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Environmental impact of lime in earthworks:

European regulations - Studies of impact of lime-treated structures on the environment

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I. Lime and REACh

I.1 Principle of REACh

The REACh (Registration, Evaluation, and Authorisation of Chemicals) regulation, which came into force in 2007, has simplified the way information about chemicals sold in the European Union is obtained.[1]

The aim of this regulation is to protect human health and the environment from possible hazards when dealing with chemicals, while still promoting innovation within the chemical industry.

The REACh regulation puts the responsibility for evaluating and managing the risks associated to the use of a chemical directly on the industrials. The companies have to provide their users with appropriate safety information and data to be compliant.

The European CHemicals Agency (ECHA) is responsible for the registration, the evaluation and the recording of the submitted chemical substances in a public database.[2]

I.2 Quicklime in the REACh regulation

As quicklime (calcium oxide) is not directly extracted from a quarry but is the result of a manufacturing process, it falls under the scope of the REACh regulation. The data available in the REACh submission file for quicklime sets the PNEC (*Predicted No Effect Concentration*) at 817.4 mg/L in soil and groundwater.

[1] REACh website : https://ec.europa.eu/environment/chemicals/reach/reach_en.htm
[2] ECHA website : https://echa.europa.eu/fr/regulations/reach/legislation

Notion tackled in the REACh program: *Technosphere zone*

The use of lime in earthworks involves usually the treatment of a small portion of the natural surrounding soil (buffer area). The process results in a significant pH increase and modifies locally the soil composition.

The combined treated and buffer areas of the soil are described as anthroposphere, also called technosphere. This concept denotes the physical portion of the environment made or modified by works of any kind of human origin. In this case, the technosphere is an earthen structure or earthwork improved by the use of quicklime. The technosphere itself is out of scope of the REACh regulation, which targets its direct environment.

II. Environmental impact of lime: Studies on the migration of the ions and impact on the direct environment of a work

The improvement in and the stabilization of soils with lime involves the preparation of an intimate soil/lime mixture, which process greatly increases the basicity of the soil.

The high pH thus achieved is maintained for a very long period of time, which can vary from several years to several decades. Certain legitimate questions, ensuing from the presence of calcium and hydroxide ions in a large body of treated soil, may consequently be posed with regard to the possible diffusion towards the direct environment of the work concerned but also towards the aquifers and ground waters possibly present underneath the work.

From the perspective of responding to these questions, a series of tests and analysis have been carried out, from laboratory experimentation up to full-scale implementation, the results of which are presented below. Some other results, coming from a literature study, are also illustrated.

II.1. Laboratory leaching tests

This study was carried out in the Lhoist R&D laboratories, where a leaching column was installed in 2002. This appliance makes it possible to measure the composition and the pH of a solution leached from a sample of treated soil; it is based on the German standard DIN 18130-1, relating to the test of permeability of a soil.

The experimental arrangement is represented in Figure 1. In a triaxial cell filled with water, the cylindrical sample of treated soil is confined in a rubber membrane, and a porous disc is placed in contact with the upper and lower surface of the sample. In order to accelerate the leaching process, a flow of pressurized water (0.1 MPa) is circulated from the bottom towards the top of the sample. The rubber membrane is maintained with a higher confinement pressure than the leaching pressure (0.12 MPa), so as to prevent circulation of water between the membrane and the sample.

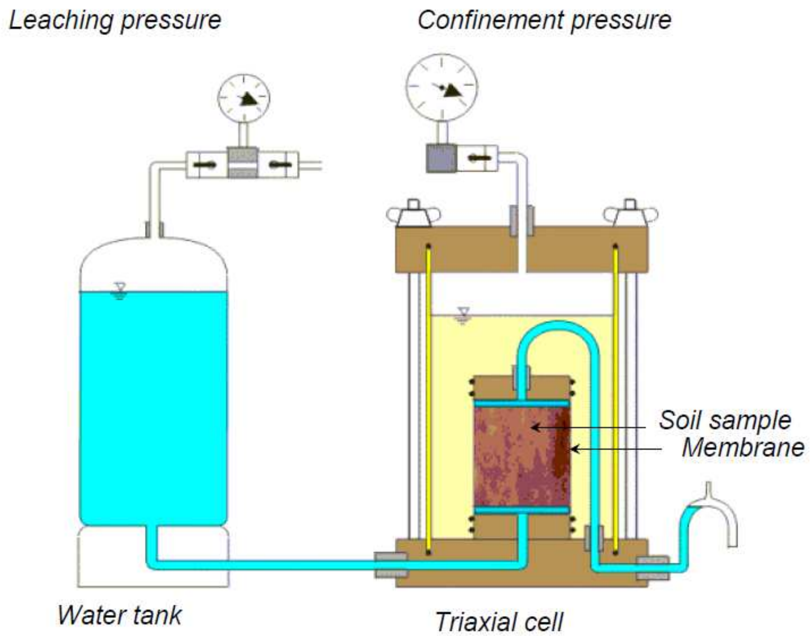

Figure 1. Experimental assembly of the leaching cell.

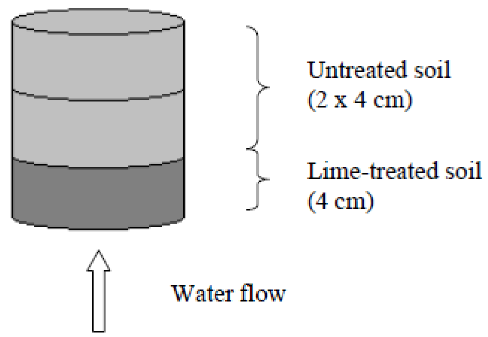

Figure 2. Arrangement of the soil sample subjected to the leaching test.

The equipment is intended to receive cylindrical test specimens with a height of 12 cm and a diameter of 10 cm, prepared in Proctor moulds. The lower layer of the sample (thickness = 4 cm) is produced from a soil treated with lime, while the two intermediate and upper layers consist of an untreated soil (Figure 2). The test specimen is positioned in the appliance so that the stream of water first passes through the treated layer and subsequently passes through the untreated soil.

The soil examined is a silty soil from the Picardy region (France). Two dosages of lime were studied: 1.5% and 3%. The water percolation tests were carried out for approximately 2 months, the change in the pH and in the concentration of calcium ions being measured in the leached solution. On conclusion of each test, the test specimen was recovered and cut into 6 slices, the Ca^{2+} content and the pH of which form the subject of measurements.

Analysis of the leached water

The change in the pH of the leached water is illustrated in Figure 3, respectively in the case of the test specimens containing a layer of treated soil containing 1.5 % and 3 % of lime. The initial pH of the distilled water is neutral (pH = 7) and increases by 1 to 2 units (up to a value equal to 9 in the case of the treatment with 3 % of lime). However, this increase remains low, in comparison with the high pH values found in a treated soil (of the order of 11-12, depending on the treatment conditions and on the age of the work). A slight decrease in the basicity of the solution is furthermore observed.

In order to establish a comparison, allowed pH values of industrial effluents neutralization, are in the range of 5.5 to 8.5, and when neutralization is made with lime, 5.5 to 9.5, which would integrate this case. For further information, see official French and European specifications.[3,4]

The amount of calcium which is passed into the percolation water is low. It is evaluated at less than 3 mg per week. Furthermore, this leached amount has a tendency to decrease with time, as is suggested by the graph of Figure 4, in the case of the sample containing the soil treated with 3 % of lime.

Once again, it is possible to have a comparison of the maximum calcium oxide amount leached with the REACh threshold, taking therefore the hypothesis that all the leached calcium is coming from lime. During the first week of the test, the level of leached calcium ions is the highest and corresponds to 2.6 mg/day. The calculation of equivalent concentration in water, taking the test parameters into account and the Darcy's law of hydraulic conductivity :

$$Q = k.S.\Delta dH/L$$

Where :

Q = water flow (m^3/s) ;
k = hydraulic conductivity (m/s) : 10^{-9} m/s is a relevant hypothesis giving a small leached volume

[3] https://aida.ineris.fr/consultation_document/8589 (France, 1996)
[4] https://circabc.europa.eu/sd/a/97d7bd92-e92e-4b42-a2b3-6a1d8d00d81d/National%20Technical%20Regulation%20%20on%20Industrial%20Wastewater.pdf (EU, 2009)

S = specimen section ($7.85.10^{-3}$ m²) ;
L = specimen height : 0.12 m
ΔH = piezometric height, or equivalent water column : 10 m (1 bar)

The calculation gives Q = $6.9.10^{-10}$ m³/s, or $5.6.10^{-5}$ m³/day (56 ml/day). The calcium concentration is, at this time, equal to 2.62/0.056 = 47 mg/liter, which also corresponds to a maximum CaO equivalent of 66 mg/L.

Taking into account that the PNEC (*Predicted No Effect Concentration*) in soil and groundwater is 817.4 mg/L according REACh, the leached lime will stay well below this value.

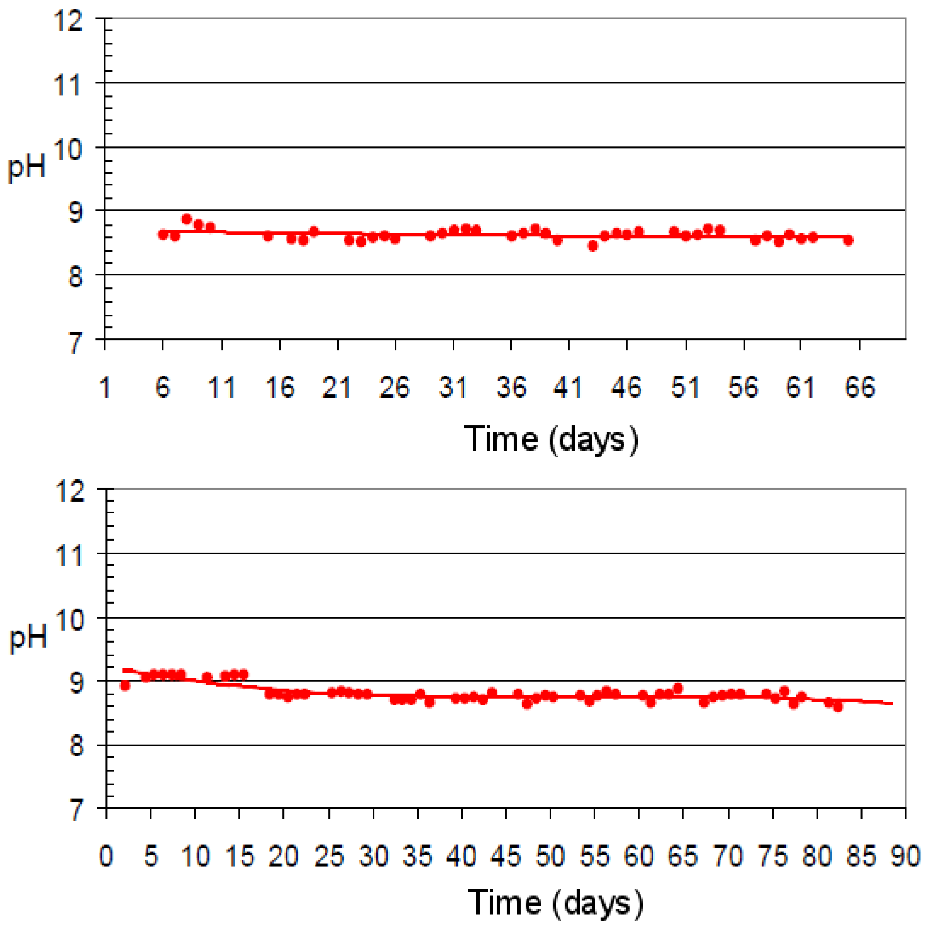

Figure 3. pH values of the leaching waters from the samples containing the silty soil treated with 1% (upper graph) and 3% (lower graph) of quicklime.

Conclusion of this series of tests: the pH and the amount of calcium (calcium oxide) leached by a solution of water injected at a relatively high pressure are both maintained at acceptable levels, even in the case of a treatment with 3% of lime.

Analysis of the recovered soil test specimens

On conclusion of leaching tests, the soil test specimens were recovered and cut into 6 slices, each having formed the subject of a measurement of pH and of calcium content. The change in these two parameters through the structure of the test specimen contained in the soil treated with 3% of lime is represented in Figure 5.

After 2 months of accelerated percolation, the pH of the treated layers remains at 10.8, while the untreated fractions exhibit a pH of 7.2 to 7.5, which is recorded as being the original pH of the natural soil. The content of calcium ions is much bigger in the treated section and decreases in the untreated sections as a function of the distance of separation from the soil treated with lime (Figure 5).

These observations tend to demonstrate the “buffering” nature of the layers of natural soil underlying a treated soil, the ions being adsorbed by a thin layer of natural soil, even under a percolation pressure of 0.1 MPa.

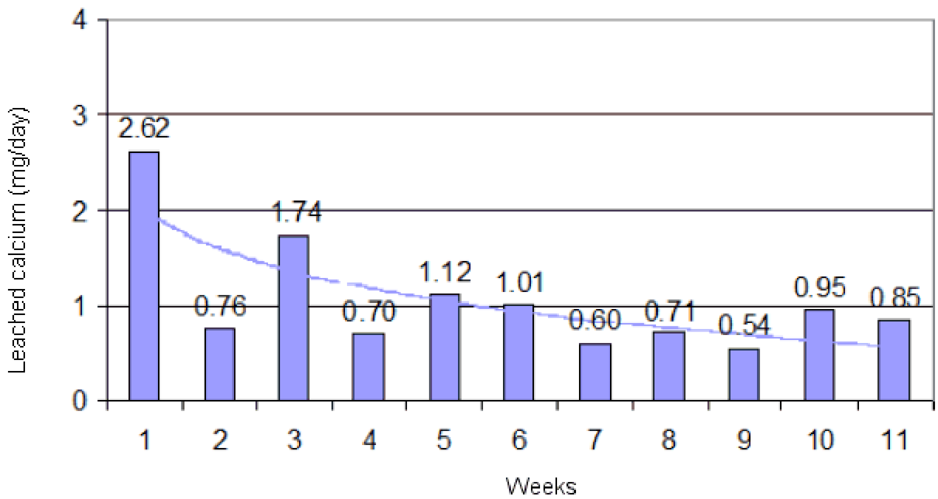

Figure 4: Amount of leached calcium as a function of the time, from the sample containing the silty soil treated with 3% of lime.

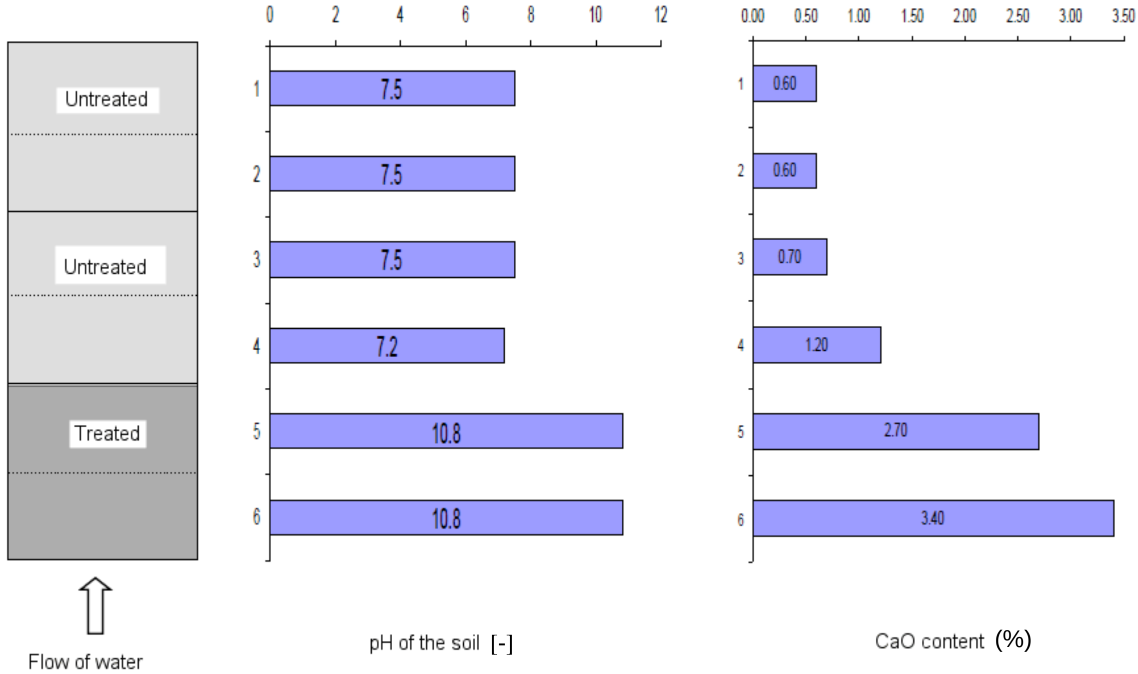

Figure 5: pH values of the different layers and total CaO content of the sample of silty soil treated with 3% of lime and untreated "buffer" soil, on conclusion of the percolation test.

II.2 Experimental trench

This trench was built with the initial aim of measuring mechanical performance qualities of excavated and recycled materials treated with lime. It was prepared and constructed in collaboration with the CER (Research and Experimentation Centre, Rouen, France), on a materials recycling platform.

Samples were taken from and in the direct vicinity of this trench, in order to evaluate the diffusion of lime from the treated zones towards the surrounding soil body.

The experimental trench was built in the following way (see Figure 6) :

- A sublayer made of untreated silt ① (35 cm)
- A layer of treated mix (silty clay excavated material treated in a mixing plant with 2% of quicklime) ② (45 cm)
- A covering layer made of untreated gravel ③(17 cm)
- Everything inserted into a sandy soil body ④

The trench, in its soil body, was soaked for 2 months. Once this period of time had passed, a transverse trench was hollowed out and samples were taken manually from the section. The sampling scheme is represented in Figure 7.

The following were withdrawn:

- samples in the layer of treated soil: Va, Vb, Vc
- 2 samples in the silt sublayer: La, Lb
- 6 sand samples: Sa, Sb, Sc, Sd, Se, Sf
- 2 reference samples: Sref (sand) and Lref (silt)

Each sample was subjected to a measurement of total calcium by atomic absorption. After stirring a suspension of 100 g of dry soil in 1 litre of water for 24 h, pH and conductivity measurements were carried out on the filtrate.

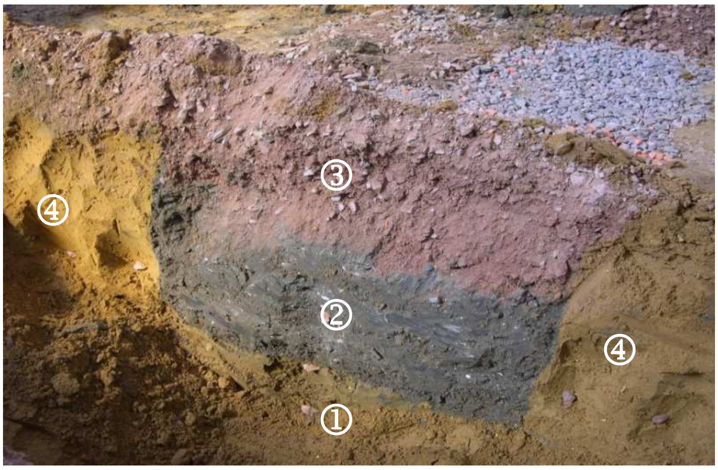

Figure 6: The experimental trench profile. The different materials of which it is composed are identified in the text.

The conductivity, which is a direct function of the ion concentration, makes it possible to qualitatively validate the total calcium measurements. The results of these measurements are taken up in , in which the contents of calcium have been converted into CaO equivalent.

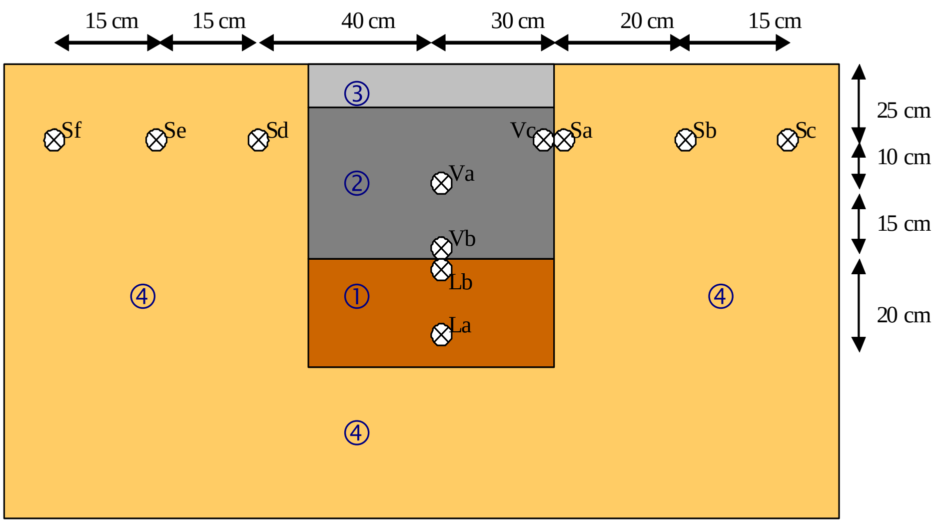

Figure 7: Scheme for withdrawing the samples from the experimental trench.

Material reference	Type of material	pH	Total Ca (%,CaO expressed)	Conductivity (µS/cm)
Va	Lime-treated material	12.2	12.43	1362
Vb		12.2	12.92	1465
Vc		12.2	16.75	1369
Sa	Sand body	9.7	0.03	80
Sb		8.4	0.03	64.7
Sc		8.6	0.04	175.4
Sd		8.7	0.03	51.3
Se		8.5	0.03	8.53
Sf		8.7	0.03	8.68
Sref		8.5	0.04	8.46
La	Silty soil sublayer	7.8	0.04	86.1
Lb		8.0	0.24	90.2
Lref		8.3	0.03	26.3

Table 1: Results of the pH, total Ca (CaO expressed) and conductivity measurements at the different sampling locations identified in Figure 7.

The pH of the treated material remains in all cases greater than 12, even after immersion for 2 months. The pH of the sand sample Sa, in direct contact with the treated material, is scarcely higher than that of the other sands (9.7 in comparison with 8.5); the same observation may be made for the layer of silt underlying the treated material.

It may be concluded therefrom that the rise in pH is only slightly noticeable and is restricted to the first centimetres of soil surrounding the portion treated with lime.

Nevertheless, the measurements of total calcium and of conductivity generally show us a high concentration of calcium ions in the lime-treated layer and a very limited concentration in the neighbouring materials. The migration of ions, with calcium and hydroxide ions, into the neighbouring materials is negligible and remains restricted to the first centimetres (Sa and Lb), despite a prolonged saturation of the site.

II.3 Tests carried out in a real work situation

Embankment of motorway in Czech Republic

The following study, carried out on a real work, was performed in December 2001 by the company Stavebni geologie – Geotechnika. The site concerned is the construction jobsite of the motorways R3509 Slavonin - Praslavice and E 0803 Nova Ves – Doksany, in the Czech Republic.

This study, in addition to the advantage of giving results for a real construction site situation, also exhibits the following benefits:

- the works are located in a nature reserve, for which the environmental impact of a treatment with lime is of obvious importance;
- the measurements on site could be correlated with laboratory and percolation tests.

The measurements carried out on the site show that, in the vicinity of the foot of the embankment, the pH of the water in nine boreholes changed, passing from a neutral value (pH = 7) to a value of 11.5 during the works for treatment with lime (from 25 July to 11 August 2000).

The values return to the initial neutrality during the following winter and subsequently remained constant. The increase in pH observed in the excavation period can be attributed to the locations where the samples were taken, open core borings exposed to runoff water and to lime dust.

The tests carried out in the laboratory made it possible to measure the change in the pH and in the concentration of soluble calcium on the soils of the work treated with 2.5% of quicklime. The results, presented in Figure 8, show relatively constant pH values and a decrease in the soluble calcium, doubtless due to the fixing thereof by pozzolanic reaction.

Similar determinations were carried out on samples of water present in soils this time withdrawn at different distances from the foot of the embankment, in the direct surroundings of the motorway R 3509 (March and June 2002). The results are illustrated in Table 2 and in Figure 9.

The values measured are fairly close to the values present in the "natural" state and do not appear to be affected by leaching of the lime from the embankment of treated soil.

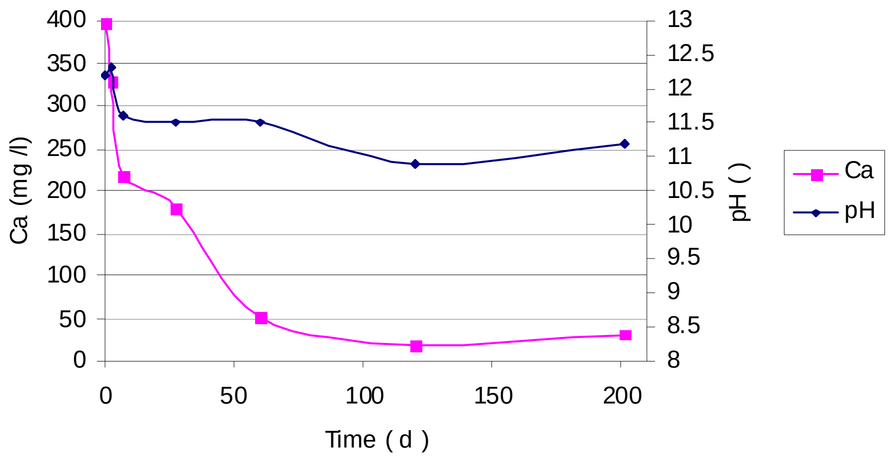

Figure 8: Change in the pH and in the calcium concentration of the materials treated with 2.5% of lime on the Czech Republic site.

Distance from the toe of the embankment (m)	Position : pK 150.149			
	pH (-)		Calcium concentration (mg/dm³)	
	March	June	March	June
10	8.05	7.84	7.6	15.4
20	8.02	7.66	10.5	14.9
30	8.5	8.17	8.7	12.4

Table 2: pH and calcium concentration in the soil bodies surrounding the zone treated with 2.5% of lime.

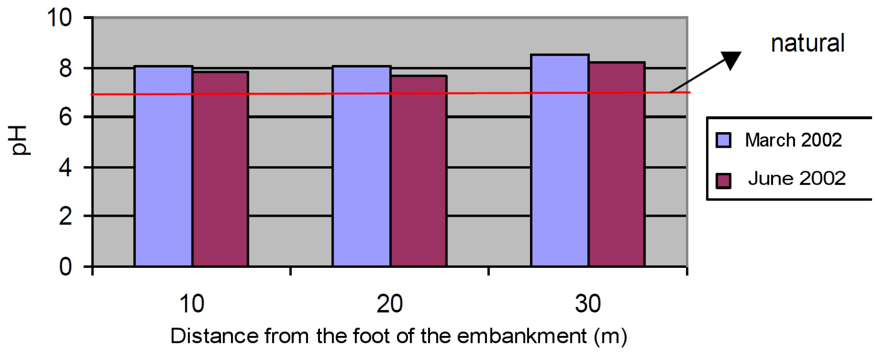

Figure 9: pH values measured in the water on conclusion of core borings at several distances from the soil body treated with 2.5% of lime.

The percolation tests were carried out simultaneously on samples of untreated soil, samples treated with lime and a sample consisting of a sublayer of untreated material covered with the soil treated with lime. All the soil samples were withdrawn on the site of the motorway R3509. The soil, which is a silty clay, was compacted with Proctor energy at its natural humidity. The dosages with lime of the treated samples are of the order of 1% or 3%. The percolation tests were carried out in a triaxial cell, with a cell pressure of 16.6 kPa and a confinement pressure of 4.6 kPa. This test required a week. The results are presented in Table 3.

Test results	Untreated	Without sublayer		With sublayer	
Lime addition (%)	0% CaO	1% CaO	3% CaO	1% CaO	3% CaO
pH	7.6	11.97	12.28	8.27	9.14
Ca (mg/l)	202	278	472	98.4	103

Table 3: Results of the percolation tests carried out on the materials from the Czech Republic site.

The sublayer of natural soil has a reducing effect both on the pH and the concentration of calcium ions of the leached solution. The pH is consequently greater by one unit with respect to neutrality (pH = 7), which tends to demonstrate that the untreated materials on which a large body of soil treated with lime rests, acts as a buffer, both for the basicity of the infiltrating water but also for the calcium ions dissolved after the treatment.

Note that the highest calcium concentration of the leachate corresponds to 661 mg/l CaO, which is again below the PNEC (*Predicted No Effect Concentration,* 817.4 mg/l).

Case of lime-treated water basin

In the context of an experimental pilot levee in lime-treated soil, a water basin was built in July 2015 in the Gard department (France). The material used for this operation was a silty soil excavated on the site and treated with 2 % quicklime. This basin of about 200 m³ capacity is also surrounded by an ambnakment and a platform built using the same lime-treated soil.

After the construction, the basin was filled with rain water and pumped water. pH measurements of water were performed 3 months after construction, from several samplings in the basin and from the water taken in the river nearby. The values are reported in Table 4, and also compared with rainwater.

Origin of water	Sample	pH value
Lime-treated soil basin	A	7.2
	B	7.3
	C	7.2
River	D	7.3
Rain water	E	6.8

Table 4. pH values of water contained in lime-treated basin, comparison with river water and rainwater from the same area.

The pH value of the water contained in the basin from 3 months is absolutely identical to the river, meaning that no impact of the constitutive lime-treated soil can be observed.

II.4. Additional literature study : case of lime columns

Several publications can be found on the literature, dealing with lime columns or lime piles, and having as first purpose to investigate the extend of improvement around these areas concentrated in lime. Some of them also report information about environmental impact in terms of lime diffusion into surrounding soils.

Rao and Mathew[5,6] investigated the permeability and the reaction products in lime-stabilized marine clay, using the lime column technique. They used the pH evolution as an indicator of lime migration. The pH was monitored for 60 days at distances up to 25 cm from the column face. The pH value, which was 7.5 after 2 days, reached 8.9 after 60 days.

These results can be compared with field tests made by Barker[7] on lime treated columns. He noticed that the pH increase near the lime piles is limited to a distance of 1.5 cm after 13 weeks. No more evolution was then observed after 13 further weeks. Laboratory tests from Rogers[8,9] confirmed that hydroxyl ion migration is limited between 2 and 3 cm far from the lime source. Barker's study[10] on 4

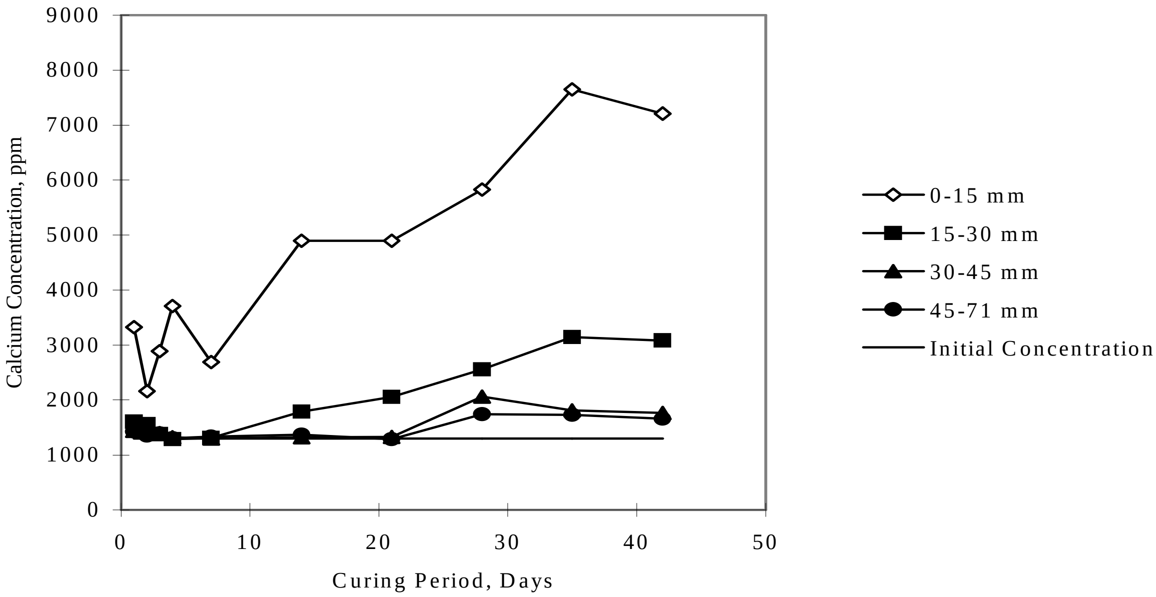

Figure 10. Relationship between calcium concentration in clay surronding the lime pile, and curing period.

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[5] Rao, Mathew , Effect of lime on cation exchange capacity of marine clay, Journal of Geotechnical and Geoenvironmental Engineering, 123, 2, 1997

[6] Rao, Mathew, Permeability studies in marine clays stabilized with lime columns, Int. Offshore and Polar Engineering, Hamilton, 1990

[7] Barker J.E., Rogers C.D.F., Boardman D.I., Physico-chemicals changes in clay soils surrounding lime piles, International Conference on Underground Infrastructure research: Municipal, Industrial and Environmental Applications, University of Waterloo, Ontario, Canada, 2001

[8] Rogers C.D.F., Glendinning S., The role of lime migration in lime pile stabilization of slopes, Quarterly Journal of Engineering Geology, 29, 1996

[9] Rogers C.D.F., Glendinning S., Troughton V.M., The use of additives to enhance the performance of lime piles, Proceedings of the 4th International Conference on Ground Improvement Geosystems, Helsinki, 2000

[10] Barker J.E., Ion Migration associated with lime piles, Ph. D. Thesis, University of Birmingham, Department of Civil Engineering, January 2002

more clayey soils with this technique led to similar conclusions: pH only increased from 5.5 to 6 after 60 days at a distance of 3 cm. The calcium ions seemed to be more mobile (Figure 10), but once again the diffusion perimeter seems to be limited to a few centimeters.

The authors provides the following explanation for lime diffusion in this type of application.[8] The actual mechanism is probably a combination of ion diffusion and mass transport depending on water flow conditions. With respect to diffusion, a possible explanation for the short distances observed is that it is controlled by a process of ion exchange. The potential for ion exchange must depend somewhat on mineralogy. The self-limiting nature of the reactions, in which stabilization of the soil adjacent to a pile inhibits further migration, must also be considered in the migration process.

II.5. Restoring the vegetation on the supporting soil mass treated with lime

As part of a research project on the long term effects of stabilizing a loam with lime, the Centre de Recherches routières [Road Research Centre] (CRR, Belgium) conducted a study, which was the extension of a test bed installed in the 1970s. At the same time as the mechanical and chemical analyses of the supporting mass, a botanic examination was carried out in 2003 on this 30 year old stabilised loam in order to study the long term impact of the treatment on vegetation, by comparing the vegetation in this treated area with that in an adjacent sample area.

The loam has a plasticity index of 8 and 95% of particles are < 74 µm.

The test bed was made in August 1972, on a section of soil at the Sterrebeek experiment site, which belongs to the CRR. A loam was thereby mixed with 7% quicklime to a depth of 30 cm over an area of 225 m² (15x15 m). This treated area was left in this condition for more than 30 years without bearing any traffic loads or without any additional treatment or changes. Only one annual trim is mentioned. It is estimated that the first 10 cm of the treated layer was recolonized by the vegetation. Exhaustive phytosociological readings have been taken by the systematic botany and phytosociology laboratory at the Université Libre de Bruxelles. The procedure used is the traditional Braun-Blanquet method; on a standard 4 m² area and in physiognomically homogeneous vegetation communities. The readings are restricted to a standard area which means that all the readings can be compared for their specific richness, i.e. the number of species they contain. In total, four readings were taken in the test area and an identical number in the lime-treated area.

Present species

The number of species recorded in the two areas did not differ significantly: 28 for the test area compared with 25 for the treated area. In addition, it was noted that many species were common to the two areas. Among the species noted, the most calciphilous were cut-leaved cranesbill *(Geranium dissectum),* common tansy *(Tanacetum vulgare),* sprawling speedwell *(Veronica prostrata),* grass vetchling *(Lathyrus nissolia),* wild basil *(Calamintha clinopodium),* alfalfa *(Medicago sativa)* and coltsfoot *(Tussilago farfara).* The calciphilous species were found just as much in the test area as the lime-treated area. When the readings were taken, the treated area had both a smaller amount of vegetation cover (on average 55% of the soil covered in vegetation compared with 85% in the test area) and less developed vegetation. The vegetation reached an average height of 80 cm in the test area and 60 cm in the lime-treated area. However, these disparities seemed to recede as the seasons progressed.

Values indicating the different species present In order to correctly describe the different units of vegetation identified and their relationship with environmental variables, the acid-alkaline coefficients and the availability of nitrogen were calculated based on the Ellenberg indicator values.

The most relevant parameter in the study seemed, in fact, to be that of acidity, whereas the nitrogen content in the soil generally reflects the overall fertility of the soil expressed by the production of vegetation.

Each species was allocated an Ellenberg indicator value in order to calculate the coefficients for a reading. The total of the values from the species is then divided by the number of species in the reading.

The coefficients thereby calculated result from the acidity (R) and the nitrogen content (N) indicator values. The scale of values ranges respectively from strong acidity (R1) to the basicity and high calcium content (R9) and from an extremely low level of nitrogen (N1) to excessive nitrogen (N9). Table 17 shows the indicator values for the areas examined.

The two areas do not differ greatly from each other. The average number of species per reading is very close and the R values are also similar. The N values are slightly lower in the treated area, which is corroborated by a lower level of biomass.

Test area				Lime-treated area			
Reading no.	Number of species	R values	N values	Reading no.	Number of species	R values	N values
1	17	6.8	4.9	5	9	6.3	4.7
2	18	6.3	5.4	6	15	6.5	4.2
3	12	6.6	6.0	7	14	6.3	5.3
4	17	6.5	5.6	8	17	6.1	4.8
Average	16	6.6	5.5		14	6.3	4.8

Table 4: Results of the R (acidity) and N (nitrogen) indicator values obtained after 30 years in the CRR bed. The lime-treated area corresponds to 30 cm of A1 loam treated with 7% lime.

<u>Conclusion from the vegetation study</u>

Even if, physiognomically, the test area and the limed area appear quite different at the time of the readings, these differences are not marked in the results. The total number of species observed in the two environments and the average number of species per reading, were not significantly different. The proportion of calciphilous species is similar in the two areas, and the R values cannot be used to distinguish them. Only the N value is slightly lower in the lime-treated area, which amounts to saying that the overall fertility of the soil is slightly less in the treated area.

II.5 Conclusions

The multiscale study, carried out through laboratory leaching tests, samples withdrawn from a saturated experimental trench and on a full-size excavation site, has made it possible to conclude that the environmental impact of a treatment with lime, via the migration/diffusion of the calcium and hydroxide ions towards the neighbouring materials and aquifers, is very limited.

The migration of these ions and the impact on the pH of the materials is limited to a zone of a few centimetres (5 to 6 centimetres for the pH, 20 centimetres at most as regards the more mobile calcium ions).

The leaching tests show a very limited effect on the percolated water. The observations tend to demonstrate the “buffering” nature of a soil, which acts as neutralizing barrier with regard to the diffusion from the structures treated with lime.

Furthermore, it has been found that the pH and the concentration of calcium ions of the treated layers was stable over time, which is evidence for the progression of the pozzolanic reactions initiated by the lime, when stabilization of the soil is the targeted objective.