

# Options for the future of cement

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Options for more sustainable cementitious materials are reviewed. The most viable option involves blending Portland cement clinker with increasing levels of supplementary cementitious materials (SCMs), but the availability of common SCMs is low compared to the demand for cement. Calcined clays are the only other materials that are potentially available in large enough amounts to continue the trend of reducing clinker content in blended cements. Recently we demonstrated that partially replacing clinker by calcined clays combined with limestone (LC<sup>3</sup> blends) can be used to achieve blended cements with good performance at much lower levels of clinker. Such blends can make a significant contribution to the reduction of CO<sub>2</sub> emission associated with cement production.

**Keywords:** *Supplementary cementitious materials; sustainability; calcined clay; limestone; CO<sub>2</sub> emissions.*

As global demand for concrete follows the growth of the middle class in the 21<sup>st</sup> century, the concrete industry faces one defining challenge: finding ways to increase the availability of concrete while at the same time reducing its environmental footprint. To have a significant global impact, solutions have to be abundant, affordable, and adapted to the users in the developing world, where most growth is expected.

## CONCRETE IS AN ECO-EFFICIENT MATERIAL

Cement and concrete are essential to the infrastructure of the modern world. No other material is able to fulfil the growing demand for building materials with such a low environmental footprint. Its widespread availability and low cost make cement by far and away the most used material on earth, with reinforced concrete accounting for more than half of all the manufactured materials and products we produce. It is only because of these

enormous volumes that overall the production of cement and concrete is estimated to account for around 5-8% of man-made CO<sub>2</sub> emissions.

Table 1 shows relative figures for the energy and CO<sub>2</sub> emissions of some common building materials [1]. Here, typical values for concrete are shown, as this is the final product used in the field. As discussed elsewhere

**Table 1. Embodied energy and associated CO<sub>2</sub> emissions for common construction material [1]**

Material	Embodied Energy (MJ/kg)	CO <sub>2</sub> (Kg CO <sub>2</sub> /kg)
Normal concrete	0.95	0.130
Fired clay bricks	3.00	0.22
Road & pavement	2.41	0.14
Glass	15.00	0.85
Wood (plain timber)	8.5	0.46
Wood (multilayer board)	15	0.81
Steel (from ore)	35	2.8

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[2], there is a huge scope also for reducing the amount of cement in concrete and it should always be borne in mind that sustainability can and should be considered at all stages of the process. The figures in Table 1 are per kg and the strength of concrete is not the same as the specific strength of steel or wood. These issues have been considered by several authors [e.g. 3]. Nevertheless, the advantages of concrete remain clear in most construction related application; even more so when the availability of materials to substitute concrete is considered. As an example, although the amount of wood used worldwide is around one tenth that of concrete, our consumption of wood is already considered to be unsustainable; we are cutting down more trees than we are planting. It follows that, independent of any other considerations; wood cannot make a significant further contribution to meeting the increasing demand for building materials worldwide.

### DEMAND FOR CEMENT IS INCREASING

Figure 1 shows the projected increase in demand for cement. Most of the increase in the coming decades will be in emerging and developing countries, which already make up over 80% of global production. Today, China's production alone exceeds that of the whole world 10 years ago. India is the country in which the increase is likely to be the largest – today the consumption per capita in India is only 1/6<sup>th</sup> that in China.

### OPTIONS TO REDUCE ENVIRONMENTAL IMPACT

Cement production accounts for the overwhelming majority of the CO<sub>2</sub> emissions associated with concrete. Unlike other materials, less than half (~40%) the CO<sub>2</sub> emitted during cement production is related to fuel and electricity. The remaining 60% comes from the decomposition of the main raw material – limestone, or CaCO<sub>3</sub>. Great improvements have been made in the last few decades in lowering energy-related CO<sub>2</sub>. Today, the production of Portland cement clinker is one of the most efficient industrial thermal processes in existence (approaching 70% of theoretical efficiency) and it is unlikely that significant further gains can be made here as discussed in more detail by Gartner [4]. Furthermore cement plants can now use a wide range of substitute

fuels, reducing the need for primary fossil fuels to below 20% in some modern plants. This versatility, the fact that the calorific value of such a wide range of waste products can be exploited in a safe manner, should be seen as another advantage of cement production.

Any attempt to reduce the remaining 60% of “chemical” emissions coming from the decarbonation of limestone will have the inevitable consequence of changing the cement's chemistry. Consequently, the reactions and performance of new materials will not necessarily be the same as the reference Portland cement. It took more than 100 years of empirical testing to develop the basis for use of Portland cement. The use of the most common blended materials – blends of Portland cement clinker with slag and fly ash – has taken more than 30 years to become established. We do not have the time to go through this long testing phase for every new material that comes along. That is why we must now move towards a more scientific basis, which can only come (on a reasonable timescale) through a systematic understanding of cementitious processes and materials at the nano-scale, extended across all the scales involved in cement and concrete production, to provide the multidisciplinary assessment and prediction tools needed to assess the functional and environmental performance of new materials.

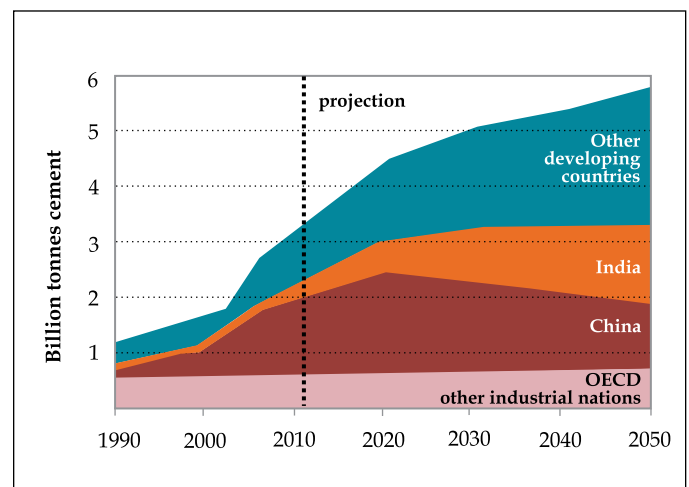


Figure 1. Forecast rise in cement production until 2050 (adapted from CEMBUREAU)

Table 2. summary of suitability of common oxides for cementitious materials.

oxide	hydrates		geology	
CaO	Good mobility, hydrates can fill space	✓	Limestone, widely distributed	✓
SiO <sub>2</sub>	Less soluble but still a major component of space filling hydrates	✓	Most common oxide, everywhere	✓
Al <sub>2</sub> O <sub>3</sub>	Variable solubility, hydrates fill space	✓	With silica in feldspars, clays, etc. also more concentrated and localised as bauxites	✓ / ?
Fe <sub>2</sub> O <sub>3</sub>	Low mobility / solubility in alkaline solutions, poor contribution to space filling	?	Very widely distributed	✓
MgO	Low mobility / solubility in alkaline solutions, poor contribution to space filling		Mainly as impurity in limestone, more concentrated sources localised.	

The options for new cement chemistries are ultimately limited by the composition of the earth. Just eight elements constitute more than 98% of the earth's crust – oxygen, silicon, aluminium, iron, calcium, sodium, potassium, and magnesium. The relative abundances are shown logarithmically in Figure 2 [5]. As a first approximation, cost will be closely related to availability, leaving only 7 oxides as possible candidates for cement making. These are summarized in Table 2, which broadly considers first their potential to form space filling hydrates and then their geological distribution.

The potential to form space-filling hydrates is the key to a hydraulic cement. First, the anhydrous cement is mixed with water. The solid content of the resulting mixture cannot exceed around 50% (W/C ~ 0.3) if the mixture is to be flowable. Then the anhydrous material must dissolve and precipitate new solids incorporating water with a higher solid volume than the original anhydrous material. The phases present in Portland cement clinker are ideal in this respect as there is roughly a doubling in solid volume, which enables most of space initially occupied by water to be filled, yielding a strong solid with low porosity.

The alkali oxides retained from Figure 2, Na<sub>2</sub>O and K<sub>2</sub>O, cannot produce hydraulic compounds as these have a very high solubility and will not deposit hydrates. Nevertheless, small amounts of these compounds play an important role in Portland cement, where they nearly all end up in the pore solution, conferring the concrete with the high pH needed to protect reinforcement. Iron and

magnesium oxide, on the other hand, lead to the opposite problem. These oxides do form insoluble hydrates, but because their mobility in the alkaline pore solution of cements is very low, these hydrates are almost exclusively deposited within the boundaries of the original grains and do not contribute to filling the space initially occupied by water or to binding the grains together. Even in very old cement pastes (even > 100 years [6]), we see bright areas corresponding to the original ferrite phase in the cement, as in Figure 3 (a). In slag cements, the magnesium component of the slag remains within the boundaries of the original slag grains and does not contribute to binding

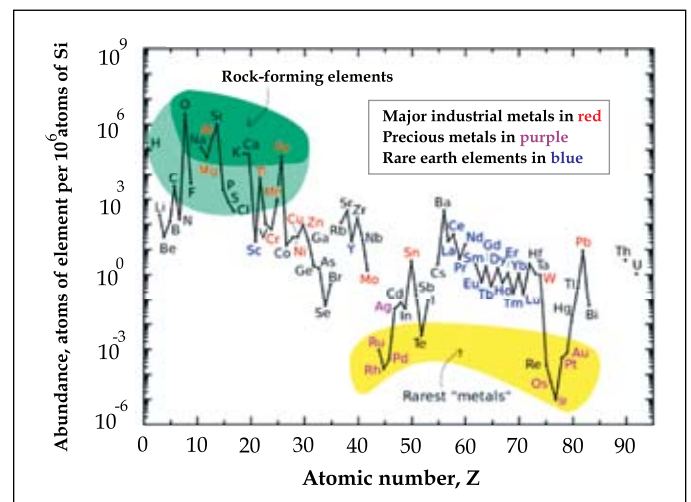


Figure 2. Relative abundance of elements, source US Geological Survey via Wikipedia [5]

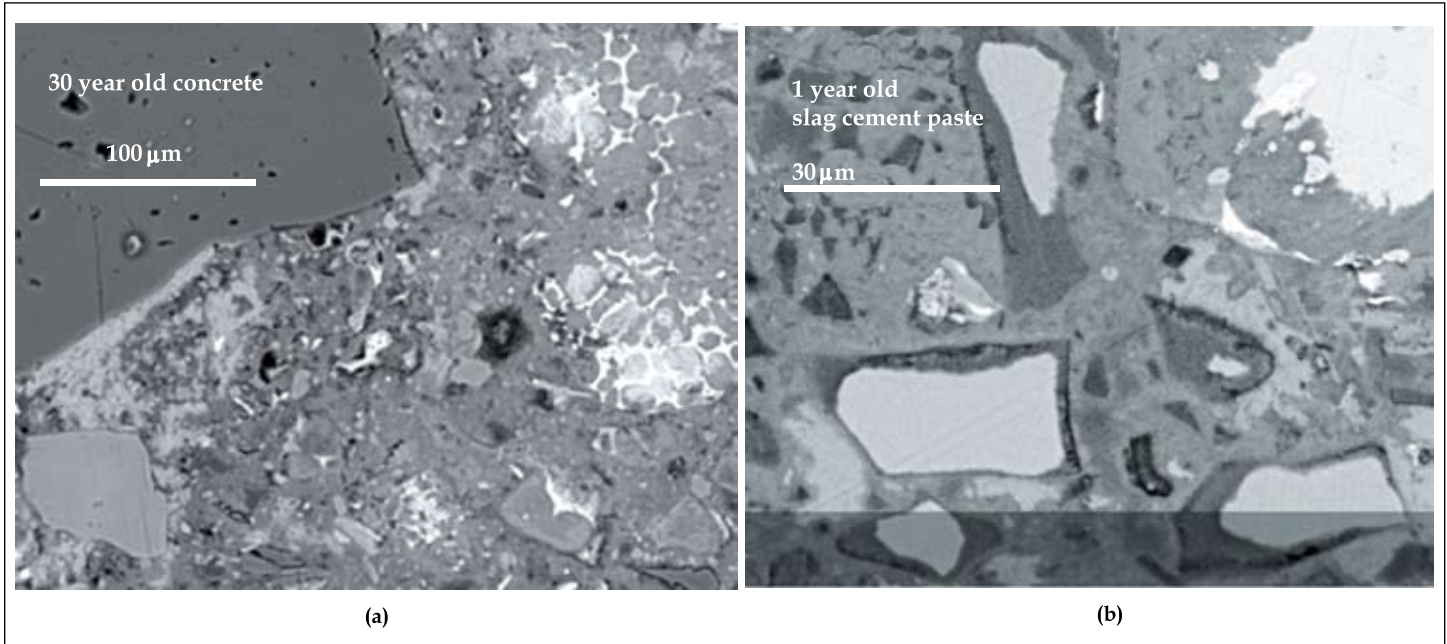


Figure 3. (a) Micrograph of old concrete shows low mobility of iron which remains in the bright areas corresponding to the ferrite phase in the original cement grains. (b) Micrograph of slag cement paste shows low mobility of magnesium, which remains within the boundaries of the original slag grains.

the materials together (Figure 3 (b)). This brings us back to the three oxides that dominate Portland cement – lime, silica and alumina.

Strategies to lower the environmental impact of cement manufacturing are already applied by the industry and their future potential has been evaluated in detail in by the IEA (International Energy Agency) for the Cement Sustainability Initiative (CSI) study of the World Business Council for Sustainable Development (WBCSD), from which Figure 4 is adapted [7]. Today the three most important approaches are:

1. Improvement of energy efficiency (red part);
2. Use of biofuels and other alternative fuels (blue part), and
3. Replacement of clinker by substitute materials or supplementary cementitious materials (SCMs). Materials used include fly ash, various slags, and natural pozzolans (green part).

Beyond these approaches the CSI study imagines the remaining reduction in CO<sub>2</sub> to come from Carbon Capture and Storage (CCS) (grey area), which is now widely thought to be, at best, very expensive (increasing cement prices 2-4 times). As seen in the Figure 4, the further gains from energy efficiency are limited as they are reaching a plateau of optimization. Gains from alternative fuels are projected to increase, but remain a fairly modest proportion of the total. The projected impact of the third option, substituting clinker by SCMs, is limited by the forecast supply of commonly used SCMs, notably slag and fly ash. For example, although slag can substitute up to 90% and typically 70% of clinker in blends, the worldwide amount of slag available is only around 5% of the amount of clinker produced. Fly ash, although available in larger amounts (around 30% of clinker worldwide, but absent in many countries), is of variable quality and much is unsuitable for cement production. So the strategy of clinker substitution could go much further if new sources of SCMs were available. However very few materials are available in the large quantities needed for cement production.

### RADICALLY DIFFERENT TECHNOLOGIES

Before discussing in detail alternative supplementary cementitious materials it is important to say something about radically different technologies such as alternative clinkers or clinker free materials.

#### Alternative clinkers

Figure 5 shows the “chemical” CO<sub>2</sub> emissions of the hydraulic minerals in the CaO – SiO<sub>2</sub> – Al<sub>2</sub>O<sub>3</sub> – (SO<sub>3</sub>) system from two perspectives: first per g of anhydrous material,

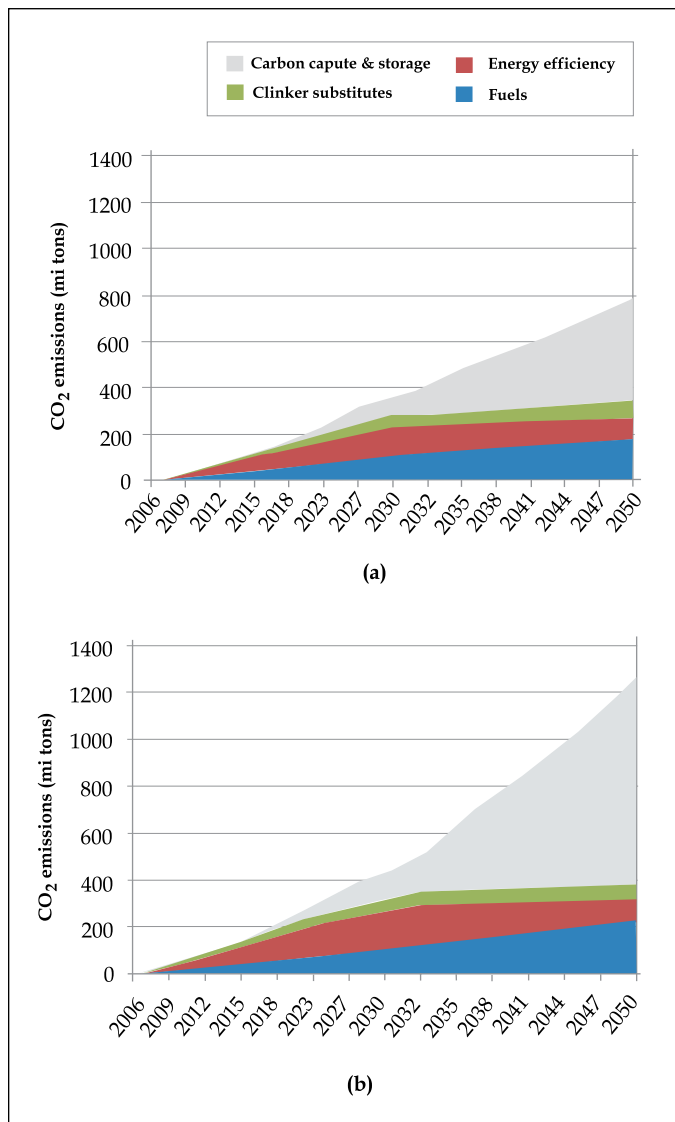


Figure 4. CO<sub>2</sub> reduction scenarios compared to 2005 baseline from CSI study Low (a) and high (b) scenarios for demand increase

then per ml of hydrates that can be formed, this latter is related to their space filling capacity and so to strength development. The volume of hydrates for the aluminate compounds (monocalcium aluminate and ye’elinite) is shown with and without added sulfate. Here it can be seen that the figures for the two most important Portland cement clinker minerals – C<sub>3</sub>S and C<sub>2</sub>S – have fairly similar values. Notably we see that C<sub>2</sub>S only presents a saving of around 10% in CO<sub>2</sub> compared to C<sub>3</sub>S. This means that for belite rich cements to represent a “real” CO<sub>2</sub> saving, they have to have more than 90% of the performance of alite rich cements. Despite intensive research efforts, we are far from achieving this in terms of the performance needed in modern construction. This table clearly shows that the most interesting minerals from the point of view of their associated CO<sub>2</sub> values are monocalcium aluminate (CA), the main component of calcium aluminate cements (CACs), and ye’elemite or Klein’s compound (C<sub>4</sub>A<sub>3</sub>\$), the main component of calcium sulfoaluminate cements (CSAs or SACs). It is therefore worth discussing the viability of clinkers based on these mineral in some more detail.

The main problem is that while alumina is very abundant in the earth’s crust, it exists mainly in feldspars and clays, where it is present alongside silica with Si:Al ~ 2 similar to that found in most portland clinkers. To obtain the desired alumina rich compounds, a more concentrated source of alumina is needed, typically bauxite. But bauxite

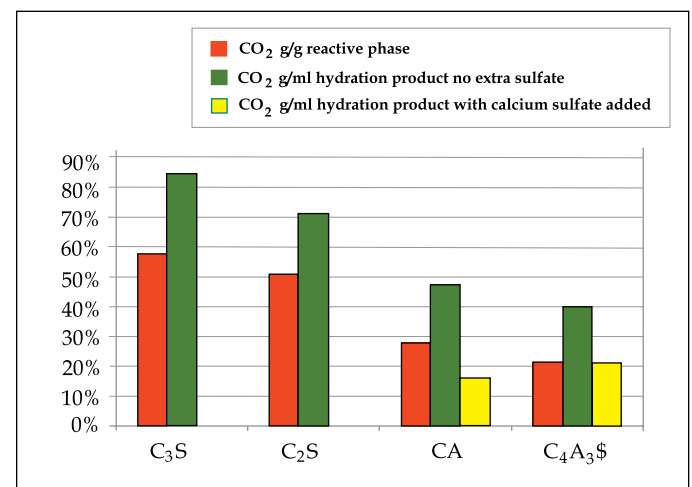


Figure 5. CO<sub>2</sub> emissions related to the major hydraulic minerals

is not nearly as widely distributed as clay minerals and is highly sought after for the production of aluminium, making it a much more costly raw material. But this is not the end of the story, as will be explained from the perspective of calcium aluminate cements.

Calcium aluminate cements (CACs) have been in commercial production for nearly 100 years, yet the total amount produced annually is less than 1/1000 of the production of Portland cement clinker, and only a small proportion of this is used in construction. This is because the cost of CAC is more than five times the cost of Portland cement. The higher cost of raw materials and production accounts for only part of this huge discrepancy. The other part lies in the number of people needed to support the use of this special material, in terms of factories producing smaller amounts, and sales and technical assistance for a specialised material that does not behave in the same way as the reference Portland cement. Consequently the use of CAC can only be justified where their special properties justify the higher cost.

Calcium sulfo aluminate cements have gained a lot of attention recently, as, in addition to the low chemical CO<sub>2</sub> of ye'elemite, the firing temperature is lower and they are more easily ground. CSAs are a very diverse family of cements. They may contain from 70% ye'elemite down to 30% or less. The main other phase that is present is usually belite. This leads to a two-stage development of properties, with the fast reaction of the ye'elemite giving early strength and the belite hydration giving later strength gain. Another complication is that the behaviour can be changed from rapid hardening to expansive by the amount of calcium sulfate added during grinding. Due to this diversity it is difficult to generalise, but it can certainly be said that today, these are specialist products along the lines of CACs rather than cements for general construction. They have been produced in China for several decades and have been used for some large-scale constructions, yet to this day, they represent less than 0.1% of the cement produced in China and are mostly used for internal partition walls.

Personally, I am doubtful that CSA cements can be competitive with blended cements discussed below on either an economic or environmental basis in general use.

The need to support consumers in their use of cements that behave differently to the Portland reference, in addition to the higher raw materials cost, will mean that the commercially viable cost will be very much more than that of Portland cement. Nevertheless such developments may be interesting in more specialist applications, for example for precast concrete.

### Clinker free binders

At present many researchers (and funding agencies) are attracted by the idea of "clinker free binders, such as alkali activated materials, also known as "geopolymers," claiming their advantages as low CO<sub>2</sub> cements. Despite the hype, it is highly unlikely that such materials will have a major impact on the sustainability of cementitious materials for many reasons:

- First, the environmental advantages of these materials are questionable. The environmental assessment made by Habert et al [8] indicates that while the production of geopolymer concrete can have a slightly lower impact on global warming than standard Ordinary Portland Cement (OPC) concrete, it has a higher environmental impact regarding other impact categories related to the production of concentrated sodium silicate solution and the dangers of handling highly alkaline solutions.
- Second, the performance of these materials is very sensitive to small variations in the starting materials, such as slag or fly ash. The setting time and rheology are difficult to control and may vary widely on a batch-to-batch basis. They are also very sensitive to contamination by conventional Portland cements. This makes use on-site by typical construction workers challenging.
- Third, the durability and long term performance of these materials is not well known.
- Finally, and perhaps most importantly, the materials used in these alkali activated materials are the same SCMs discussed above – slag, fly ash, etc., which are only available in relatively low amounts compared to cement clinker. The materials that perform best

in these systems are the same as those that perform best in cement blends.

So from a practical point of view, one is faced with a choice of how to use slag: either (a) as a component of a Portland based blend in a well-known and tested technology with proven durability that can be used in existing mix designs with conventional equipment, or (b) in a clinker free system with the attendant problems of sensitivity, strongly alkali materials, risks of contamination, and unknown long-term performance. Again the cost of providing technical support to users also means that a price significantly higher than that of Portland cement will have to be charged for such materials to be a commercially viable.

**ALTERNATIVE SCMS: LC<sup>3</sup> – LIMESTONE, CALCINED CLAY, CLINKER CEMENTS**

Faced with the limited supply of traditional SCMs, the key question becomes: what alternative SCMs are available in the very large quantities needed? Materials such as plant ashes, notably rice husk ash, can be interesting, but the quantities available are relatively small and localised (both geographically and to certain periods of the year). There are extensive reserves of natural pozzolans worldwide (e.g. volcanic ashes), but generally these have a fairly low reactivity and are very variable from place to place. The one material which has a real potential to make a significant contribution to lowering the clinker (and so CO<sub>2</sub> emissions) of cement is clay, which becomes reactive when calcined at temperatures between 600 – 800 °C.

The use of calcined clays as pozzolans has been known for some time, it was widely used in India in the 1970s (before fly ash was widely available) and is currently being used in countries without supplies of the common SCMs, notably Brazil. However, substitution just as a pozzolan is only practical up to about 30%, at which level the cost of calcination usually does not make this an economically viable option. Recently, it has been shown that by making a coupled substitution of calcined clay with limestone, an additional 15% or so of clinker can be replaced by limestone with no decrease in mechanical performance so that the extra saving of clinker can offset the cost of calcination. This is because the aluminate component of the calcined clay can react with calcium

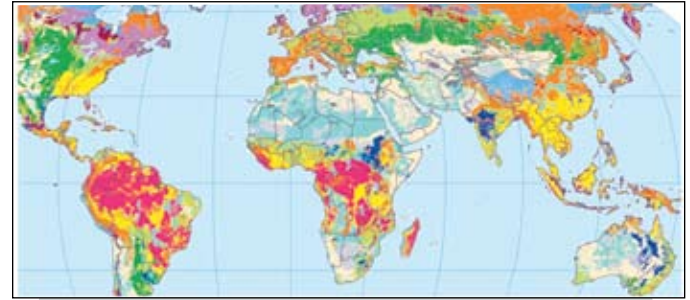
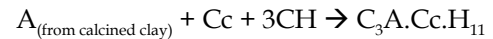


Figure 6. World soil map. Kaolinite rich clays are most likely to exist near the surface (top 5m) in the yellow and pale green regions [9]

carbonate (limestone) and calcium hydroxide to produce space filling carbo aluminate hydrates:



Clays are the weathering product of all rock types and as such are abundantly available close to the earth’s surface in all geological settings. They are made up of silicon and aluminium oxides, which together constitute around three quarters of the earth’s crust. The most suitable clay types, those containing kaolinite, are typical in tropical and subtropical environments, where most of the increase in demand for cement is forecast to occur. Kaolinite clay occurs in abundance in association with soils that have formed from the chemical weathering of rocks in hot, moist climates, Alfisols (pale green) and Ultisols (yellow) on the map (Figure 6). This confirms the abundance of clay, especially in India and South East Asia.

The potential for ternary blends of limestone, calcined clay and clinker, which we call LC<sup>3</sup> (limestone calcined clay cement), to give good performance at 50% clinker content or less has been demonstrated in the collaborative research between the Laboratory of Construction Materials (LMC) at EPFL, Switzerland, and CIDEM in Cuba [10].

The kaolinite content needed for LC<sup>3</sup> blends is much lower than for the “pure” kaolinitic clays used in the ceramic or paper industries. This means that the use of such “low grade” clays would not compete with demand for resources by other industries. Our experience in India [11] has shown that abundant amounts of low-grade clays exist as over or under burden in existing

quarries, currently regarded as waste. The use of such resources would not require opening new quarries nor deplete agricultural soils. With an optimum at around 700 to 800°C, the calcination temperature is much lower than the 1450°C needed for clinker manufacturing, hence consuming less fuel. Furthermore, this calcination process does not emit chemical CO<sub>2</sub> and can be carried out with existing equipment (rotary kilns or tower calcination units).

Limestone is always available at cement plants. The LC<sup>3</sup> blends typically contain around 15% limestone, and limestone unsuitable for clinker production can be used. For example, high dolomite contents produce periclase during clinker production, which causes expansion. Such materials can be used safely in interground materials, leading to more efficient use of limestone quarries. Depending on the exact scenario, the amount of cement that can be produced from the same identified limestone reserve could be increase by up to a factor of two.

Further important advantages of LC<sup>3</sup> technology are:

- Cheaper or similar production costs

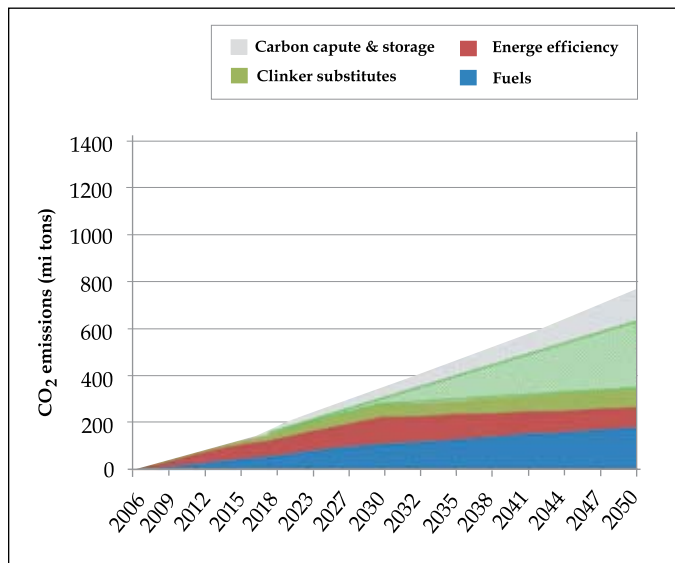


Figure 7. Potential additional reduction of CO<sub>2</sub> by clinker substitution through the use LCC ( calcined clay, limestone ) technology.

- LC<sup>3</sup> can be produced using the existing equipment in a cement plant
- Use of LC<sup>3</sup> also does not need any major changes in concrete technology.

This leads to an enormous potential for rapid uptake of the technology with significant potential for CO<sub>2</sub> reduction and efficient use of resources.

LC<sup>3</sup> has the potential to provide a large-scale solution to the two main challenges facing the production of building materials today. A rough estimation of the impact of LC<sup>3</sup> on the CO<sub>2</sub> reduction scenarios is evaluated in relation to the results of the WBCSD CSI study [7] (Figure 7). This assumes that the supply of SCMs can be increased by a fairly modest 600 million tonnes worldwide by 2050. This graph shows how LC<sup>3</sup> can significantly contribute to the growth of the cement sector without increasing emissions.

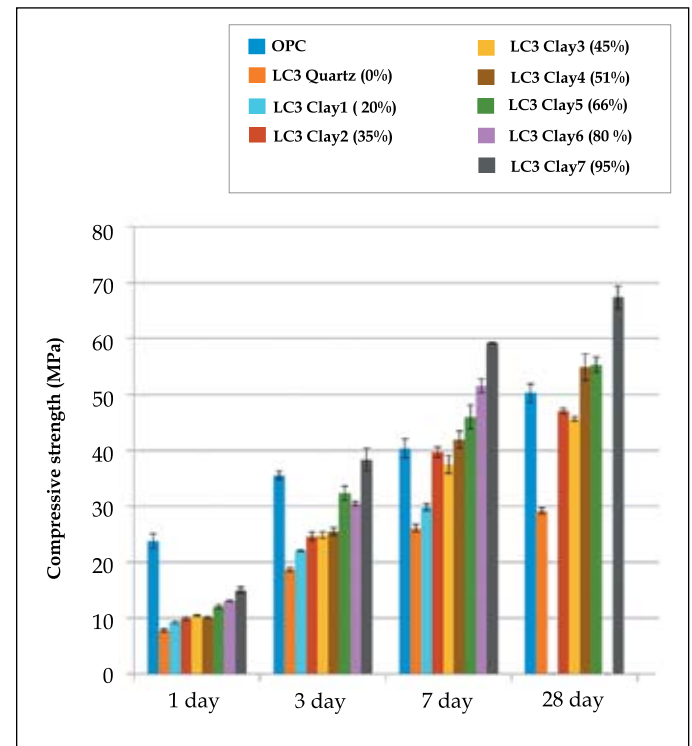


Figure 8. Strength development for a range of blends all containing 50% clinker, 5% gypsum, 15% limestone and 30% calcined clay. The calcined clays originate from India, Brazil, Thailand and Cuba.



This is more feasible than compensating a business as usual strategy with carbon storage and capture. Importantly, the uptake of LC<sup>3</sup> would allow developing countries to satisfy rapidly growing demand while reducing the CO<sub>2</sub> intensity of their activities and is thus a politically very attractive global environment option.

### Strength of LC<sup>3</sup> blends prepared from low grade clays

Further investigating the potential of blends based on limestone, calcined clay, and clinker (LC<sup>3</sup>), blends incorporating calcined clay from several countries (India, Brazil, Thailand, Cuba) were analyzed. An example of the strengths obtained is shown in Figure 8.

Plotting these results against the kaolinite content of the clays, it can be seen that kaolinite content is the main parameter determining strength development (Figure 9).

In these experiments the cements were prepared by intermixing based on the same ground clinker. It can be seen that the one day strengths are still rather low, but, for a kaolinite content of 50% or more, higher strengths than the reference are achieved by 7 days. In practice one day

strength can be improved by intergrinding as is currently done for many fly ash blended cements.

### Durability of LC<sup>3</sup> cements

When considering a new cement formulation, the question of durability is of prime importance. Will these materials perform as well as existing materials over the lifetime of a typical building or structure? A wide-ranging and detailed study of the durability of LC<sup>3</sup> is underway in Switzerland, India and Cuba. This study will look at the underlying scientific mechanisms as well as full scale exposure. However, we have every reason to expect that these materials will have good durability:

1. First, the phase compositions of the materials is very similar to existing Portland and blended cements. The principal hydrate is calcium silicate hydrate, C-S-H, whose long-term behavior is well known and understood. The other aluminate containing phases, mono and hemi carbonate and ettringite are also formed in limestone cements, widely used in Europe for many years.
2. Second, analysis of the pore structure shows that, as for other blended cements [e.g. 12], the pores

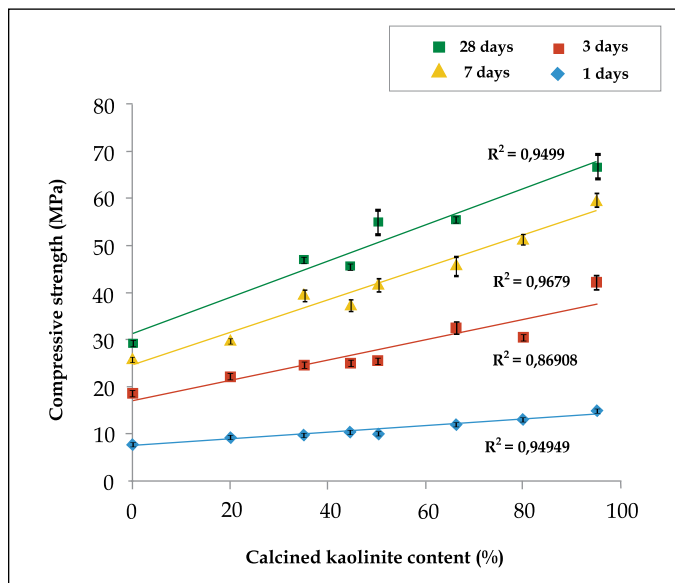


Figure 9. Relation between kaolinite content of the clay which is calcined and strength

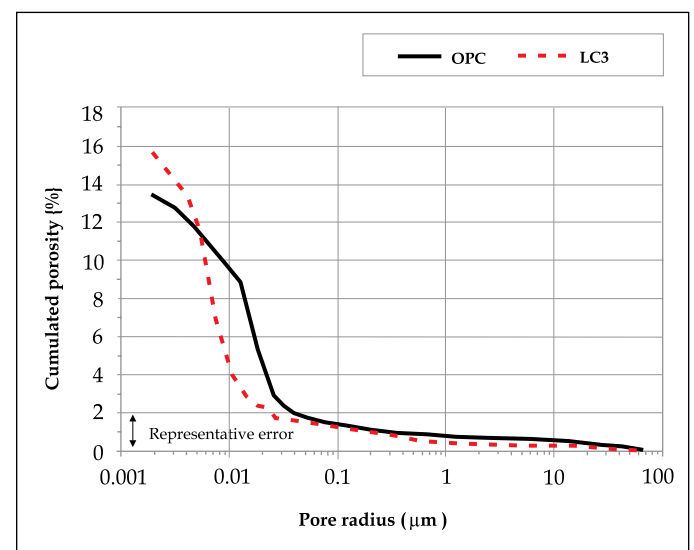


Figure 10. Mercury Intrusion Porosity results for LC<sup>3</sup> blend and reference Portland cement

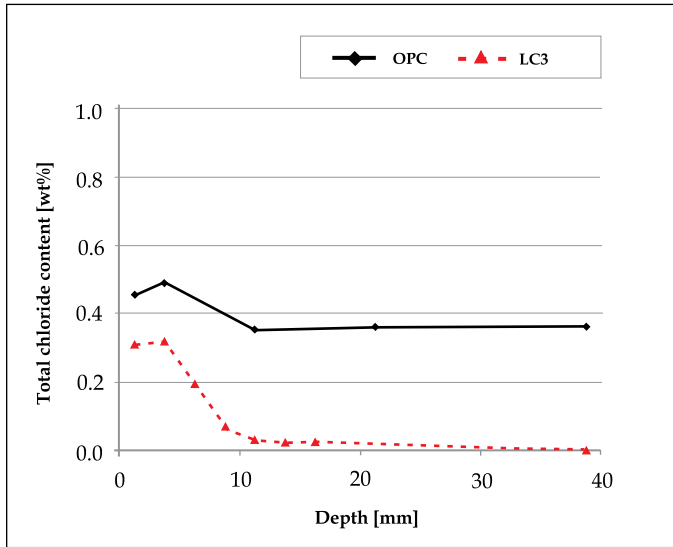


Figure 11. Chloride profiles after 2 years ponding in 0.5M NaCl solution

are smaller, even though the overall porosity may be slightly higher (Figure 10) [13].

- Third, preliminary results on the resistance to penetration of chloride ions are extremely good. Figure 11 shows the chloride profiles after two years ponding in 0.5M NaCl solution [13].

## CONCLUDING REMARKS

Fulfilling the needs of the growing world population for food and habitation and at the same time avoiding drastic climate change due to the emission of greenhouse gasses is the most important challenge facing the world today. In the field of building materials, cement and concrete remain the best option as they can be produced in large quantities almost anywhere in the world and have an intrinsically low environmental footprint compared to alternatives. Nevertheless, they are the focus of intensive research efforts to reduce the 5-8% of CO<sub>2</sub> emissions attributable to their production. This paper tries to present a brief but comprehensive analysis of options for cements of the future, starting from the composition of the earth and considering the practicalities of producing

such a low cost material that can be used by anyone, even with little or no training. This shows that blends in which around half of the conventional Portland cement clinker is replaced by a combination of limestone and calcined clay - *LC<sup>3</sup> blends* - present an extremely promising option to achieve lower CO<sub>2</sub> emissions, lower costs, and increase supply - particularly in tropical and subtropical countries where the increase in demand will be the highest in coming decades. We have already received strong expressions of interest in this technology from many cement producers worldwide.

More work is needed to provide a solid scientific and engineering foundation for these new materials. A large body of work is already underway, undertaken by a network of researchers in Switzerland, India, and Cuba. The core of this work has been made possible through significant funding provided by the Swiss Agency for Development and Collaboration. In the coming years, we hope to extend this network to more countries as a co-ordinated worldwide effort is needed to accelerate the introduction of such environmentally friendly options.

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