from Rogers and Giral²⁷. *From Radhakrishna and Vaidyanadhan²⁴.

reaction kinetics and hydrological parameters in the eq. (1). Similarly, other ultramafic/mafic lithologies can be quantified for their carbon storage capacity and their relevance to carbon storage can be anticipated. Some important aspects about carbon sequestration that have to be considered in mineral carbonation reactions as well as other sequestering options are energy consumption and cost, wherein current technology should aim at the development of alternative separation techniques that are simple and cost-effective. The temporary nature of storage, safety in case of accidental releases and reduction in risk hazards, environmental impact, particularly when sequestered in oceans, monitoring and verification technology for effective long-term storage, and legal awareness and outreach to public regarding details about sequestration in an area of interest also require considerable attention.

Conclusion

This assessment provides the probable amounts of CO₂ that can be stored as mineral carbonates in ultramafic rocks of Kolar and Chitradurga greenstone belts. If the effective sequestration accounts to be 20% of the total, from this computation it is observed that about 2.94 million metric tonnes of CO₂ can be sequestered in the ultramafics rocks of Kolar greenstone belt and the Chitradurga ultramafics can store ~4.7 million metric tonnes of CO₂. For 2002, the carbon emission rate of India was 1.2 Gt and in the background of this assessment of ultramafic rocks for their CO₂ storage capacity, ~0.25% of CO₂ can be disposed of in Kolar and 0.39% in Chitradurga greenstone belts of Dharwar Craton. The estimates shown here exemplify the sequestration potential capacity of the ultramafic rocks. The low porosity, structural features and chemical compositions of the mafic and ultramafic rocks of the greenstone belts in the Archean terrains constrain them to be the best-suited carbon storage reservoirs in comparison to other geological provinces in India, particularly the Cretaceous Deccan Basalts, which have greater surface area, porosity, favourable structural features and abundance of mafic rocks with subordinate ultramafics. A similar assessment for the Deccan Vol-

canic Province in India, based on its geology, structure, mineral and chemical composition for sequestering CO₂ may provide a viable option as another important medium to mitigate the greenhouse gas effect of CO₂. However, the estimates provided here enable us to assess the CO₂ storage potential of ultramafic rocks in the greenstone belts, thus further contributing towards the potential assessment of other geological reservoirs for carbonating the Mg, Fe, Ca silicates under controlled geological and geochemical conditions relevant to the Indian subcontinent.

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