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Impact of different climates on the resistance of concrete to natural carbonation

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HIGHLIGHTS

- Sheltered natural carbonation can be similar between different climates.
- Huge impact of curing on natural carbonation for very early age (i.e. 1 day)
- Impact of curing is negligible for advanced curing age, regardless of cement type.
- Strong impact of precipitation on natural carbonation.
- The higher the number of rainy days, the lower the carbonation speed.

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ABSTRACT

This paper describes a unique international inter-laboratory study on the carbonation resistance of concrete prepared with different supplementary cementing materials. Concrete specimens – from 45 different concrete mixtures – prepared centrally in Lafarge Centre de Recherche (France) were shipped in a sealed condition to 4 other academia research laboratories (located in USA, Canada, India and China). The specimens were exposed to the ambient environments and atmospheric CO₂ concentrations in the five locations, including Lafarge Centre de Recherche in France, in both sheltered and unsheltered condition for a period of 5 years. Measurements of carbonation depth were performed at periodic intervals, and the data was analyzed to assess the influence of climatic conditions on the resistance to carbonation. The results indicate that the general trend of carbonation is not much different irrespective of the macroclimate. Further, the number of rainy days seems to have a more significant influence on the progress of carbonation than the total rainfall in the region.

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1. Introduction

The dominant cause of premature deterioration and reduced service life of reinforced concrete structures is corrosion. According to the National Academy of Corrosion Engineers (NACE), they estimated the global cost of corrosion at a staggering \$2.5 trillion in

Abbreviations: LS, limestone filler; S, slag; FA, fly-ash; PZ, pozzolan; w_{eff}/b , effective water to total binder ratio.

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2013 (NACE International, 2016). These costs include corrosion incurred in the infrastructure and transportation sectors which are the domains of reinforced concrete structures. Carbonation and chloride ingress are the two causes of corrosion in reinforced concrete structures, the dominant cause being dependent on the geographical location and environmental effects to which the concrete structure is subjected.

Reinforced concrete structures located away from coastal regions are expected to be affected by carbonation-induced corrosion rather than chloride-induced corrosion. Carbonation-induced corrosion can result in uniform cross-sectional loss of the rebar

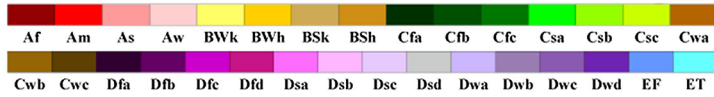
reducing the residual capacity of reinforced concrete structures. Carbonation is a physicochemical process that involves the diffusion of CO₂ through the concrete pores and its reaction with the hydrated cement products, such as calcium hydroxide and calcium silicate hydrates, lowering the alkalinity of the concrete.

The need for the reduction of annual CO₂ emissions due to cement production, and better strength and durability performance of concretes made with blended cements, are reasons to opt for cement clinker substitution in concretes [1–3]. However,

the carbonation resistance of such cementitious systems is found to be reduced with high volume clinker substitution [4–9]. This could be attributed to the reduction in CaO buffer capacity of the cementitious system with respect to increase in the partial substitution of clinker fraction [10,11]. Similarly, a decrease in the duration of the initial moist curing could increase the carbonation rates in concrete mixes with moderate binder content (i.e. less than 380 kg/m³) and low w/c ratio [12,16–18]. In addition, increase in atmospheric CO₂ levels due to increase in man-made industrial

World Map of Köppen–Geiger Climate Classification

updated with CRU TS 2.1 temperature and VASCLimO v1.1 precipitation data 1951 to 2000



Main climates

- A: equatorial
- B: arid
- C: warm temperate
- D: snow
- E: polar

Precipitation

- W: desert
- S: steppe
- f: fully humid
- s: summer dry
- w: winter dry
- m: monsoonal

Temperature

- h: hot arid
- k: cold arid
- a: hot summer
- b: warm summer
- c: cool summer
- d: extremely continental
- F: polar frost
- T: polar tundra

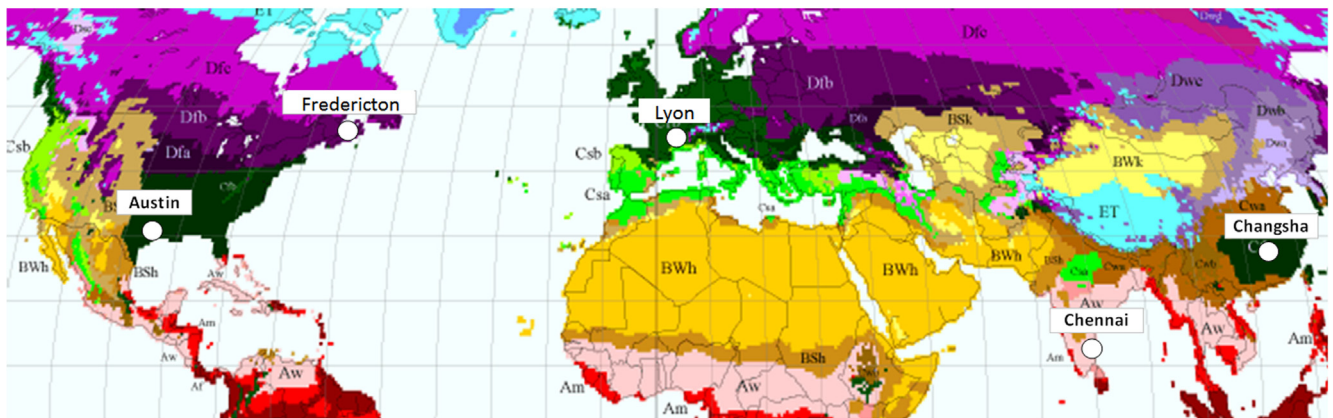


Fig. 1. Geographical location and climate type of exposure sites.

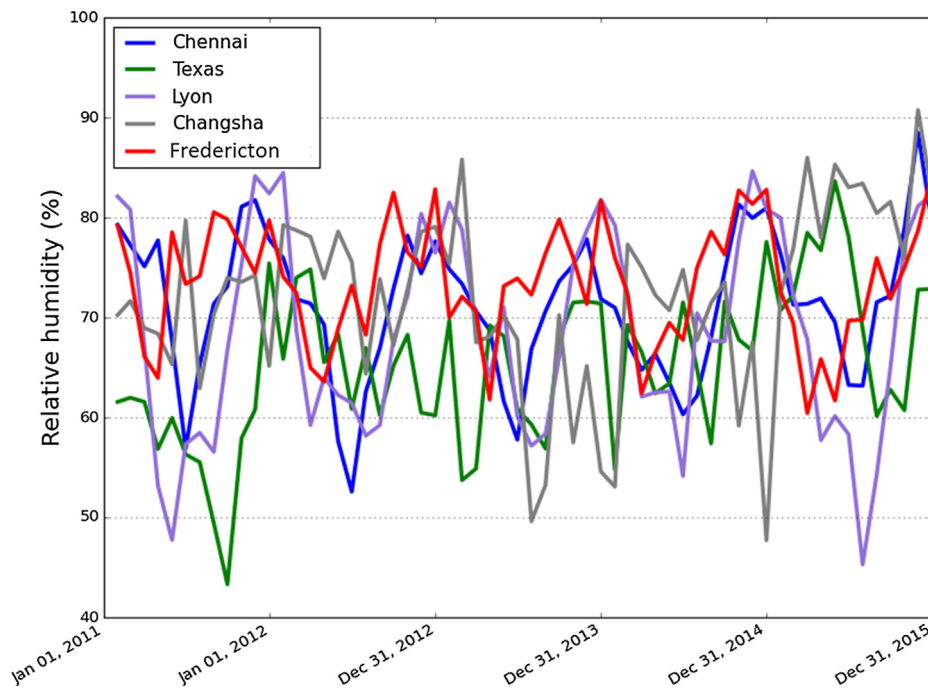


Fig. 2. Monthly relative humidity from 2011 to 2015.

and vehicular emissions in urban areas increase the potential threat to reinforced concrete structures by accelerating carbonation [19–23]. Thus, the estimation of potential carbonation resistance based on prevailing exposure conditions, as well as type and composition of materials used in the structure is necessary to construct a building with intended life expectancy.

In the past two decades, many researchers have attempted to develop models to estimate carbonation depth through empirical, numerical and reaction-kinetics based approaches, primarily based on accelerated carbonation data [24–28]. Carbonation depth pre-

dictions based on natural carbonation data are limited due to lack of long-term durability data. Based on the linear relationship observed between natural and accelerated carbonation coefficients calculated using Tutti’s square root of time law, empirical relationships were proposed by researchers [6,29,30]. These conversion factors between accelerated and natural carbonation coefficients vary with respect to the local climate of exposure site, CO₂ concentration, and type of exposure (i.e. sheltered or unsheltered) [5]. Reinforced concrete buildings in tropical climatic zones are reported to have higher carbonation than buildings in temperate

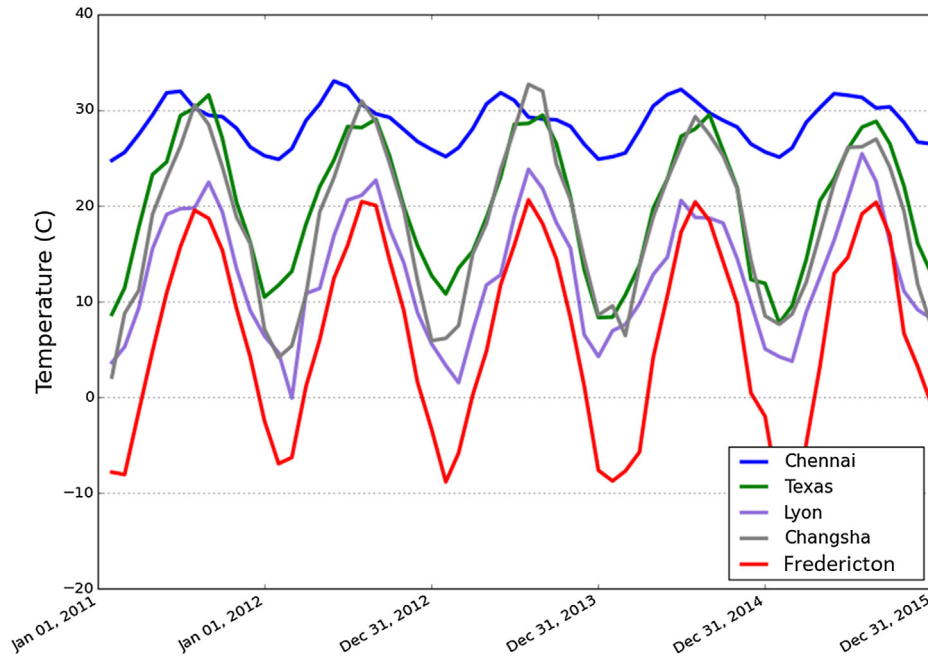


Fig. 3. Monthly temperature from 2011 to 2015.

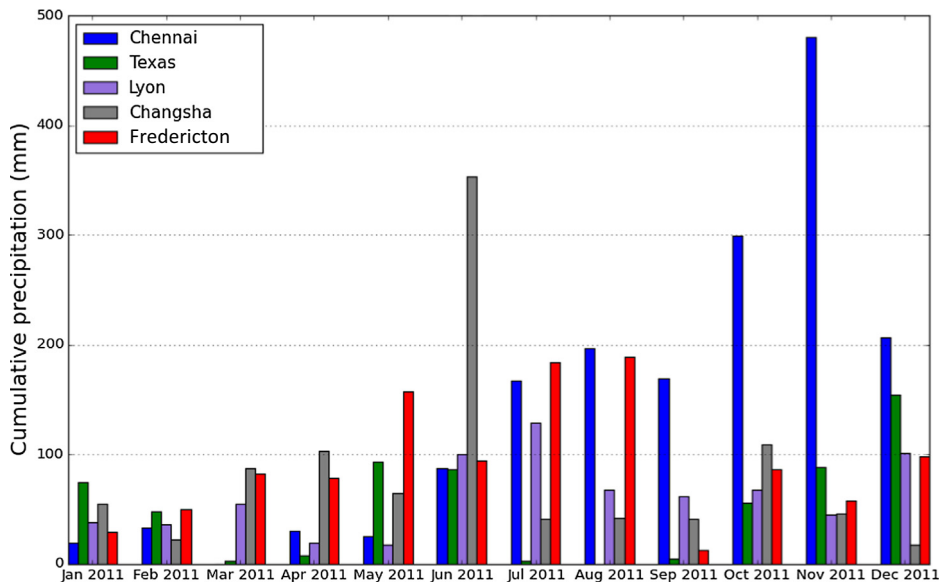


Fig. 4. Monthly cumulative precipitation during 2011.

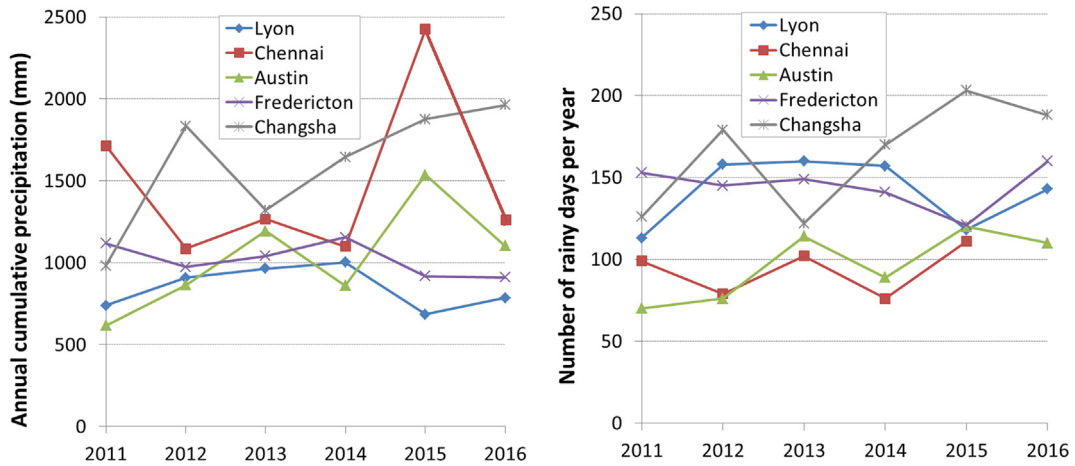


Fig. 5. Amount of annual cumulative precipitation and number of rainy days per year in 5 locations from 2011 to 2016.

Table 1
Binder properties.

Parameters	Unit	Cement I 52.5 N (OPC)	Limestone (LS)	Slag (S)	Fly ash (FA)	Pozzolan (PZ)
LOI at 950 °C	[%]	1.13	40.74	–	4.82	7.66
SiO ₂	[%]	20.28	6.43	36.09	55.61	68.05
Al ₂ O ₃	[%]	4.76	0.36	11.44	25.6	13.04
Fe ₂ O ₃	[%]	3.05	0.16	–	4.94	2.28
CaO	[%]	64.47	51.4	42.09	2.76	2.09
MgO	[%]	0.81	0.32	7.62	1.39	0.91
SO ₃	[%]	3.49	0.02	2.03	0.18	0.04
K ₂ O	[%]	0.78	0.04	0.41	1.95	3.03
Na ₂ O	[%]	0.17	0	0.27	0.49	2.78
Free lime	[%]	1.13	–	–	0.11	–
Specific gravity	[g/cm ³]	3.14	2.72	2.91	2.58	2.38
SSB	[cm ² /g]	3561	–	3656	4540	4340
BET	[m ² /g]	–	2.6	–	5.15	10.6
Glassy phase	[%]	–	–	99.6	37.5	–

Table 2
Aggregate properties.

Property	Sand [0–5 mm]	Gravel [5–20 mm]
Specific gravity [g/cm ³]	2.58	2.59
Water absorption at 24 h [%]	0.85	0.98

Table 3
Range value for main characteristics of 45 concretes.

	Characteristics	Min–Max
Substitution rate of addition	w _{eff} /b ratio	0.45–0.65
	Air voids content [%]	0.9–7.0
	Paste volume [L/m ³]	210–320
	Clinker [%]	50–100
	Limestone [%]	0–30
	Slag [%]	0–50
	Fly ash [%]	0–50
	Pozzolan [%]	0–30

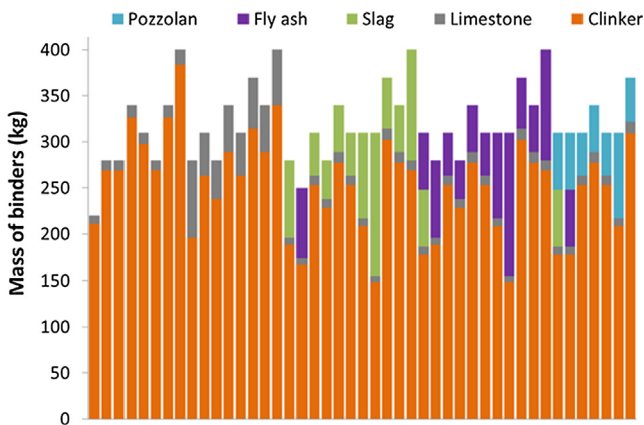


Fig. 6. Summary of the mix-design of 45 concretes.

climate due to higher mean daily temperature prevailing in those regions [31]. However, there is no distinct understanding of the mechanisms.

This paper presents an experimental program that was devised to address the shortcomings addressed above. This is an invaluable international collaborative study that gives valuable insights into the role of the local climatic conditions on the progress of natural carbonation in sheltered and unsheltered conditions.

2. Experimental program

The objective of the experiment was to evaluate the impact of different climates on the natural carbonation of concrete. Together

Table 4
Grading curve of concrete C5.

Sieve [mm]	0	0.16	0.25	0.32	0.4	0.5	1	2	5	8	16	20	25
Passing [%]	0	15	17.5	22	27	31	34	41	57	62	80	92	100

Table 5
Distribution of the production of 45 concretes.

Production number	Date of production	Concrete reference	Number of prisms 100 × 100 × 400 mm per mixture	Number of cylinders 110 × 220 mm per mixture
1	1/11/11	C4, C7, C8	9	3
2	2/8/11	C1, C2, C3, C5*	9	3
3	2/22/11	C6, C9*, C10, C11	9	3
4	3/15/11	C14, C15*, C16	9	3
5	6/7/11	C12, C21, C32	9	3
6	9/6/11	C25, C26, C27	9	3
7	10/4/11	C36*, C37*, C38*	9	3
8	3/14/12	C42*, C45*	9	3
9	3/28/12	C13, C17*, C19, C20	9	3
10	4/18/12	C22, C23, C24, C28	9	3
11	4/26/12	C29*, C30, C18, C31	9	3
12	5/31/12	C33, C34, C35*, C39	9	3
13	6/27/12	C40, C41, C43, C44	9	3



Fig. 7. Concretes exposed to (a) unsheltered and (b) sheltered conditions at Lyon.

with LCR in Lyon (France), 4 partners from Chennai (Madras-India), Austin (Texas-US), Fredericton (New Brunswick-Canada) and Changsha (Hunan-China) have participated in the experimental campaign.

2.1. Climatic conditions in chosen locations

The geographical location and the climate type of each city are presented in Fig. 1. The average, minimum and maximum values of temperature and relative humidity for each location are indicated in Table 12. It is observed that Austin and Changsha are both classified warm temperate, fully humid and hot summer climate. Lyon is of warm temperate, fully humid and warm summer climate. Chennai is of equatorial and winter dry climate while Fredericton is of snow, fully humid and warm summer climate [32].

In order to obtain a detailed evaluation of the real climatic condition of these locations, details on temperature, relative humidity and precipitation are presented in Figs. 2–5. These data were taken from [33]. Data can be provided on a daily, monthly or yearly basis. However, due to the important fluctuation of temperature (T°) and relative humidity (RH) in daily data which makes different curves strongly tangled, only monthly data are presented to better visualize all the data together.

It is observed that the relative humidity is very unstable from one year to another in all 5 locations while it is not the case for

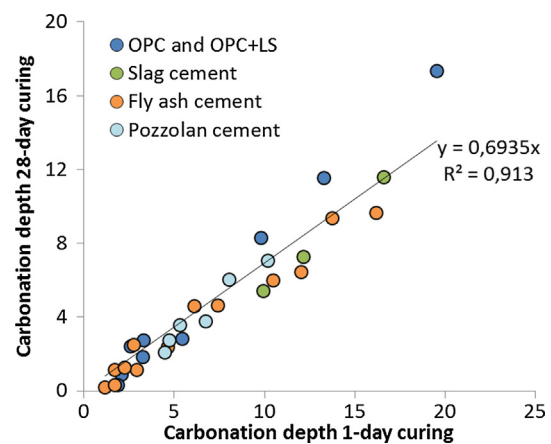


Fig. 8. Carbonation depth (mm) between 1 and 28 day curing (data from Lyon).

the temperature. However, similarity of temperature between Texas and Changsha, a large difference is observed for both relative humidity and temperature between these locations.

The amount of monthly cumulative precipitation is very different between locations (Fig. 4). It is also worth mentioning that the amount of cumulative precipitation as well as the number of rainy

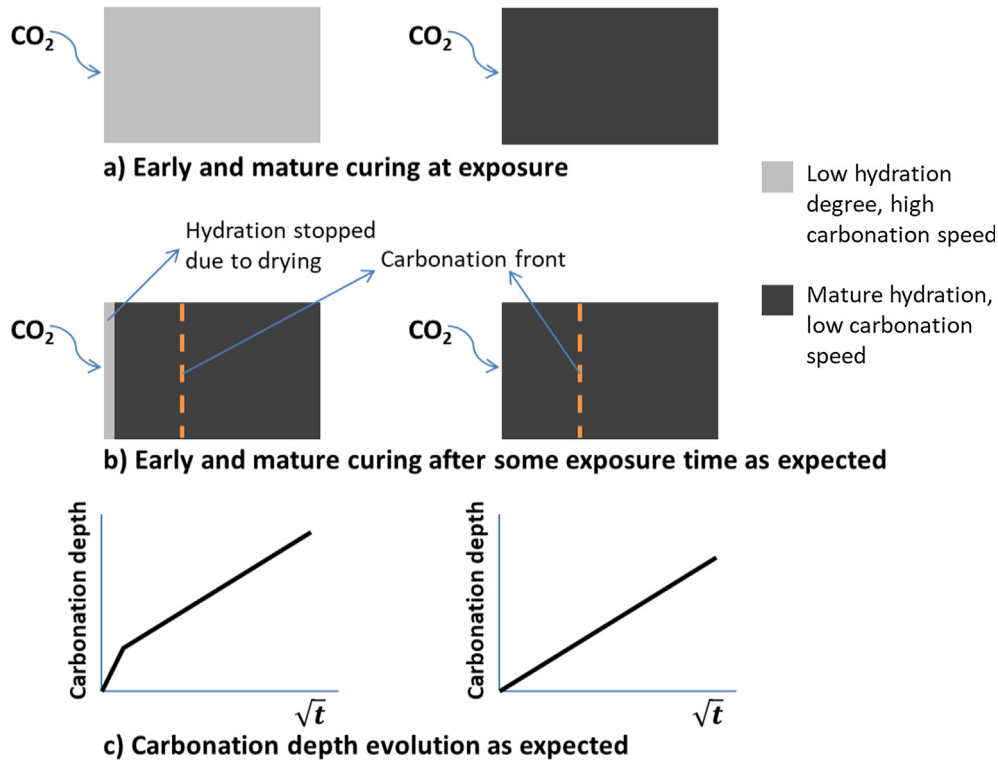


Fig. 9. Natural carbonation and impact of drying/hydration coupling.

days is very unstable from one year to another for all the 5 locations (Fig. 5). No correlation is observed between these two parameters in general. The amount of annual precipitation is quite comparable between Lyon, Fredericton and Austin while it is higher in Chennai and Changsha. The annual precipitation in 2015 for Chennai is abnormal because of the floods. The number of rainy days per year is however low in both Chennai and Austin, but significantly higher in the 3 other locations.

2.2. Materials

Type I 52.5 N Ordinary Portland Cement (OPC), Limestone filler (LS), Ground granulated blast-furnace slag (S), type F fly-ash (FA) from thermoelectric power plant in France, natural pozzolan (PZ) from Greece, a polycarboxylate based superplasticizer and an air entrained agent (for some mixes) were used. The main properties of these cementitious materials are listed in Table 1.

A silico-calcareous rounded aggregate was selected with a maximum diameter of 20 mm. The main properties of the sand and gravel are listed in Table 2.

2.3. Concrete composition and production

Forty-five various compositions were produced at LafargeHolcim Research Center between 2011 and 2012 with varying weff/b ratios (0.45–0.65) and types of binder according to EN 197-1. In addition, various percentages in weight of supplementary cementing materials such as limestone, slag, fly ash and pozzolan were incorporated. Detailed mix designs are presented in the Appendix (Table 6). Some main information concerning the mix-design of concretes is summarized in Fig. 6 and Table 3.

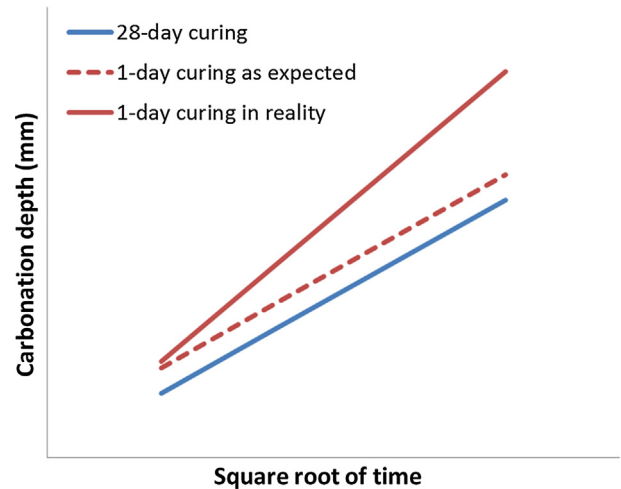


Fig. 10. Illustration of carbonation depth evolution for 1 day and 28 day curing.

Concretes were designed from the Faury method with a targeted consistency S4 (160–210 mm). The grading curve of concrete C5 was used as a reference for the other 44 concretes (Table 4).

Concretes were produced in a concrete pilot plant with a mixer capacity of 120 L. Prior to mixing, aggregates were previously dried at 110 ± 5 °C until a constant mass was reached (i.e. two successive weighing separated by at least 1 h did not differ by more than 0.1%). The aggregates were mixed for 4 min by adding the pre-wetting water for the first 30 s. The binders were then introduced and mixed with the aggregates for 1 min. The mixing water con-

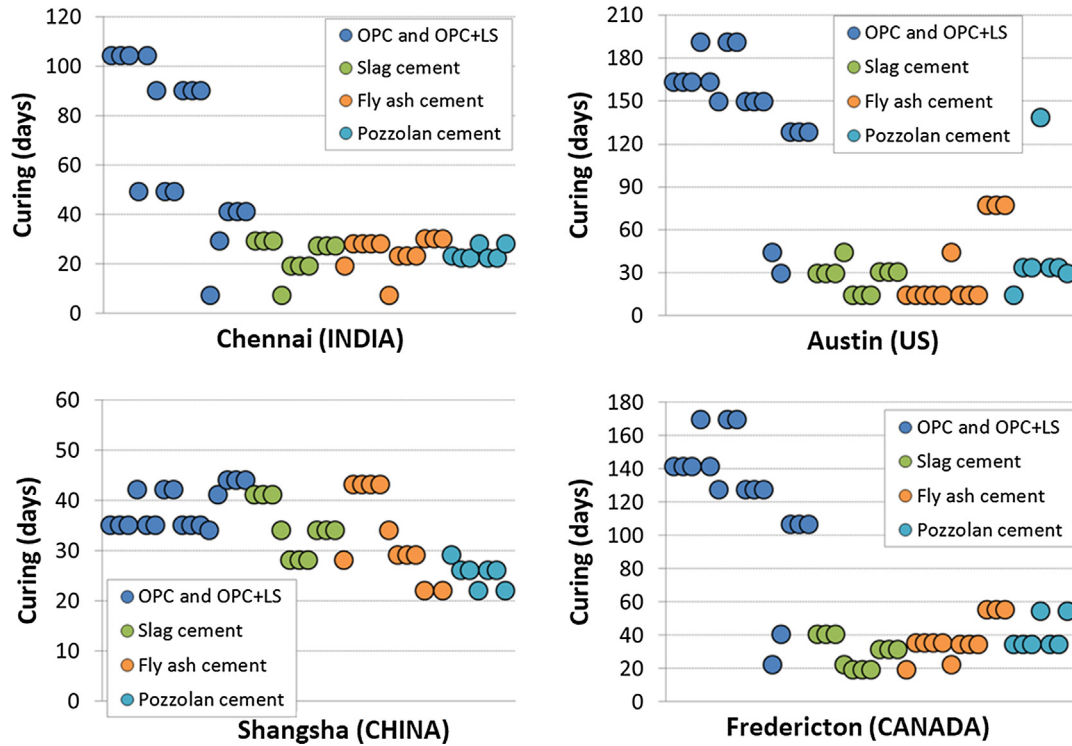


Fig. 11. Curing age of concretes before exposure for different locations.

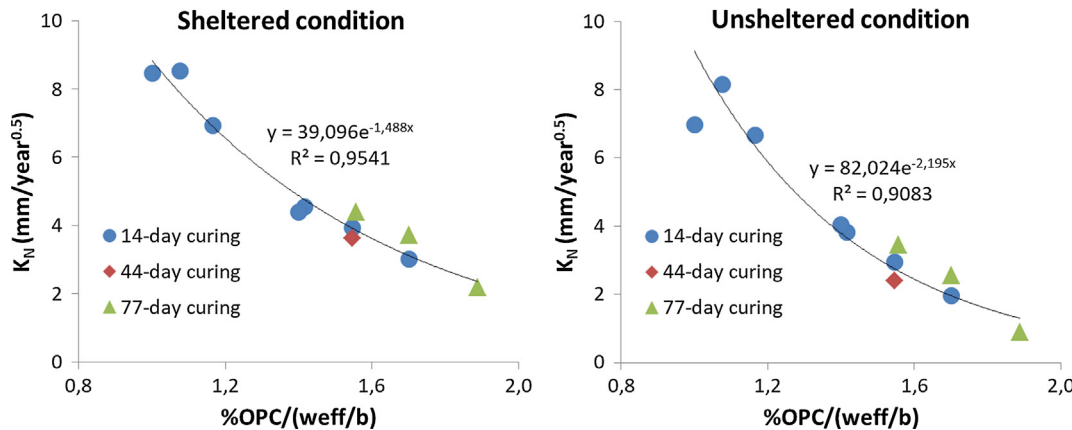


Fig. 12. Carbonation rate of fly cement concretes as a function of OPC content to water ratio (US data).

taining admixtures was then added and mixed for an additional 2 min.

2.4. Specimen preparation and test methods

The following tests on fresh concretes were performed: consistency at 5 min with the slump test according to EN 12350-2; and entrapped air at 5 min according to EN 12350-7.

The following specimens were produced for each mixture: three 110 × 220 mm cylindrical specimens for compressive strength at 28 days in accordance with EN 12390-3; nine 100 × 100 × 400 mm prismatic specimens to determine the natural carbonation depth.

For logistical reasons, the 45 concretes were cast in 13 batches running from the beginning of 2011 to the middle of 2012. Each production thus includes 2–4 concretes for all the laboratories as indicated in Table 5.

2.4.1. Natural carbonation test

Following 24 h of curing under wet burlap and plastic, all specimens were demolded. One 100 × 100 × 400 mm prismatic specimen per concrete from C1 to C45 was placed outside at LCR and unprotected from direct rainfall and another one protected from rainfall (Fig. 7). This condition corresponds to Lyon climate with a 1 day curing time in unsheltered and sheltered conditions respectively. Among these concretes, 11 ones (mix designs with

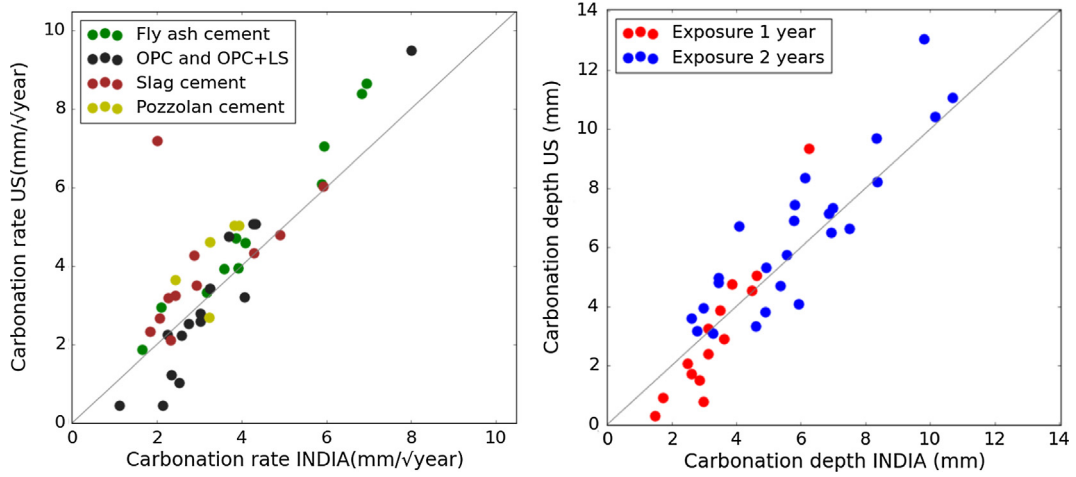


Fig. 13. Comparison of carbonation in sheltered condition between Chennai and Austin.

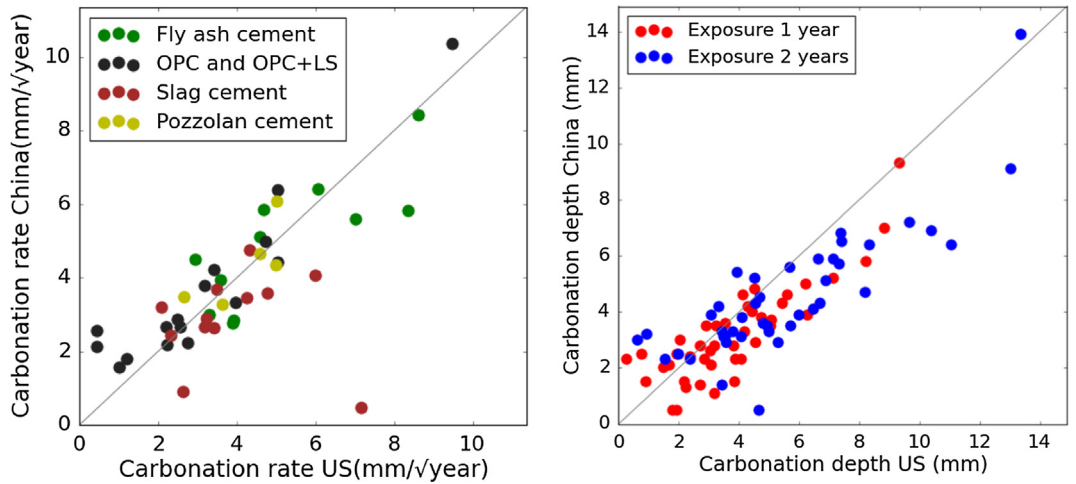


Fig. 14. Comparison of carbonation in sheltered condition between Austin and Changsha.

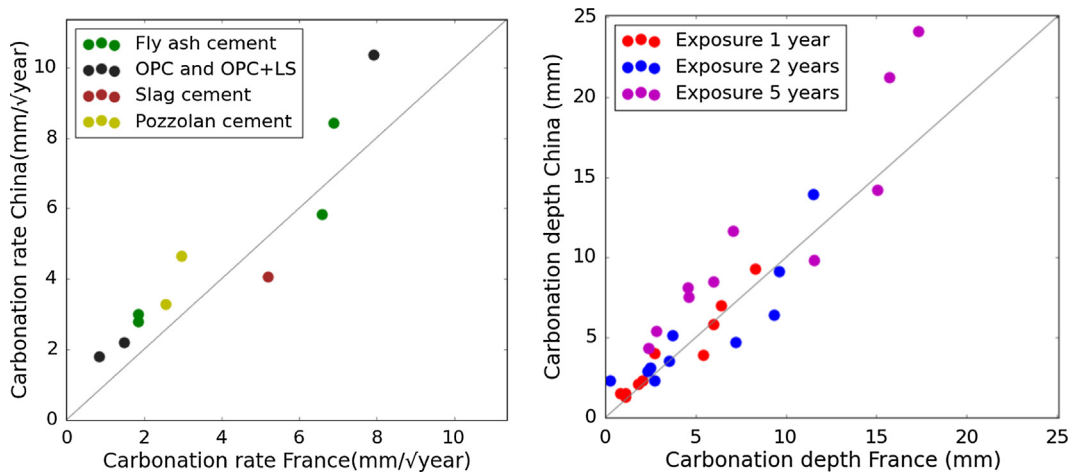


Fig. 15. Comparison of carbonation in sheltered condition between Lyon and Changsha.

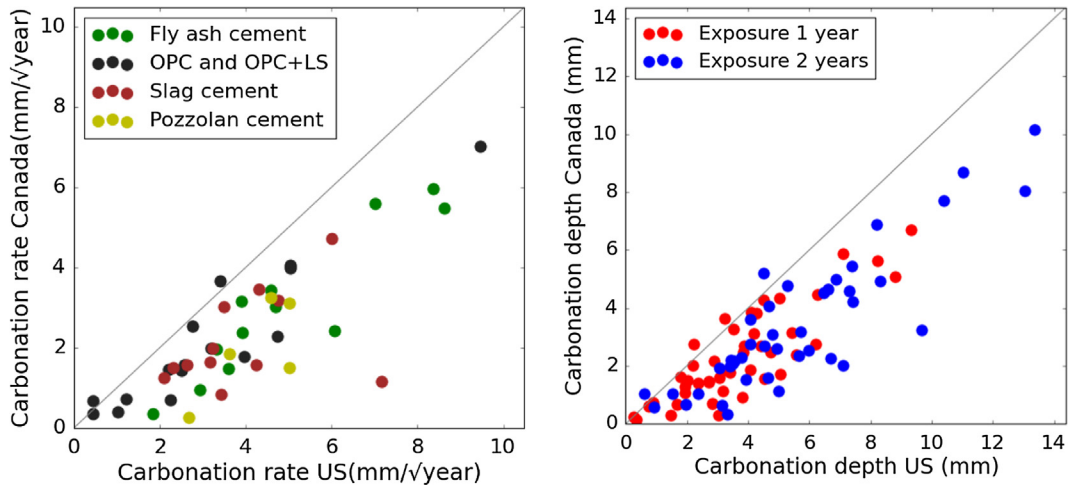


Fig. 16. Comparison of carbonation in sheltered condition between Austin and Fredericton.

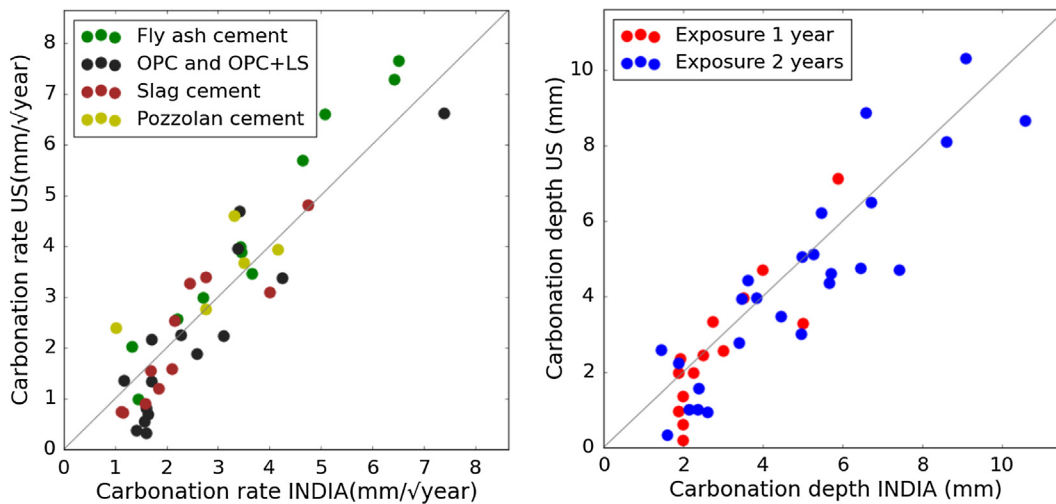


Fig. 17. Comparison of carbonation in unsheltered condition between Chennai and Austin.

an asterisk in Table 5) were chosen to produce an additional specimen per mix. They were then wrapped in a damp cloth and stored in a sealed plastic bag at 20 ± 1 °C for 28 days and then placed outside at LCR and protected from direct rainfall. This condition corresponds to Lyon climate with a 28-day curing time in sheltered conditions. The four other sets of 45 specimens were individually wrapped in a damp cloth, 24 h hours after demolding and placed in a sealed plastic bag and delivered to the other countries. Upon arrival at the other research laboratories, specimens were exposed for 5 years in both sheltered (outside and protected from direct rainfall) and unsheltered conditions except Fredericton where only sheltered exposure was possible. Exposure of the same concretes in sheltered and unsheltered conditions always started at the same time for each location.

During exposure concrete samples were placed vertically. Following 1, 2 or 3, and 5 years of exposure, a slice (approximately 80–100 mm thick) was broken off each prism. The freshly broken surface of the split slice is then brushed and the depth of carbona-

tion was determined by spraying the freshly fractured surface with a 0.5% phenolphthalein solution. Following $1 \text{ h} \pm 5 \text{ min}$, a picture of the colored surface was taken and the carbonation depth was determined with a ruler at 5 equidistant points on each of the four sides of the slice giving a total of twenty measurement points. Measurements were not made within 15 mm of the corners. If the measurement falls to an aggregate, it is extrapolated on both sides of this aggregate. Before putting the sample back, the fractured surface was protected with a sealing product. The average carbonation depth for each side was calculated and the average carbonation depth of the concrete mix is reported.

3. Assessment of experimental results

In this section, the carbonation results from the considered climates as well as several aspects potentially impacting on carbonation are investigated.

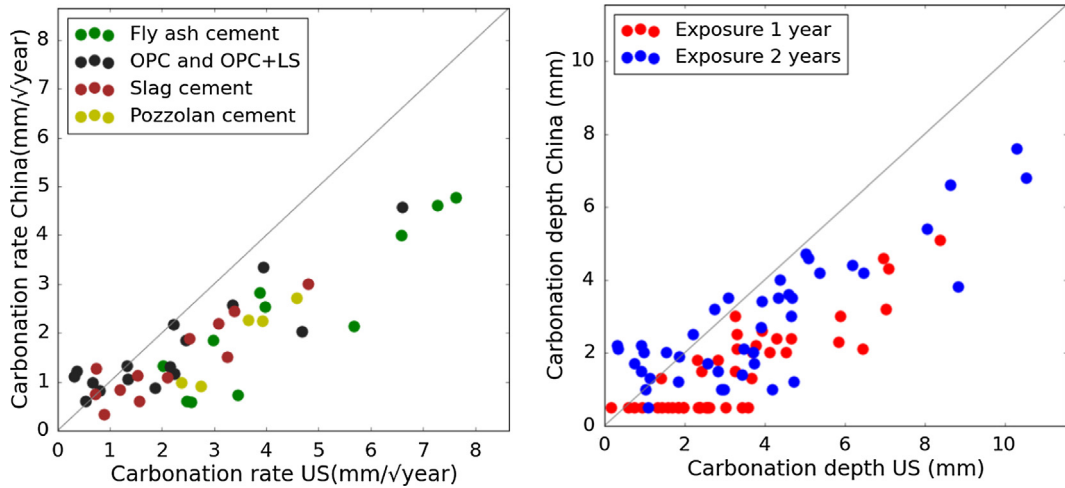


Fig. 18. Comparison of carbonation in unsheltered condition between Austin and Changsha.

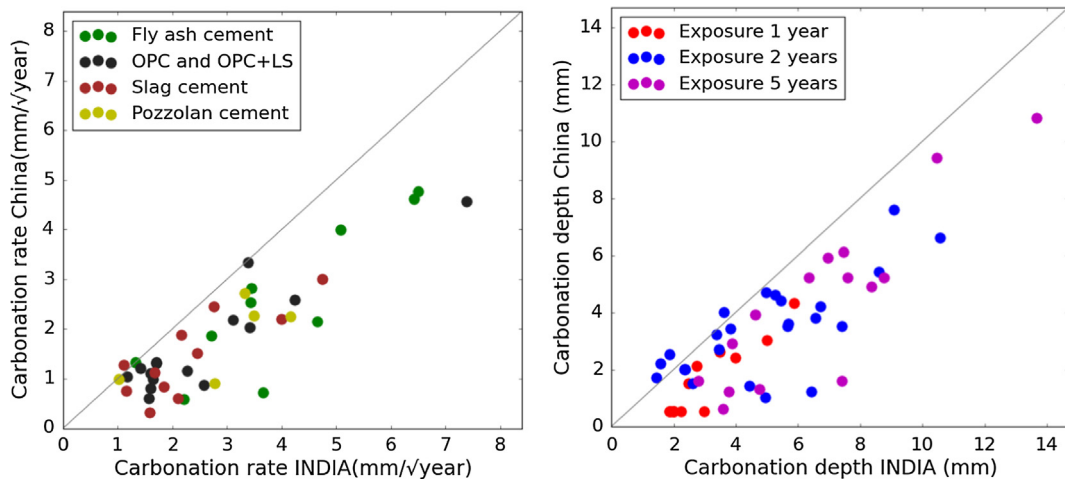


Fig. 19. Comparison of carbonation in unsheltered condition between Changsha and Chennai.

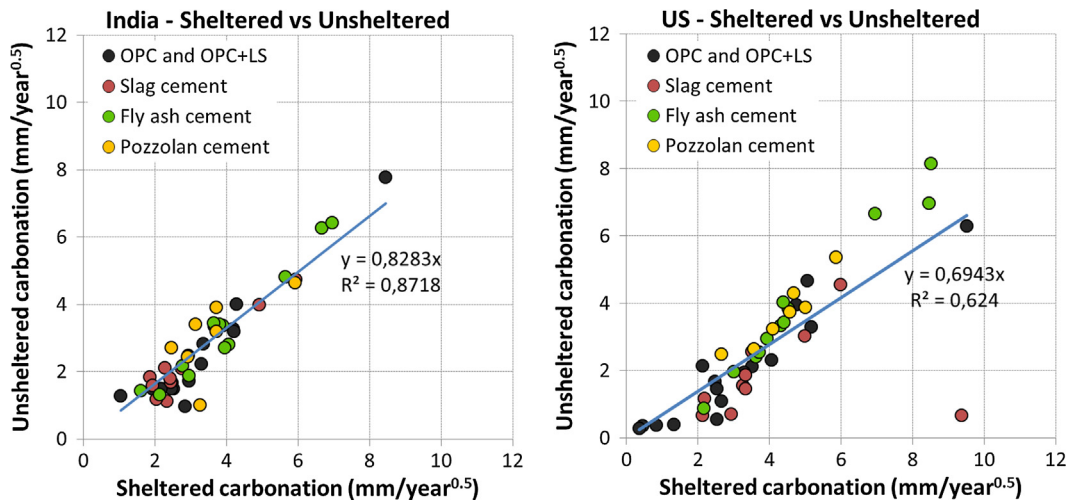


Fig. 20. Comparison of carbonation speed between sheltered and unsheltered condition for Chennai and Austin.

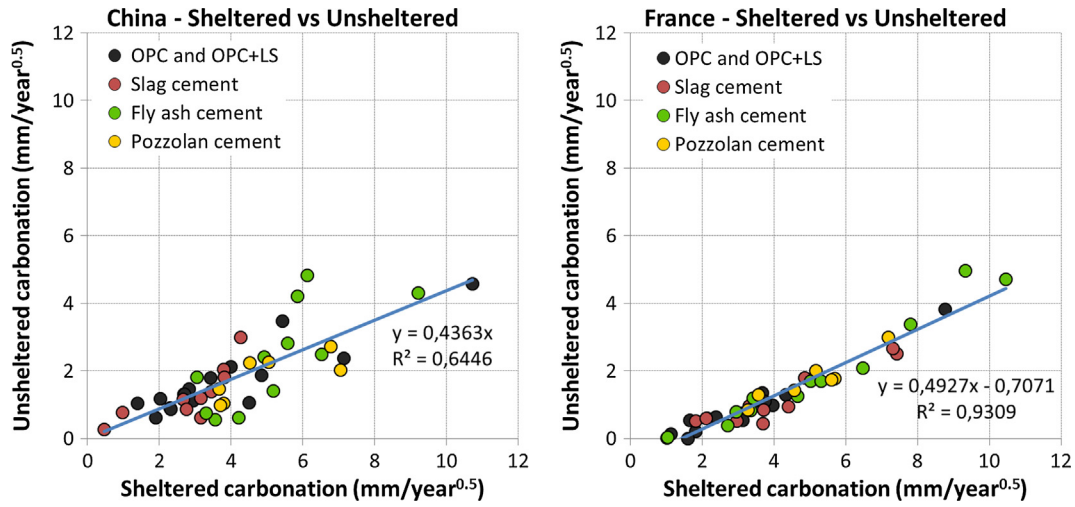


Fig. 21. Comparison of carbonation speed between sheltered and unsheltered condition for Changsha and Lyon.

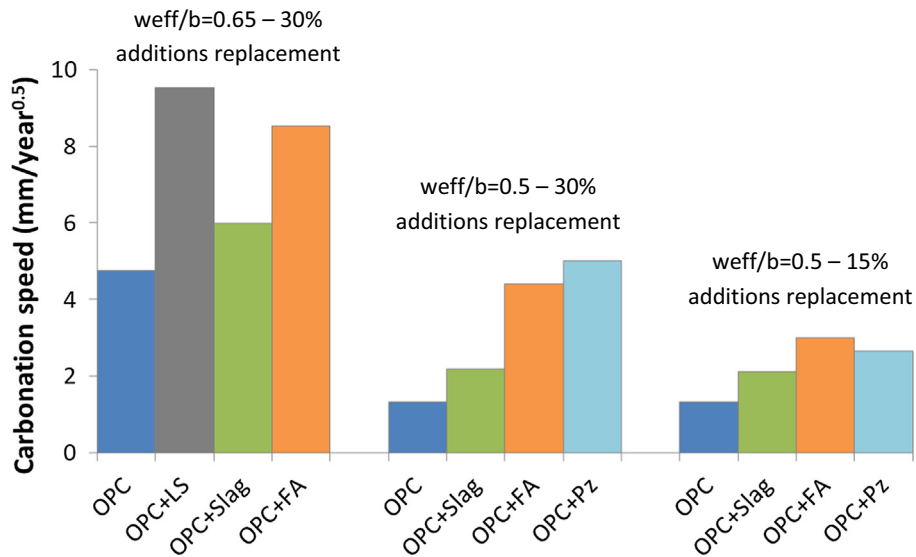


Fig. 22. US sheltered condition: impact of supplementary cementing materials at different weff/b ratios.

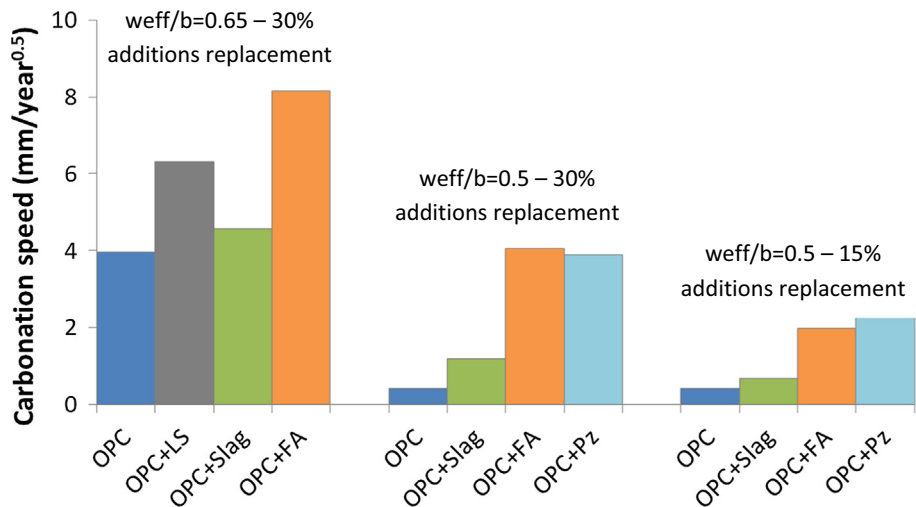


Fig. 23. US unsheltered condition: Impact of supplementary cementing materials at different weff/b ratios.

3.1. Impact of curing

3.1.1. Early curing age

The study of the impact of curing was intentionally realized only for Lyon climate. In Lyon, concretes were exposed after 1 day curing for unsheltered condition. For sheltered condition, 2 curing ages of 1 and 28 days were considered. Among 45 concretes which were all exposed after 1 day curing as for unsheltered condition, 11 concretes were chosen to produce additional specimens that were exposed later after 28 day curing. The comparison of carbonation depth up to 5 year exposure between 1 and 28 day curing for these 11 concretes is presented in Fig. 8. It is observed that the carbonation depth is much higher for 1 day curing than for 28 day curing. Moreover, the carbonation depth between 1 and 28 day curing is proportional with almost the same proportional factor for different types of cement.

Similar results can be found in [12] for natural carbonation in lab controlled conditions. However, the authors have not shown

explicitly the proportionality of carbonation depth between 1 and 28 day curing which appears as a surprising result.

Indeed, early age curing should impact the region close to the concrete surface where hydration will be stopped due to drying (Fig. 9a and b). As the drying of concrete is a very long and slow process [13] the impacted region was expected to be limited. At further depths far enough from the concrete surface, hydration should continue to reach the maturity during exposure, regardless of initial curing. The carbonation speed is thus expected to be faster at the beginning when carbonation progresses through the poorly cured depth and to stabilize when carbonation reaches well cured depths (Fig. 9c). Therefore, the difference of carbonation depth between 1 and 28 day curing should be generated only at the beginning of exposure. At later exposure ages, carbonation is expected to progress at a constant speed for both 1 and 28 day curing, which means the carbonation depth evolution curves should be parallel (Fig. 10). Nevertheless, this is not the case in reality as

Table 6
Concrete mix designs and properties on fresh and hardened concretes.

Concrete [-]	Addition [%]	w_{eff}/b [-]	M_{binder} [kg/m ³]	M_{sand} [kg/m ³]	M_{gravel} [kg/m ³]	Air content [%]	f_{cm} [MPa]
C1	0	0.65	220	1042	963	3.4	34.8
C2	0	0.65	280	964	890	1.8	34.1
C3	0	0.55	280	1002	923	2.4	46.0
C4	0	0.55	340	932	868	2.0	36.3
C5	0	0.5	310	987	910	2.0	48.7
C6	0	0.5	280	1018	940	2.0	52.2
C7	0	0.45	340	904	904	6.0	40.8
C8	0	0.45	400	874	815	3.9	42.0
C9	30% LS	0.65	280	960	883	1.5	23.1
C10	11% LS	0.6	310	945	868	1.2	33.4
C11	11% LS	0.55	280	1000	919	2.8	42.0
C12	11% LS	0.55	340	923	858	1.5	37.1
C13	11% LS	0.5	310	1007	879	1.8	48.5
C14	11% LS	0.5	370	879	809	4.0	34.6
C15	11% LS	0.45	340	934	860	4.6	44.6
C16	11% LS	0.45	400	873	801	4.1	37.3
C17	30% S	0.65	280	983	859	1.2	37.5
C18	30% FA	0.6	250	1031	900	2.2	32.5
C19	15% S	0.6	310	965	843	1.0	40.3
C20	15% S	0.55	280	1023	892	1.6	47.2
C21	15% S	0.55	340	922	861	1.5	42.3
C22	15% S	0.5	310	1007	881	1.5	56.4
C23	30% S	0.5	310	1007	879	1.4	55.7
C24	50% S	0.5	310	1005	877	1.6	47.7
C25	15% S	0.5	370	870	829	5.2	30.7
C26	15% S	0.45	340	926	870	6.6	37.5
C27	30% S	0.45	400	862	811	4.8	30.0
C28	20% S + 20% FA	0.5	310	1002	870	1.4	41.1
C29	30% FA	0.65	280	976	850	0.9	26.7
C30	15% FA	0.6	310	962	839	1.0	37.5
C31	15% FA	0.55	280	1020	887	1.9	43.7
C32	15% FA	0.55	340	920	855	1.2	38.6
C33	15% FA	0.5	310	1002	873	1.5	46.0
C34	30% FA	0.5	310	996	869	1.3	37.5
C35	50% FA	0.5	310	988	861	1.5	29.7
C36	15% FA	0.5	370	887	796	4.5	32.2
C37	15% FA	0.45	340	944	845	5.5	39.3
C38	30% FA	0.45	400	873	782	5.3	30.0
C39	20% S + 20% PZ	0.5	310	997	869	1.6	40.4
C40	20% FA + 20% PZ	0.5	310	991	864	1.8	30.6
C41	15% PZ	0.6	310	959	838	1.2	30.8
C42	15% PZ	0.55	340	944	824	1.5	34.2
C43	15% PZ	0.5	310	1001	873	1.4	43.4
C44	30% PZ	0.5	310	995	867	1.6	34.7
C45	13% PZ	0.5	370	893	779	3.4	33.9

the carbonation depth always remains proportional between 1 and 28 day curing, regardless of the exposure age.

The most likely explanation for the proportionality of carbonation depth between 1 day and 28 day curing is that the carbonation front following 1 day curing always remains within the poorly cured region impacted by drying. This suggests that the impacted region is not as limited as believed but much larger, up to 20 mm in this case. To explain how drying could reach immediately such an important concrete depth and stop hydration there, a hypothesis was made that at very early ages, the size of pores within paste remains as large as capillary drying can occur. Capillary drying is much faster than diffusion drying and can reach significant depths very quickly [14]. The hydration process is then drastically slowed or stopped within the whole concrete depth affected by capillary drying.

It is worth mentioning that the huge impact of early age curing on carbonation as observed here may not remain valid in unsheltered condition. Indeed, rain water penetrating into concrete can result in a recovery of hydration within the dried depth. In this case, the difference of carbonation depth between early age and advanced age curing is expected to be reduced significantly.

3.1.2. Advanced curing ages

Due to shipment duration and technical reasons, concrete age at exposure was varying between countries and between different concretes as shown in Fig. 10. Since concrete specimens remained covered with damp cloth in sealed plastic bags during shipment and storage until exposure, they can be considered to be in wet curing during this period.

Table 7
Natural carbonation depths in sheltered and unsheltered conditions in Lyon.

Concrete	Lyon (France)										
	Curing time	Sheltered			Unsheltered			Curing time	Sheltered		
		[days]	X _{C1year} [mm]	X _{C2years} [mm]	X _{C5years} [mm]	X _{C1year} [mm]	X _{C2years} [mm]		X _{C5years} [mm]	[days]	X _{C1year} [mm]
C1	1	5.0	6.5	8.8	1.9	1.7	2.3	–	–	–	–
C2	1	4.2	5.1	8.3	2.4	2.3	3.1	–	–	–	–
C3	1	2.8	2.9	4.1	0.2	0.3	0.5	–	–	–	–
C4	1	3.2	3.8	4.8	1.2	0.9	1.4	–	–	–	–
C5	1	2.1	1.9	2.6	0.0	0.1	0.3	28	0.9	0.3	2.4
C6	1	2.5	3.0	3.6	0.0	0.0	0.0	–	–	–	–
C7	1	1.3	2.4	1.8	0.2	0.0	0.0	–	–	–	–
C8	1	1.5	1.6	2.3	0.2	0.2	0.0	–	–	–	–
C9	1	9.8	13.3	19.6	5.5	7.3	8.4	28	8.3	11.5	17.4
C10	1	5.1	6.5	9.7	2.3	2.3	2.9	–	–	–	–
C11	1	4.2	4.8	7.0	1.0	1.1	1.7	–	–	–	–
C12	1	3.9	4.7	8.5	1.6	1.9	2.5	–	–	–	–
C13	1	3.9	4.7	7.0	0.9	0.7	1.3	–	–	–	–
C14	1	4.2	5.1	7.7	1.9	1.9	2.5	–	–	–	–
C15	1	3.3	3.3	5.5	1.1	0.7	1.5	28	1.8	2.7	2.8
C16	1	3.1	3.4	3.7	1.7	1.2	1.3	–	–	–	–
C17	1	10.0	12.2	16.6	4.4	4.7	5.6	28	5.4	7.3	11.6
C18	1	9.7	11.6	17.6	4.7	5.3	7.6	–	–	–	–
C19	1	6.5	7.3	11.0	2.4	2.7	4.0	–	–	–	–
C20	1	5.8	6.3	10.1	1.9	1.1	2.3	–	–	–	–
C21	1	3.8	4.9	7.4	1.2	1.7	2.1	–	–	–	–
C22	1	5.2	4.5	6.9	0.7	0.6	1.2	–	–	–	–
C23	1	5.1	6.2	8.3	1.2	1.5	1.9	–	–	–	–
C24	1	9.2	11.8	16.3	3.3	3.5	6.1	–	–	–	–
C25	1	2.4	2.8	8.6	1.0	0.6	1.1	–	–	–	–
C26	1	2.8	2.7	4.2	1.2	0.9	1.2	–	–	–	–
C27	1	2.7	3.2	4.7	1.6	1.0	1.4	–	–	–	–
C28	1	7.8	10.7	14.3	3.2	3.5	4.7	–	–	–	–
C29	1	10.5	13.8	20.9	6.9	8.2	11.1	28	6.0	9.4	15.1
C30	1	6.0	7.7	11.2	2.5	2.8	3.8	–	–	–	–
C31	1	5.4	7.1	10.4	2.3	2.0	2.9	–	–	–	–
C32	1	4.5	5.1	7.8	1.9	1.7	2.8	–	–	–	–
C33	1	3.4	4.5	6.6	1.0	1.4	1.7	–	–	–	–
C34	1	6.1	8.3	11.8	2.4	2.4	3.9	–	–	–	–
C35	1	12.0	16.2	23.3	6.7	8.5	10.4	28	6.4	9.7	15.7
C36	1	1.7	2.8	6.1	1.0	0.5	0.9	28	1.2	2.5	4.6
C37	1	1.2	1.7	2.3	0.3	0.1	0.1	28	0.2	0.3	1.3
C38	1	2.9	4.7	7.4	0.9	1.1	1.9	28	1.1	2.4	4.7
C39	1	6.8	9.0	12.7	2.8	3.4	3.9	–	–	–	–
C40	1	8.2	10.7	16.1	3.9	4.8	6.7	–	–	–	–
C41	1	5.5	7.4	11.6	2.7	3.3	4.5	–	–	–	–
C42	1	4.7	6.8	10.2	2.4	2.7	3.2	28	2.7	3.8	7.1
C43	1	3.7	5.0	7.3	1.1	1.3	1.9	–	–	–	–
C44	1	6.1	8.4	12.5	2.9	3.0	3.9	–	–	–	–
C45	1	4.5	5.3	8.0	1.7	2.3	2.8	28	2.1	3.6	6.0

For advanced curing ages, capillary drying is unlikely to occur but diffusion drying occurs as pore sizes are well densified. Since diffusion drying is a very long process, it is believed that the impact of curing would be little because only a thin depth at the concrete surface can be dried before hydration degree reaches the maturity within the whole sample during exposure. To demonstrate this, consider the case of concretes with fly ash cement exposed in Austin (US) in Fig. 11. The variation of curing age is very large, from 14 days to 77 days. It worth noting that the hydration degree of fly ash can be neglected for 14 day curing while it is about $30\% \pm 40\%$ at 77 day curing [15]. Fig. 12 shows carbonation rate plotted as a function of OPC content to water ratio $\alpha = \frac{\%OPC}{(w_{eff}/b)}$. It is observed that the higher the OPC content to water ratio, the lower the carbonation rate. A very good trend is found for both sheltered and unsheltered condition, regardless of curing age. This means that the carbonation speed depends only on the concrete

mix-design, not on the curing age. It can be concluded from this finding that for advanced curing age from 14 days in this case, the curing has no impact on the natural carbonation of concretes with fly ash.

The same result as above was found for other cement types and other countries. However, either the difference in curing age between concretes is much less significant or the hydration degree is already high within the curing age range. Given the fact that fly ash may be the addition whose hydration degree is the most sensitive to curing age, it is assumed that the same conclusion on the impact of curing remains valid for other cement types.

This finding is interesting as it allows for the comparison between the carbonation of concrete with different climates despite the difference in curing age, as long as the curing age is advanced enough.

Table 8

Natural carbonation depths in sheltered and unsheltered conditions in Chennai.

Concrete	Chennai (India)								
	Curing time	Sheltered				Unsheltered			
		[days]	Xc _{1year} [mm]	Xc _{2years} [mm]	Xc _{3years} [mm]	Xc _{5years} [mm]	Xc _{1year} [mm]	Xc _{2years} [mm]	Xc _{3years} [mm]
C1	104	4.6	–	7.2	–	4.0	–	5.6	–
C2	104	3.9	–	6.3	–	3.5	–	5.8	–
C3	104	3.1	–	4.2	–	1.9	–	2.9	–
C4	49	3.5	–	5.0	–	3.0	–	4.2	–
C5	104	1.8	–	4.4	–	1.9	–	2.6	–
C6	90	1.5	–	1.8	–	2.0	–	2.1	–
C7	49	3.0	–	3.2	–	2.0	–	2.5	–
C8	49	2.9	–	4.2	–	2.3	–	2.5	–
C9	90	6.3	–	14.9	–	5.9	–	13.7	–
C10	90	4.5	–	7.8	9.4	5.0	–	7.8	8.8
C11	90	3.1	–	5.7	–	2.5	–	3.8	–
C12	7	–	–	6.0	7.4	–	–	4.9	6.3
C13	29	–	4.9	–	6.4	–	2.4	–	3.9
C14	41	3.6	–	7.3	–	2.8	–	5.6	–
C15	41	2.6	–	3.7	–	2.0	–	2.6	–
C16	41	2.5	–	5.0	–	1.9	–	1.6	–
C17	29	–	8.4	–	–	–	6.7	–	–
C18	28	–	10.2	–	12.2	–	8.6	–	10.5
C19	29	–	7.0	–	–	–	5.7	–	–
C20	29	–	3.5	–	–	–	2.4	–	–
C21	7	–	–	4.2	–	–	–	3.1	–
C22	19	–	3.3	–	–	–	1.6	–	–
C23	19	–	2.6	–	–	–	2.6	–	–
C24	19	–	5.4	–	5.9	–	3.4	–	4.6
C25	27	–	–	4.1	5.0	–	–	3.6	4.8
C26	27	–	–	3.8	4.4	–	–	1.8	2.8
C27	27	–	–	4.3	3.9	–	–	2.7	3.6
C28	19	–	7.5	–	8.7	–	3.6	–	6.4
C29	28	–	10.7	–	14.6	–	10.6	–	13.7
C30	28	–	7.0	–	8.4	–	5.3	–	7.5
C31	28	–	5.6	–	–	–	3.8	–	–
C32	7	–	–	4.8	–	–	–	3.8	–
C33	23	–	3.0	–	–	–	1.9	–	–
C34	23	–	5.8	–	8.4	–	5.0	–	7.6
C35	23	–	9.8	–	–	–	9.1	–	–
C36	30	–	5.9	–	6.2	–	5.0	–	3.8
C37	30	–	2.8	–	3.5	–	2.1	–	3.2
C38	30	–	4.9	–	8.1	–	6.4	–	7.4
C39	23	–	4.1	–	–	–	3.5	–	–
C40	22	–	8.3	–	–	–	6.6	–	–
C41	22	–	6.1	–	8.1	–	5.5	–	7.0
C42	28	–	5.8	4.7	–	–	5.7	5.5	–
C43	22	–	4.6	–	–	–	1.4	–	–
C44	22	–	6.9	–	8.0	–	7.4	–	8.4
C45	28	–	3.5	4.3	–	–	4.5	4.4	–

3.2. Impact of climatic conditions

3.2.1. Sheltered condition

As mentioned above, the curing age before exposure is strongly different from one country to another. However, it was also shown that for advanced curing age beyond 14 days, the curing had no impact on the natural carbonation. Therefore, the comparison of carbonation between two locations on the same concrete could be made as long as the initial curing is not lower than 14 days in both locations.

The comparison results are presented in Figs. 13–16. It is found that the carbonation is globally comparable between Chennai, Austin, Changsha and Lyon for both carbonation rate and carbonation depth despite some noticeable difference in climatic condi-

tions. In general, the trend is consistent between different cement types as well as between different ages. To explain this result, it should be considered that temperature and relative humidity (RH) can impact carbonation in two opposite ways. Indeed, increase of temperature speeds up the carbonation progress while for RH above approximately 50%–60%, the carbonation rate decreases as CO₂ diffusivity decreases. Therefore, if the impact of T° and RH is compensating between these climates, carbonation will remain similar.

While carbonation is comparable between Chennai, Austin, Changsha and Lyon, it is significantly lower in Fredericton (Fig. 15). This can be explained by the very low temperature there. However, Fredericton results appear quite scattered compared to other locations as no clear trend can be found.

Table 9
Natural carbonation depths in sheltered and unsheltered conditions in Austin.

Concrete	Austin (US)						
	Curing time	Sheltered			Unsheltered		
[–]	[days]	X _{C1year} [mm]	X _{C2years} [mm]	X _{C5years} [mm]	X _{C1year} [mm]	X _{C2years} [mm]	X _{C5years} [mm]
C1	163	5.1	6.5	7.7	4.7	5.2	4.7
C2	163	4.8	5.6	9.0	4.0	3.8	3.3
C3	163	2.4	3.5	4.1	2.0	3.5	4.1
C4	191	3.9	4.1	4.9	2.6	2.9	3.3
C5	163	0.9	1.6	2.7	1.0	1.1	0.7
C6	149	0.3	0.9	0.8	0.2	0.9	0.6
C7	191	0.8	0.6	0.7	0.6	0.4	0.6
C8	191	1.5	2.0	1.5	2.0	0.8	0.9
C9	149	9.3	13.4	19.1	7.1	10.6	12.1
C10	149	4.5	7.4	10.2	3.3	5.4	6.3
C11	149	3.3	4.5	7.2	2.4	3.8	4.0
C12	44	3.4	6.0	8.0	2.3	4.7	4.1
C13	29	2.7	3.8	5.0	1.4	1.0	3.3
C14	128	2.9	4.6	6.6	3.3	3.1	4.0
C15	128	1.7	2.4	5.4	1.4	1.9	0.9
C16	128	2.1	3.4	5.4	2.4	1.9	2.2
C17	29	6.3	8.2	12.1	5.9	6.5	9.3
C18	14	7.1	10.4	13.7	7.0	8.1	13.8
C19	29	4.2	6.5	10.1	3.3	4.4	6.1
C20	29	3.1	4.8	6.4	1.9	1.6	3.4
C21	44	3.8	5.0	6.6	2.6	3.7	3.5
C22	14	2.0	3.1	4.2	1.5	0.3	1.7
C23	14	2.7	3.6	4.3	1.7	0.9	2.7
C24	14	3.6	4.7	7.1	2.9	2.8	5.5
C25	30	3.2	3.6	7.1	1.6	2.9	2.7
C26	30	1.8	3.5	6.0	0.8	1.2	1.4
C27	30	2.0	4.7	20.9	1.9	1.1	1.4
C28	14	4.1	6.6	8.4	3.8	4.4	6.8
C29	14	8.2	11.1	17.4	7.0	8.7	17.2
C30	14	4.3	7.3	8.7	4.3	5.1	7.8
C31	14	3.9	5.7	7.8	3.3	4.0	6.0
C32	44	3.2	5.7	7.0	2.4	4.2	4.5
C33	14	2.9	4.0	6.1	2.6	2.2	4.2
C34	14	5.4	7.4	8.5	4.1	5.0	8.3
C35	14	8.8	13.1	16.6	8.4	10.3	14.0
C36	77	2.2	4.1	7.7	3.0	3.0	5.4
C37	77	0.4	3.2	4.1	1.6	1.0	2.0
C38	77	2.2	5.3	8.9	3.6	4.7	7.0
C39	14	4.6	6.7	7.9	3.7	3.9	6.8
C40	33	6.2	9.7	11.3	6.5	8.9	10.4
C41	33	5.6	8.3	8.9	5.9	6.2	8.8
C42	138	4.4	6.9	9.0	3.7	4.6	7.7
C43	33	3.1	3.3	5.5	2.4	2.6	5.3
C44	33	5.1	7.1	10.0	4.6	4.7	8.1
C45	29	4.1	5.0	7.2	3.5	3.5	5.5

3.2.2. Unsheltered condition

For unsheltered condition, comparison was made only between Chennai, Austin and Changsha, because concretes were all exposed after 1 day curing in Lyon, while exposure was not realized in Fredericton.

For concretes exposed at advanced curing ages, the impact of unsheltered exposure on carbonation, compared to sheltered exposure, relies mainly on the fact that rain water saturates the concrete surface and prevents CO₂ from penetrating into concrete. It is interesting to see that carbonation remains comparable between Chennai and Austin for both sheltered and unsheltered conditions (Figs. 13 and 17). Previously, it was shown that the amount of annual precipitation is higher in Chennai than in Austin while the number of rainy days is quite similar between both locations (Fig. 5). This suggests that the number of rainy days may be a relevant parameter impacting the carbonation of concrete.

The carbonation is lower in Changsha than in Chennai and Austin. This is consistent with the fact that both the amount of annual precipitation and the number of rainy days are higher in Changsha. It can also be noted that the result appears quite scattered for Changsha, especially for carbonation depth (Figs. 17–19).

3.2.3. Comparison between sheltered and unsheltered condition

The comparison between carbonation rate with respect to both sheltered and unsheltered conditions is presented in Figs. 20 and 21, respectively. It can be seen that the carbonation rate is much lower in the unsheltered condition than in the sheltered condition as the saturated concrete surface and prevents CO₂ from penetrating into the concrete. However, the degree of impact of rain is very different from one location to another.

For early age curing concrete, the impact of rain may be two-fold. Indeed, in being exposed to the environment, a certain con-

Table 10
Natural carbonation depths in sheltered and unsheltered conditions in Changsha.

Concrete	Changsha (China)						
	Curing time [days]	Sheltered			Unsheltered		
		X _{C1year} [mm]	X _{C2years} [mm]	X _{C5years} [mm]	X _{C1year} [mm]	X _{C2years} [mm]	X _{C5years} [mm]
C1	35	3.5	5	11.1	2.4	3.3	4.1
C2	35	3.8	6	12.3	2.6	5	7.6
C3	35	2.4	3.1	6.5	0.5	2.1	3.1
C4	42	1.5	3.8	4.9	0.5	1.5	1.9
C5	35	1.5	2.3	4.3	0.5	1	1.3
C6	35	2.3	3.2	6.1	0.5	2.2	2.7
C7	42	2.5	3	4.6	0.5	2.1	2.4
C8	42	2	2.5	3.1	0.5	1.7	2.2
C9	35	9.3	13.9	24.1	4.3	6.8	10.1
C10	35	4.8	6.8	16.4	3	4.2	5.2
C11	35	3.5	5.2	10.2	1.5	1.7	2.4
C12	34	3.4	3.9	7.9	1.8	3	3.9
C13	41	2.8	3.3	6.2	1.3	2	2.9
C14	44	3.5	4.3	9.2	2.1	3.5	4.6
C15	44	2.1	2.3	5.4	0.5	1.2	1.9
C16	44	3	3.3	6.8	0.5	1.9	2.3
C17	41	3.9	4.7	9.8	3	4.2	6.7
C18	43	5.2	6.9	13.3	3.2	5.4	9.4
C19	41	3.3	4.1	8.7	2.5	3.5	4.5
C20	41	2.1	3.6	7.1	0.5	2	2.5
C21	34	2.8	3.3	6.1	0.5	2	2.4
C22	28	2.5	3.9	7.8	0.5	2.2	2.9
C23	28	1.4	2.9	6.2	0.5	1.5	1.8
C24	28	3.6	4.5	8.7	1.8	3.2	3.9
C25	34	1.1	3.1	7.1	0.5	1	1.3
C26	34	0.5	1.4	2.1	0.5	1.3	1.6
C27	34	0.5	0.5	1.1	0.5	0.5	0.6
C28	28	4.6	5.9	11.2	2.2	4	5.2
C29	43	5.8	6.4	14.2	4.6	6.6	10.8
C30	43	4.2	5.7	12.8	2.4	4.6	6.1
C31	43	2.3	3.5	6.9	1.5	3.4	3.8
C32	34	2.8	5.6	9.3	0.5	1	1.3
C33	29	2.3	5.4	11.6	0.5	2.5	2.9
C34	29	4.3	6.5	14.9	2	4.7	5.2
C35	29	7	9.1	21.2	5.1	7.6	9.4
C36	22	1.5	3.1	8.1	0.5	1	1.2
C37	-	-	-	-	-	-	-
C38	22	1.3	2.9	7.5	0.5	1.2	1.6
C39	29	2.9	4.3	8.3	1.3	2.7	3.1
C40	26	5	7.2	16.1	2.1	3.8	4.3
C41	26	4.6	6.4	15.6	2.3	4.4	5.9
C42	22	4	5.1	11.6	2	3.6	4.9
C43	26	2.6	4.2	8.6	0.5	1.7	2.2
C44	26	3.7	5.9	10.1	2	3.5	4.9
C45	22	2.3	3.5	8.5	0.5	1.4	2.1

Table 11
Natural carbonation depths in sheltered and unsheltered conditions in Fredericton.

Concrete	Fredericton (Canada)			
	Curing time	Sheltered		
		X _{C1year} [mm]	X _{C2years} [mm]	X _{C5years} [mm]
[-]	[days]			
C1	141	4.3	5.5	–
C2	141	2.5	3.1	–
C3	141	1.4	2.1	–
C4	169	2.5	3.6	–
C5	141	0.7	1.0	–
C6	127	0.2	0.6	–
C7	169	0.6	1.0	–
C8	169	0.3	0.6	–
C9	127	6.7	10.1	–
C10	127	4.3	5.4	–
C11	127	3.6	5.2	–
C12	22	1.8	2.5	–
C13	40	1.4	2.3	–
C14	106	2.2	2.7	–
C15	106	0.7	1.0	–
C16	106	1.5	2.0	–
C17	40	4.5	6.9	–
C18	35	5.9	7.7	–
C19	40	3.1	4.5	–
C20	40	1.6	3.1	–
C21	22	0.9	1.1	–
C22	19	1.0	1.9	–
C23	19	1.4	2.2	–
C24	19	3.3	4.1	–
C25	31	1.8	2.2	–
C26	31	1.6	2.2	–
C27	31	1.3	1.6	–
C28	19	3.8	4.6	–
C29	35	5.6	8.7	–
C30	35	3.8	4.6	–
C31	35	2.7	3.2	–
C32	22	1.1	2.3	–
C33	34	0.7	1.5	–
C34	34	3.1	4.2	–
C35	34	5.1	8.0	–
C36	55	2.0	2.7	–
C37	55	0.1	0.6	–
C38	55	2.7	4.8	–
C39	34	1.5	2.2	–
C40	34	2.7	3.2	–
C41	34	2.4	4.9	–
C42	54	2.7	5.0	–
C43	34	0.3	0.3	–
C44	34	1.7	2.0	–
C45	54	1.9	2.6	–

crete depth will be dried rapidly and the hydration will stop there. Once rain water has penetrated into this region, a hydration recovery can occur within this depth that modifies its resistance to carbonation. This may be the reason why the trend for Lyon, with curing of 1 day, is different from that for other locations as the trend line there does not pass by 0.

It is observed that the impact of rain is more important in Changsha and Lyon than in Chennai and Austin. This result is consistent with the comparison result between countries in unsheltered condition mentioned above as the number of rainy days is higher in Changsha and Lyon than in Chennai and Austin.

3.3. Impacts of supplementary cementing materials

The effect of supplementary cementing materials at various replacement levels is presented for Austin in Figs. 22 and 23.

It can be seen in Figs. 22 and 23 that replacing portland cement with alternative cementitious materials at various replacement

levels reduces the resistance to natural carbonation; where the higher the replacement level, the higher carbonation depth is observed. At the same substitution rate, the impact of additions is stronger for lower carbonation speed of the reference mix with 100% OPC (lower water to binders ratio, unsheltered condition instead of sheltered one).

The impact of fly ash and pozzolan is comparable and shown to result in increased carbonation rates than only using slag. The impact of limestone is also important but more data is needed for comparison with fly ash and pozzolan.

4. Conclusions

From the results presented herein, the following conclusions can be made following five year of natural carbonation is various geographic locations around the world:

- Natural carbonation in sheltered condition is similar between Lyon, Chennai, Austin and Changsha despite the climatic condition is noticeably different.
- The impact of curing on natural carbonation is vast for young concrete (i.e. 1 day) and is negligible for curing duration over 14 days, regardless of cement type. The carbonation depth remains proportional between 1 and 28 days curing up to five years. The proportionality coefficient is almost the same for different cement types.
- For concrete exposed after 1 day curing, hydration seems to be stopped quickly within a concrete depth of at least 20 mm once exposed, regardless of cement type.
- The impact of precipitation on natural carbonation is strong but the degree of impact depends on the climate. The number of rainy days per year seems to be a relevant indicator, not the cumulative precipitation as the concrete pores are blocked in saturated (or near-saturated) concrete. The higher the number of rainy days, the lower the carbonation rate. For advanced curing ages, carbonation rate is shown to almost be proportional between sheltered and unsheltered condition as the linear trend line passes thru 0 while it is not the case for very early age (i.e. 1 day). This may be explained by the fact that rain water can result in a recovery of hydration within concrete depth where hydration has been stopped in being exposed at early age.
- The impact of fly ash and pozzolan is comparable and much greater than that of slag. The impact of limestone is also important but more data is needed for comparison with fly ash and pozzolan.

Declaration of Competing Interest

We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

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Appendix

Details on the concrete mix-designs and natural carbonation results are given in the following Tables.

Table 12
Average, minimum and maximum values of temperature and relative humidity for each location.

Location	Year	T average [°C]	T min [°C]	T max [°C]	RH average [%]	RH min [%]	RH max [%]
Lyon (France)	2011	13,6	−3,0	29,8	67,6	32,0	96,0
	2012	12,9	−7,6	29,7	67,8	33,0	99,0
	2013	12,2	−2,9	30,1	70,1	31,0	98,0
	2014	13,1	−2,1	27,4	69,8	33,0	100,0
	2015	13,3	−1,3	30,9	66,6	29,0	99,0
	2016	12,8	−0,4	27,3	71,5	34,0	99,0
Chennai (India)	2011	28,3	22,7	33,8	73,7	44,0	96,0
	2012	28,8	22,3	35,6	69,2	42,0	95,0
	2013	28,3	23,4	34,4	70,3	41,0	92,0
	2014	28,6	23,2	33,9	70,0	42,0	98,0
	2015	28,9	23,3	34,6	73,1	48,0	98,0
	2016	29,1	22,2	34,6	69,1	48,0	96,0
Austin (US)	2011	20,9	−5,5	34,7	58,4	24,0	99,0
	2012	20,8	0,6	32,0	65,8	28,0	96,0
	2013	19,8	−1,5	32,6	64,7	30,0	96,0
	2014	19,4	−1,9	31,4	66,1	28,0	99,0
	2015	19,7	−1,3	31,6	71,4	33,0	100,0
	2016	20,7	0,0	31,4	70,1	31,0	96,0
Fredericton (Canada)	2011	6,7	−19,6	23,8	75,1	34,0	96,0
	2012	7,1	−17,2	23,9	73,1	30,0	100,0
	2013	6,2	−21,1	26,2	73,2	34,0	98,0
	2014	6,0	−23,8	25,2	74,3	26,0	99,0
	2015	5,7	−20,5	24,8	71,2	33,0	99,0
	2016	6,7	−19,1	24,2	72,1	30,0	97,0
Changsha (China)	2011	18,0	−0,8	34,8	70,3	31,0	95,0
	2012	17,7	−0,6	33,4	75,0	28,0	97,0
	2013	19,3	−2,1	36,2	65,3	28,0	96,0
	2014	18,7	−1,2	33,7	67,3	20,0	98,0
	2015	17,5	−0,5	32,0	81,4	39,0	99,0
	2016	17,7	−1,6	32,4	81,3	33,0	100,0

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