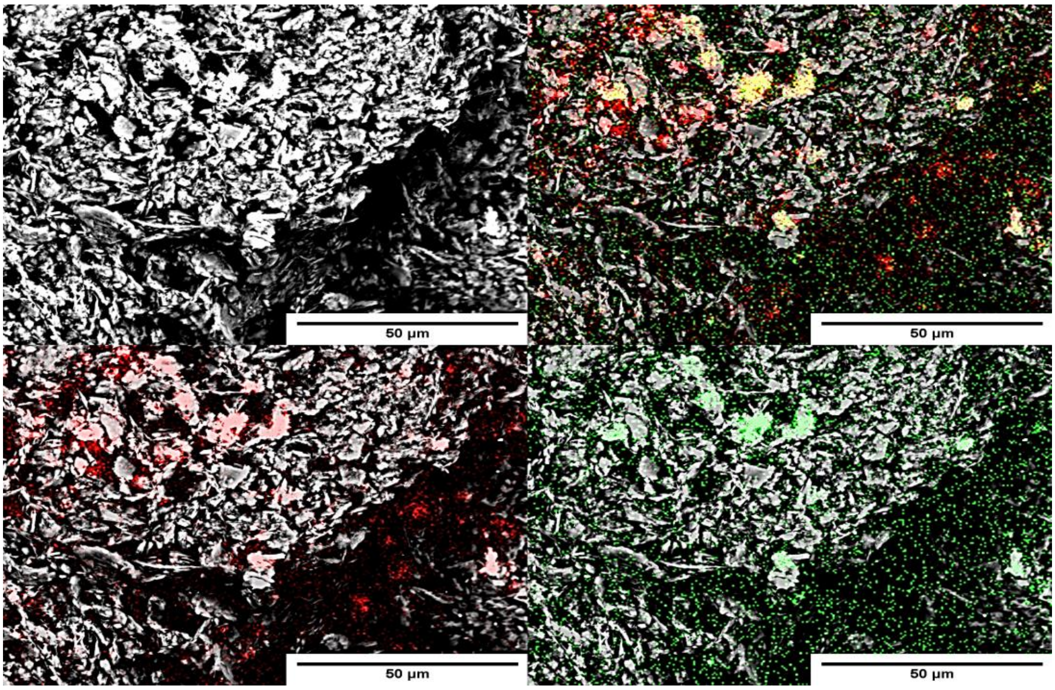

Effects of heavy metal contamination on pozzolanic reaction in clay and challenges with sustainable remediation

Master's Thesis in the Master's Programme Infrastructure and Environmental Engineering

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CHALMERS UNIVERSITY OF TECHNOLOGY
Gothenburg, Sweden 2021
www.chalmers.se

MASTER’S THESIS ACEX30

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Cover:
SEM-Photo of stabilised contaminated kaolin clay, indicating that copper ions binds to the calcium ions in the binder.
Department of Architecture and Civil Engineering
Göteborg, Sweden, 2021

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ABSTRACT

Sustainability is more relevant than ever before, hence having sustainable remediation methods is essential. Today, most of the remediation is done through "dig and dump", an old and a non-sustainable method. Instead, using waste and natural materials, and utilising natural pozzolanic reactions, can minimise the remediation footprint and maximise its results.

The purpose of this study is to evaluate the influence of heavy metals on pozzolanic reactions on reconstituted kaolin and natural sensitive clay from Gothenburg, by mixing the clays with copper nitrate ($Cu(NO_3)_2$) and adding calcium hydroxide ($Ca(OH)_2$) and silicon oxide (SiO_2), acting as a binder that creates a cementitious mass. The effect of contamination and binding was investigated by performing geotechnical tests to compare the strength and stiffness of clay with different mixtures of the binders. Different curing methods were also evaluated to examine governing factors during sample preparation. Furthermore, another aspect was to investigate the challenges with implementing sustainability when remediating. This was done through interviews with Skanska and governing authorities such as the Swedish EPA. A thorough literature study was also conducted to evaluate previous studies on the subject.

Copper contamination affected both the physical and chemical properties of the clays tested. The clay mineralogy is crucial, since the behaviour of the Gothenburg clay and kaolin clay was opposite to each other. This has been proven by having higher strength and stiffness for the uncontaminated Gothenburg clay compared to the contaminated clay, while the opposite was observed for kaolin.

Finally, sustainability management is lacking in knowledge and usage of new technologies. There is a wide knowledge in the sector, but it needs to be spread and more widely available in form of better handbooks. It was also shown that more sustainable guideline values are needed, to prevent unnecessary remediation. The literature showed a great lack of usage of sustainable remediation methods, and it must be more beneficial to use sustainable methods prior to old "dig and dump" methods.

Key words: Stabilisation, kaolin clay, heavy metals, pozzolanic reaction,
sustainability management, sustainable remediation.

Tungmetallers påverkan på puzzolanska reaktioner i lera och utmaningar med hållbar sanering

Examensarbete inom masterprogrammet infrastruktur och miljöteknik

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SAMMANFATTNING

Hållbarhet är mer relevant än någonsin, att ha hållbara saneringsmetoder är även det lika viktigt. Idag sker majoriteten av sanering genom schaktsanering, en gammal och tämligen ohållbar saneringsmetod. I stället, genom att använda överskottsmaterial eller hållbara material, och utnyttja naturliga reaktioner, kan saneringsmetoder bli avsevärt bättre.

Syftet med den här studien är att undersöka tungmetallers påverkan på puzzolanska reaktioner i kaolinlera och Göteborgslera, genom blandning av kopparnitrat med kalciumhydroxid och kiseldioxid som agerar bindemedel och skapar ett cementliknande material. Detta undersöktes genom olika geotekniska tester för att jämföra hållfasthet samt styvhet hos lerorna, med olika blandningar av bindemedel. Även olika härdningsmetoder testades, för att undersöka styrande faktorer. Syftet var också att undersöka utmaningar med implementering av hållbar sanering. Detta undersöktes genom intervjuer med Skanska och styrande myndigheter så som Naturvårdsverket. Dessutom gjordes en omfattande litteraturstudie, för att undersöka tidigare studier inom området.

Att ha koppar i leran visade sig ändra så väl fysiska, som kemiska egenskaper hos leran. Typen av lera är väsentlig då det visade sig vara motsatt effekt mellan kaolinleran och Göteborgsleran. Detta bevisades genom att den rena leran hade högre hållfasthet än den förorenade leran för Göteborgsleran, medan motsatt resultat för kaolinleran.

Gällande hållbar miljöhantering visade det sig finnas kunskapsbrist och brist på användning av ny teknik gällande hållbar sanering. Det finns en bred kunskap i branschen men denna behöver spridas och vara mer åtkomlig för användare, genom att ha bättre handböcker. Det visade sig även vara brister i tillämpning av mer hållbara riktvärden, för att undvika onödig sanering. Litteraturstudien visade att det är en stor brist på användning av hållbara saneringsmetoder och att det måste vara fördelaktigt att använda dessa jämfört med gamla schaktsaneringsmetoder.

Nyckelord: Stabilisering, kaolinlera, tungmetaller, puzzolanska reaktioner, hållbarhetshantering, hållbar sanering.

Table of contents

Preface

This study was carried out during the spring of 2021 as a part of the Master's programme at Chalmers and as a part at Skanska and the Skanska 21 student programme. All laboratory work was conducted at the Geomechanics laboratory at Chalmers; therefore, I would like to give a big thanks to all involved to help and support me. Especially Georgios Birmpilis and Reza Ahmadi Naghadeh for all their help in the laboratory with testing and their advice, but also for the support with the report and being patient with all my questions. I would also like to thank Jelke Dijkstra for being positive and suportive. At Skanska, I would like to thank Linus Andersson with the help with inspiration, ideas and support with sustainability management questions. Last but not least I would like to thank Anders Karlsson for supplying laboratory equipment and material to make all the testing possible.

Finally, I want to thank Chalmers and my classmates for the joy I have had during my years at Chalmers.

Göteborg, June 2021

Folke Arvidsson

1. Background

This chapter will describe relevant background information for explaining the topic and understand why it is relevant. It will also describe the aim and objectives of the study.

1.1. Introduction

Urbanization is a commonly used term and refers to that more of the population move closer to large cities, to achieve a better way of living (UN, 2020). This often enhances the economy both for the region and the residents (UN, 2020). However, due to limited space in cities, the city needs to expand geographically which can be difficult, due to the previous land use (Boverket, 2019). In the past, industrial areas were often located on the outskirt of the city, leading to heavily contaminated land causing a need for remediation before constructing residential areas (Länsstyrelsen Stockholm, 2003). Today there are almost 85 000 contaminated sites, of which 1100 pose a great risk, indicating that contaminated sites are a significant problem (Naturvårdsverket, 2019). More sustainable remediation technologies are needed and old technologies such as "dig and dump" are soon to be outdated. This due to several reasons but mainly the fact that the soil must be dug up and requires vast space for the masses, as well as having little to no recycling (Hudson, 2010). Instead, the soil can be remediated *in-situ* leading to less energy spent on the remediation process and no space required for the soil masses. However, some *in-situ* technologies require perfect soil conditions to work, making it less versatile than the user-friendly "dig and dump" (Geosolv, 2020).

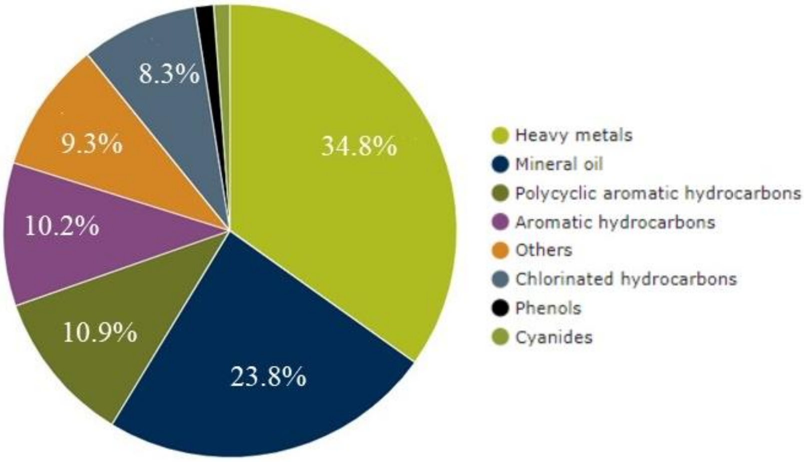


Figure 1. Proportions of different contaminants in the soil in Europe (EEA, 2011).

There are many different types of contaminants found in the soil, as shown in Figure 1, where heavy metals and mineral oil are the most commonly found in Europe (EEA, 2011). Heavy metals can often be found at locations where old industries have been and can be both toxic and vital for the human body (Naturvårdsverket, 2006). Copper is one of those heavy metals that is needed for essential enzymes to function properly but is toxic at higher concentrations. Since copper is not biodegradable it tends to adsorb in humus substances which can be transported by water or accumulated in soils through binding to particles (Naturvårdsverket 2006). The reason that copper is of particular interest is because of its great adsorption potential to clay particles. The adsorption is related to pH where a lower pH results in lower adsorption, hence clays with high pH are likely to bind strongly to copper (Atanassova & Okazaki, 1997).

There are several options for remediating heavy metals from soil, as mentioned before the soil could be remediated *in-situ*, dug up and disposed or treated *ex-situ* (Naturvårdsverket, 2009). This study will focus on the *in-situ* method of solidification/stabilisation since it is the most applicable in practice. How this remediation method affects non-contaminated soil is known, but less is known how it is affected by contaminants.

1.2. Problem definition

Having more intensified constructions on sensitive clays can be a future problem, both in terms of stability and settlements. Constructing on soil that has been remediated and that once was contaminated can cause uncertainties in how the soil behaves, in comparison to non-contaminated, non-remediated “natural” soil. Also, making sustainable remediation more comprehensive and more attainable for construction companies is vital. This by having a more straight-forward procedure in terms of regulations and laws, making it easier to implement sustainable remediation.

1.2.1. Aim and objectives

This study aims to evaluate how heavy metals influence the performance of a sustainable stabilisation/solidification method and to understand how remediation is managed today and what has to be done to implement more sustainable remediation. This is achieved by i) performing a series of comprehensive laboratory testing using both micro and macro scale laboratory techniques and ii) literature review and industry-based questionnaires with a general focus on sustainability and sustainability management.

In line with the objective, the following questions will be answered in this study:

What is the effect of heavy metals, *i.e.* copper, on the shear strength and stiffness of clay?

- Perform geotechnical tests on contaminated clay sample and compare with the reference non-contaminated.

What effect has the stabilised remediation process on the strength and compressibility of clay, and is it affected by heavy metals?

- Perform geotechnical tests on the remediated contaminated clay sample and compare with the reference non-contaminated stabilised sample and contaminated sample.

How is the situation with sustainability management in Sweden and how could it be improved?

- This is based on both literature review as well as questionnaires and interviews from construction companies and governing authorities.

What should be done to have a more easily “user-friendly” process when planning and performing remediation? Are construction companies limited by regulations and laws, making it difficult to implement more sustainable remediation more easily?

- This is based on both literature review as well as the more practical aspect and regulations when it comes to remediation and contaminated soil. Also, by a questionnaire to understand how remediation is perceived in different construction projects.

1.2.2. Limitations

The study will not discuss or compare how other remediation methods affect the clay, as well as evaluate the effect of having different types of contaminants in the soil. It will also mainly focus on the management and implementation of sustainable remediation in Sweden, not other countries.

2. Contaminated Clay and Stabilisation Remediation

In this chapter are all necessary and relevant aspects described, based on previous studies and available literature regarding the subject. The methodology and research questions are, to an extent, based on this literature review, which is divided into separate chapters to include all relevant parameters for answering the aim.

2.1. Geotechnical properties and mechanical response of contaminated clay

Clay is the most fine-grained soil material, with particles smaller than 2 µm in diameter, and consists of one or several types of clay minerals. Depending on the mineral composition, water content and void ratio (the ratio between the volume of solids and volume of voids), the clay's properties will significantly differ in terms of strength and stiffness. The most common clay minerals are kaolinite, illite and smectite, which all have different composition of minerals, such as alumina, silica, potassium and magnesium, as well as structure. This structure of the clay minerals can vary from being a simple two-layered mineral to a complex mineral with several layers. Therefore, different clays will react differently with chemicals and have different geotechnical and hydrogeological properties. Clay can be a stiff, almost rock-like, material with a strength of 200 kPa but can also be a sensitive material with almost no stiffness and low strength of 10 kPa. This variation, in combination with very low permeability, makes clay a complex soil to work with when constructing. The mechanical response for clay is anisotropic where the Poisson's ratio often is around 0.2. Due to the low permeability and relatively high water content, the pore water will dissipate due to consolidation as a result of applying pressure. How the pore water pressure changes have a great effect on the response of clay, since the effective stress is directly related to the pore water pressure. Therefore, the drainage conditions are vital for the clay's response since it controls the behaviour of the pore water pressure. How clay is affected by contaminants, especially heavy metals, will be described in the upcoming chapters. (Mitchell & Soga, 2005)

2.1.2. Heavy metals

There are numerous studies regarding heavy metals, Muththalib and Badouet (2019) as well as Karkush *et al.* (2013) are two of the studies that have investigated the effect of having heavy metals in clay. According to Di Maio (1996), having a higher concentration of alkali metals such as sodium and cadmium changes the Atterberg limits (liquid limit, LL, and plasticity index, PI). As mentioned in the introduction, soil binds strongly to metals and the adsorption depends on different parameters. Mineralogy, cation exchange capacity (CEC) which is the amount of cations the negative clay particles can bind to, and the organic matter determines how strong metal ions can bind to the soil (Martínez *et al.*, 2000; Muththalib and Badouet, 2019). Anson and Hawkings (1998) and Di Maio *et al.* (2004) have also shown that the mineralogy of the clay and the clay fraction has an important role in how the clay reacts to alkali metals in the pore fluid. According to them, having salt (NaCl) in smectite clay decreases the compressibility of the soil whereas, for kaolinite and illite, the result is the opposite. The explanation of this is partly due to the higher water content in smectite but it is mainly the CEC that influence the most, this since a mineral with lower CEC binds weaker to metals (Ridge, 1983). Further, Ridge (1983) showed that smectite has roughly ten times higher CEC than kaolinite and roughly three times higher than illite, thus explaining the lesser strong bonding for kaolinite. pH is also vital for the soils ability to bind to metals, a higher pH in the pore fluids decreases the charge of the negatively charged clay particle, resulting in a stronger binding with the positively charged metal ion (Muththalib & Badouet, 2019).

In more detail, the area around a clay particle consists of a negatively charged clay particle, a positively charged layer surrounding the particle (Stern layer) and outside that layer is a mixture of positively and negatively charged ions (the diffuse layer), both these layers are referred to as the double layer, see Figure 2 (Pouralhosseni, 2014). When a positively charged cation, a metal ion, binds to the negatively charged clay particle, the double layer is penetrated and compressed, which is why the clay should become less compressible, at least in theory. Further, depending on how strong the clay binds to the metal ion, the clay will be more or less compacted (Muththalib and Badouet, 2019).

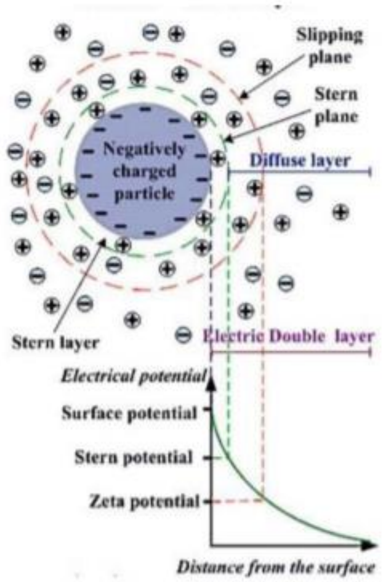


Figure 2. Different layers surrounding the negatively charged clay particle (Pouralhosseni, 2014).

Regarding heavy metals, the behaviour is similar to when having alkali metals, but they tend to bind stronger due to their higher positive charge (Ouhadi, 2006). Ouhadi (2006) proved the same changes in compressibility as Di Maio (1996), when adding lead and zinc ions to bentonite clay. As previously stated, is kaolinite less affected by the pore fluid due to its lower CEC, but previous studies have shown that the geotechnical properties of clay were affected by heavy metals. Here, the unconfined compressive strength decreased almost 40% and the compression and swelling indices doubled, for a natural clay consisting of mainly montmorillonite (Nazir, 2011).

According to Muththalib and Badouet (2019), there is a complex relationship in how the clay reacts to different concentrations and different compositions of heavy metals. Having two different metal ions (Pb and Cu) of the same concentration affect the liquid limit more than having one metal ion (Cu) with twice the concentration. Further, bentonite reacts significantly different from kaolinite, where the PI greatly decreases for bentonite and slightly increases for kaolinite. It was also shown that copper and lead affects kaolinite the most, in terms of plasticity and liquid limit. Lastly, it was stated that some heavy metals bind so strong to the clay that remediation is unnecessary, if the pH does not increase significantly, which is unlikely. (Muththalib and Badouet, 2019)

Karkush *et al.* (2013) have also tested how heavy metals affect the Atterberg limits, but also the strength, stiffness and compressibility indices. This was done by artificially contaminating the clay by adding copper sulphate and lead nitrate, which was done similar to (Muththalib and Badouet, 2019). Even though the study of Karkush *et al.* (2013) lacked details of the clay mineralogy and sampling procedure, the result showed great significance. According to Karkush *et al.* (2013), bonded copper stronger to clay than lead, due to its ability to create cation bridges (that is connecting the diffuse layer with the stern layer) between the clay particles and hence making the clay particles coarser. Finally, the results based on Karkush *et al.* (2013) show an increase in compressibility properties (void ratio and compression indices) whilst a significant decrease in shear strength. However, Karkush *et al.* (2013) found that the samples contaminated with lead did not show an increase in liquid and plastic limits or compressibility properties, which is not in line with what Muththalib and Badouet (2019) presented.

So, based on the studies above, contaminants such as heavy metals have a significant influence on the geotechnical properties of clay. How heavy metals affect remediation methods will be mentioned in oncoming chapters.

2.2. Stabilisation remediation method

Stabilisation is a way to physically prevent a contaminant to move and spreading. This is done by inserting a large auger into the soil that mixes it with a stabilising additive, often cement, see Figure 3 (Grobbel & Wang, 2013). The soil can also be extracted from the soil to be treated *ex-situ* but this procedure is rarely used, since the soil often is treated *in-situ*. Thus, the contaminant is not extracted out of the soil but is encapsulated and prevented from further spreading (CPEO, n.d.).

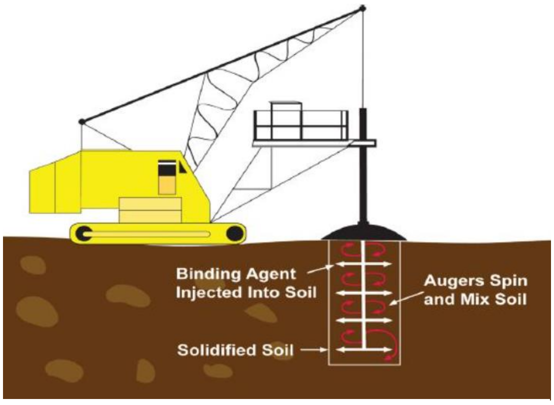


Figure 3. Stabilisation method (USEPA, 2012).

In 2001, stabilisation was accounting for the second most used remediation method in USA where 25% of all remediation were done with stabilisation, which states that it is a commonly used method. It benefits for its versatility since it can be used for many contaminants in several different soil types. The requirements of tools and technology are low which means that it is relatively cheap and easy to use, the mixing itself is also fast, making it effective. However, the soil’s hydrogeological properties are important, especially the hydraulic conductivity since if there are large differences in the injection area, the mixing can suffer from being inhomogeneous. Also, the long-term safety is not evaluated and could facing problems if the soil conditions changes (Geoengineer, 2013).

2.2.1. Alternative stabilisation methods

The common procedure when stabilising soil is to use ordinary Portland cement (OPC) and lime as stabilisers but due to the relatively high carbon footprint, more sustainable options are preferred. Using ash of some sort as a binder was studied by Coudert *et al.* (2019) and Atahu *et al.* (2019). The ash used by Coudert *et al.* (2019) was fly ash which originates as a biproduct from coal combustion. This ash has become an Alkali Activated Material (AAM) by having silicate powder (fly ash) that have reacted with an alkali metal. AAM has equal mechanical properties to OPC but a lower carbon footprint, hence a more sustainable option. Coudert *et al.* (2019) have not studied how the fly ash binder affects the strength at clay compared to OPC, but how it affects the clay on its physicochemical properties. Note that no contaminant was used, which could affect the physicochemical properties further. Coudert *et al.* (2019) concluded that no change in physicochemical properties were to be seen but due to reactions in the clay with calcium-silicon and highly solvable salts, the long-term stability could be negatively affected.

A more region-specific solution is to use of ash from coffee husks as an alternative in coffee-growing countries such as Ethiopia, where 30-50% of the mass final coffee produced is waste (Oliviera & Franca, 2015). Atahu *et al.* (2017, 2019) investigated how the coffee husk ash affects the geotechnical properties which showed that it improved the plasticity and swelling behaviour of the soil, as well as significantly increasing the bearing capacity, decreasing the compressibility and making the soil more cementitious.

Reddy *et al.* (2020) examined the effects on clay when having limestone calcinated clay cement (LC^3), instead of ordinary lime cement. By replacing 50% of the OPC with equal parts limestone and cement clay, the carbon footprint is reduced by up to 40% (Tironi *et al.*, 2013; Fernandez *et al.*, 2011; Cancio *et al.*, 2017). Reddy *et al.* (2020) evaluated the effect on LC^3 when having both zinc and lead where only the effect of having zinc has been investigated in previous studies. Further, the tested soil was a natural soil from India, that was artificially contaminated with dissolved zinc nitrate and lead nitrate and was stored for 14 days (Reddy *et al.*, 2020). Using metals in form of dissolved nitrates are a common method for contaminate the soil, used in most of the referred studies mentioned above. Treating the soil with LC^3-stabilisation was proven to be successful both in terms of strength and leachability, where the unconfined compression strength was almost doubled and the leachability met the requirements (Reddy *et al.*, 2020).

Avet and Scrivener (2020) proved that the performance of LC^3 is related to pH, where a higher pH reduces the binding in the solidifier and hence a higher risk for spread of contaminants. Reddy *et al.* (2020) found that pH was increased when adding the binder to the contaminated soil and then was stabilised.

Since 60-70% of the wood produced is waste in form of sawdust and bark, a more relevant stabilisation binder for countries such as Sweden, could be to use rest products from sawmills (Naturvårdsverket, 2010). Otoko *et al.* (2014) showed that having 4% sawdust content in clay almost doubled the compressive strength and lowered the plasticity index, Akinwumi *et al.* (2016) showed the same result but that there was no optimum sawdust content. Butt *et al.* (2016) also showed an increase of compressive strength at a 4% sawdust content and an increase for both plasticity and liquid limit. However, it could be questioned if sawdust is a representative binder compared to lime cement in terms of leachability. This has not been studied but since heavy metals, especially copper, binds strongly to organic matter, having a higher concentration organic matter in the soil could result in a lower leachability, in line with Atahu *et al.* (2017, 2019) (Naturvårdsverket, 2006).

2.2.2. Enhanced sustainable stabilisation

Using lime, fly ash or calcinated clay is a common substitute for cement, as described above. Some compounds, in reaction with water, such as calcium carbide residue (CCR), with high content of calcium hydroxide have proven to be a successful, more sustainable additive to use as a binder in clays. This due to the cementation of the added calcium hydroxide and the pozzolanic materials in the clay. A pozzolanic material consist of high amounts of silica and alumina, which in reaction with calcium hydroxide creates calcium silicate ($CaSiO_3$). Calcium silicate creates calcium silicate hydrate (C-S-H) when reacted with water. There are several different variants on calcium silicates which is why there are many different reactions, but the simplest are described by Equation 1 and Equation 2. The C-S-H acts as a glue and bind strongly to the clay particles and becomes cement, hence a higher strength and stiffness will be achieved. (Latifi *et al.*, 2018; Baltakys *et al*, 2009; Dodson, 1990)

$$Ca(OH)_2 + SiO_2 \rightarrow CaSiO_3 + H_2O \tag{1}$$

$$CaSiO_3 + H_2O \rightarrow CaO * SiO_2 * H_2O \tag{2}$$

CCR is a rest product of acetylene production, and when reacted with water calcium hydroxide is produced which properties is much like the ones for lime but without the not so environmental-friendly carbon, making it more sustainable than *e.g.* LC^3 (Suksiripattanapong *et al.*, 2017; Kampala *et al.*, 2013). Studies showed that having CCR as a binder resulted in higher strength at the soil, than for using lime (Horpibulsuk *et al.*, 2014; Kampala *et al.*, 2013). Latifi *et al.* (2018) studied how the unconfined compression strength and compression indices of kaolin clay was affected by CCR. Their study showed that the strength increased with increasing content of calcium carbide and curing time (7-90 days). However, the maximum strength occurred at 12% CCR content and 90 days. Regarding compressibility, both swelling and compression indices decreased with curing time, as well as the particle size. Since the clay have become more denser it would probably result in lesser leachability of contaminants, but this was not investigated by Latifi *et al.* (2018) and will be discussed later.

The reason why ash, especially fly ash, is commonly used for replacing cement as a binder is because its high pozzolanic content of silica. Hence having a combination of pure pozzolanic material and calcium hydroxide should be the best alternative, this since the more pozzolanic material there is the finer the cement will be, probably resulting in better performance for both leachability and strength. Due to high pH, the natural pozzolanic material that exists naturally in clay dissolves and reacts with the calcium ions and creates a cement-like material (Horpibulsuk *et al.*, 2013).

Further, by adding pozzolanic material, the effectiveness is increased since more silica and alumina can react hence more cement can be created (Horpibulsuk *et al.*, 2013). This is also most likely why having a too high CCR content resulting in no further increase of strength, as shown by Latifi *et al.* (2018) and Horpibulsuk *et al.* (2012). This is visualised in Figure 4, where the graph is divided into three different zones: active, inert and the deterioration zone (Horpibulsuk *et al.*, 2012). It is evident that in the active zone (< 8% content), all calcium carbide content has reacted with the pozzolanic material in the soil. The soil is "saturated" and cannot react with more additive, hence it will result in a decrease instead of an increase in strength, since the additives have no strength on their own and thus weaknesses the clay (Horpibulsuk *et al.*, 2013).

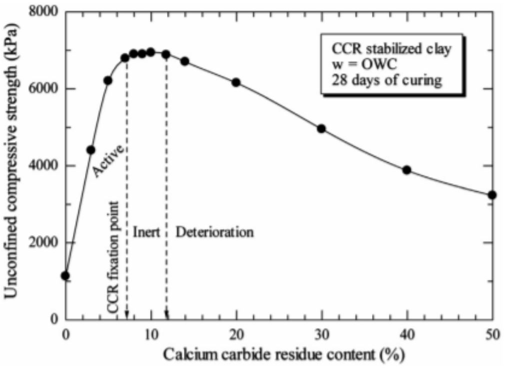


Figure 4. The different zones when having pozzolanic reactions in the soil between calcium hydroxide and clay (Horpibulsuk et al., 2012).

As could be assumed, the fly ash did only affect the inert zone, this since the amount pozzolanic material in the clay is not limiting the reaction in the active zone. Since the reaction takes time, the strength increased with longer curing times, soaking the clay with water was proven to reduce the strength due to the interference with repulsive forces in the water. Having a higher content fly ash (0, 6 & 12%), showed a linear increase in the inert zone (10% CCR content) and a slight linear increase in the deterioration zone (20%). Where having 10% CCR content resulted in higher strength than for 20% content. So, having a pozzolanic ash in combination with an additive rich in calcium hydroxide has proven to enhancing both strength and leachability of the soil (Horpibulsuk *et al.*, 2013).

2.2.2.1. Biochar

Biochar, a rest product from pyrolysis of any type of biomass, is commonly used as an additive in soil, to enhance the binding of nutrients and making it more fertile. Biochar is, like fly ash, a material that increases the pozzolanic activity and which in combination with cement (or just itself) and a binder, such as calcium hydroxide, enhances the strength and can encapsulate heavy metals (Nair *et al.*, 2020). It has proven great potential in remediation efficiency, with a reduction of bioavailable heavy metals of up to 92% and is on paper a more sustainable substitute to fly ash, since it also acts as a fertilizer for plants (Houben *et al.*, 2013; Wang *et al.*, 2020).

However, it is discussed if biochar is as stable to use as fly ash, this since its binding potential and stabilisation potential can differ greatly even though the biochar originates from the same production batch (Wang *et al.*, 2020). Further, Bian *et al.* (2014) and Jia *et al.* (2017) found a variation in stabilisation efficiency from as low as 18.8% to 54.5%, which could result in poor remediation. This variation is due to that biochar is sensitive and the slightest difference in production could result in different properties in the biochar, even from the same feedstock (Uchimiya *et al.*, 2010; Cha *et al.*, 2016; Qian *et al.*, 2016). Also, since biochar can be made from any type of combusted biomass, the chemical composition can vary significantly from being simple and containing few chemicals, to complex and containing several different chemicals (Oliviera *et al.*, 2017). Furthermore, parameters such as combustion temperature will greatly influence the remediation potential for heavy metals, this since it can change the CEC, pH and surface area, all related to how well metals bind to the material (Sizmur *et al.*, 2017; Li *et al.*, 2018; Zhang *et al.*, 2018). If the rate of temperature increases or decreases, have also a significant influence on the binding potential, this since it affects the functional groups on the surface on the biochar and thus preventing heavy metals to bind (Ding *et al.*, 2014; Sizmur *et al.*, 2017).

Having a sufficient level of remediation of heavy metals requires high concentrations of biochar, resulting in an imbalance in the soil nutrient content and may lead to soil alkalinity and a reduction in biomass. Hence making it a more inaccurate additive to use today, but by having a more controlled production and more knowledge about the soil environment, it is the most sustainable additive to use in future. (Wang *et al.*, 2020)

2.2.3. Remediation efficiency

The leachability or influence of contaminants have not been discussed in the mentioned studies above, which could be a concern. However, the pozzolanic reaction between the alumina and silica in the soil, increases the pH. This since the pore water dissolves the calcium hydroxide in the additive, and thus creating dissolved calcium ions. These positively charged calcium ions penetrates the double layer, as mentioned earlier, and binds to the negatively charged clay particle. By doing this, the clay becomes more compacted, since the particles are stronger bonded together. Further, the pozzolanic material in the clay dissolves at an increased pH, and reacts with the dissolved calcium ions, causing a stronger bond with time. When the calcium ions have reacted with the pozzolanic material, the pH will slightly decrease, thus indicating that no further curing is needed. If the pH continues to decrease the C-S-H and the binding between the clay and the metal ions could be affected negatively and posing a risk for higher leachability, this have however not been studied. (Siddiqua and Barreto, 2018)

Further, the solubility of copper decreases with increase in pH, hence lesser leachability is to expect at high pH Cuske *et al.* (2013). Since the buffering capacity is related to the CEC, where a larger CEC results in a higher buffering capacity, a soil with lower CEC is less resistive to pH changes (Luo *et al.*, 2015). Moon *et al.* (2013) studied this by investigating the remediation efficiency of heavy metals with an enhanced stabilisation in clayey sand. Copper sludge was used as additive, in combination with fly ash as pozzolanic material. The results showed that over 95% of the added copper and lead contaminants were removed, hence a very efficient method. By SEM-photos, it was evident that the soil particles have formed ettringite particles, that is a hydrous calcium aluminium sulphate mineral, as a product of pozzolanic reaction (Moon *et al.*, 2013). According to Gougar *et al.* (1996), two worth metal ions can replace the two worthy calcium ions in the ettringite, which could be a reason for the efficient remediation. Furthermore, Moon *et al.* (2013) stated that the basic conditions are favourable for remediation of heavy metals, hence a low leachability should be expected when considering all parameters.

2.3. Influence of silica to calcium ratio

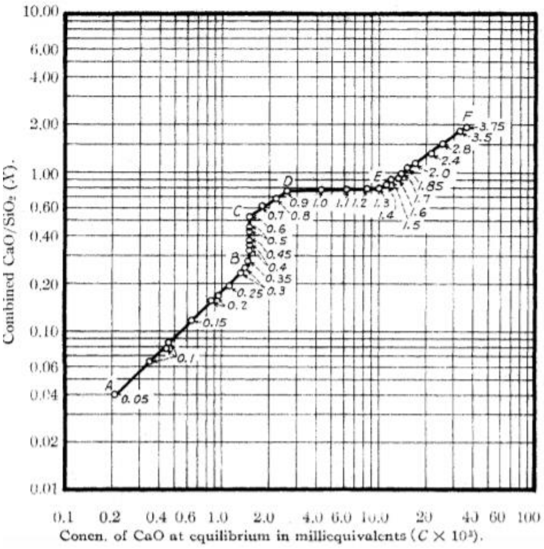


Figure 5. Adsorption of calcium on silica, at increasing concentration of calcium oxide.An increase means that more calcium is adsorbed. Note that no increase in adsorption is obtained when increasing the ratio from 0.9 to 1.3. (Beitlich, 1938)

As mentioned earlier, a certain ratio of fly ash to CCR was evaluated and discussed by Horpibulsuk *et al.* (2012, 2013). However, this ratio was only related to the increase in strength and not the actual ratio where the additives bond the strongest. Beitlich (1938) investigated how the adsorption changed with different ratios of calcium oxide and silica, ranging from 0.05 to 3.75. This was done by using Freundlich adsorption isotherms, that is a method to present the relationship between the ratio of adsorbed material (adsorbate) and the mass of adsorbent with an increase in concentration (Sparks, 2003). Further, Beitlich (1938) found that an increase in concentration of calcium oxide resulted in more mass adsorbed. As can be seen in Figure 5, the relationship is not linear where the adsorption increases significantly for some ratios and not increase at all for other ratios. To conclude, the adsorption increases with an increase of calcium oxide to silica ratio, showing great binding potential even at high concentrations (Beitlich, 1938).

Further, Greenberg (1961) studied what factors that affects the reaction between silica and calcium hydroxide. Temperature, concentration of calcium hydroxide and silica respectively, type of silica and the surface area of the silica was evaluated. This was conducted by measuring the conductance in a solution with both silica and calcium hydroxide, where the conductance is related to the concentration of calcium hydroxide. To simplify it can be said that the less calcium hydroxide a solution contains, the lower is the conductance. Hence, the more effective reaction between the silica and the calcium hydroxide, the lower is the conductance since more of the calcium hydroxide has bonded and created calcium silicate hydrates. It was shown that the conductance decreased faster when having a higher ratio of calcium hydroxide and silica, also when having finer particles of silica. It was also shown that having a higher concentration of calcium hydroxide on its own did not increased the rate of decrease of conductance. This could be an explanation why Horpibulsuk *et al.* (2012) showed no further increase in strength at higher CCR concentrations. Temperature was also influencing the reaction, where a higher temperature increased the reaction time. Lastly, the surface area was shown to have the greatest influence on the reaction speed whit differences over 100 times. However, having a too high surface area could be less efficient, this since the pores are faster filled with calcium hydroxide than the silicate dissolves and it does not react as much as it could do. Greenberg (1961)

2.4. Heavy metal reactions and adsorption properties

Pure kaolin clay consists of kaolinite, $(OH)_8Si_4Al_4O_{11}$, which is a 1:1 clay mineral, that is that it has one octahedral silicate layer ($(Si_2O_5)^{-2}$) and one tetrahedral hydroxide layer ($(Al_2[OH]_4)^{+2}$). Thus its cations, the ions that can react, is silica and alumina and binds through hydrogen bonds between the hydroxyl group and the oxygen molecules. Due to its slight imbalanced charge, weak van der Waals bindings can also occur between the particles. See Figure 6 of the layout of kaolinite. The charge on the kaolinite particles is pH dependent, where a higher pH results in negatively charged edges and high cation exchange and vice versa for lower pH, making its ability to bind to positively charged ions pH dependent. (Mitchell & Soga, 2005)

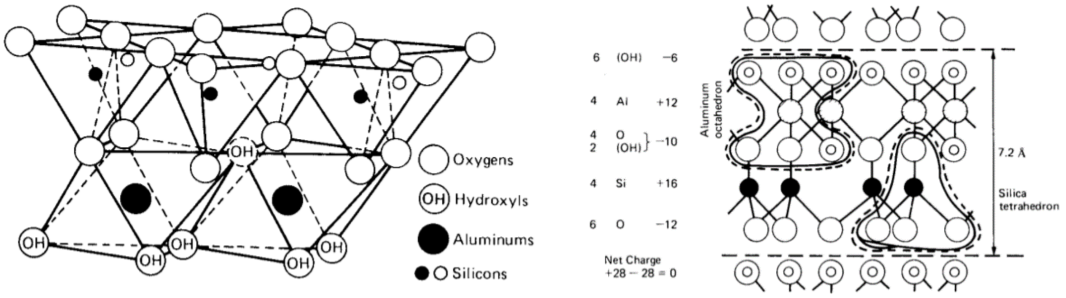


Figure 6. Composition of kaolinite to the left, and charge distribution to the right. The slight imbalance between the silica tetrahedron and the alumina octahedron makes it possible to create van der Waals bindings. (Mitchell & Soga, 2005)

Calcium hydroxide, $Ca(OH)_2$, also referred to as slacked lime, is an inorganic chemical compound with high pH (12.5). As for the kaolinite, it binds strongly to each other by hydrogen bonds and is structured 2:1, that is two hydroxyl groups at the edges and calcium ions in centre. (NCBI, 2021)

Silica, SiO_4, is a mineral in tetrahedral form, this due to its highly charged, but small, cation (as can be seen in Figure 6). Most of the minerals with a highly charged cation are formed in tetrahedral or octagonal shape, this to have the cations far from each other, since the molecules are repelled from each other due to the strong charge. The silicon binds to oxygen ions by covalent and ion binding and the silicates binds to each other by ionic binding at the edges, making it possible to have several different types of silicates such as islands, rings, chains etc. (Mitchell & Soga, 2005).

However, at high pH these silicates are broken down, when adding calcium hydroxide, where the hydroxyl group becomes water and the calcium and silicon binds by covalent binding and the molecules binds to each other by strong hydrogen bonds (Hou *et al.*, 2018). When reacted with water, the silica transforms into silanol, that is silicon with a hydroxyl group (S-O-H), and the alumina to aluminol, that is alumina with a hydroxyl group. Where the calcium only reacts with the silicon and not the silanol group, and creates C-S-H, which is a complex compound shown in Figure 7. (Dalstein *et al.*, 2017). (Mitchell & Soga, 2005)

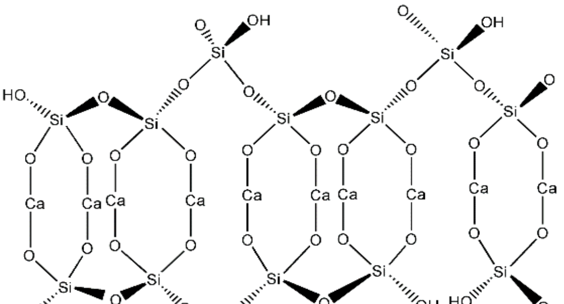


Figure 7. Calcium silicate hydrate (C-S-H), a reaction between calcium hydroxide and silica. Note the silanol groups at the edges. (Wang et al., 2018)

Further, when calcium ions dissolve the pozzolans (the silica and alumina) in kaolinite, the edges become more negatively charged due to the aluminol, silanol and hydroxyl groups that have been created due to the breakdown of the pozzolans. This results in that more of the added calcium now binds to the edges of the kaolin, to these groups, instead of continuing to dissolve the internal pozzolans, which slows down the pozzolanic reaction. (Chemeda *et al.*, 2018)

Few have studied how heavy metals react with calcium and silica in soil. Cairns *et al.* (2006) investigated the uptake of copper ions on C-S-H, in form of copper hydroxide nitrate and copper chloride hydroxide. According to them, it is the hydroxyl group on the surface of the silicate, the silanol group, that binds to copper. Also, that it is unlikely for the copper to bind to the same location on the silicate as calcium does, this since the copper was found to bind to the calcium silicate and not the silicates on their own. Hence, copper nitrate will most probably not interfere with calcium reacting to silicate but calcium silicate itself. Since both hydroxyl groups and copper ions lower the pH, if the copper binds to the silica's hydroxyl groups, the pH will decrease, which was also proven to happen when testing. This was shown by removing the hydroxyl group on the silanol by acid washing, that binds to the hydroxyl groups, where it was found that less copper was able to bind. Also, when removing the calcium, more copper would in theory be able to bind, if it binds to the silica rather than the calcium ions, which was proven to happen by SEM-analyses. Hence the copper binds to the silica rather than the calcium, on the calcium silicates (Borrmann *et al.*, 2005; Cairns *et al.*, 2006).

Based on this, there is low probability that the copper interferes with the pozzolanic reaction between the silica and the calcium hydroxide, but instead interferes with the resulting calcium silicates. However, since copper nitrate (not hydroxide) does not have a hydroxyl group, and thus creating weaker hydrogen bonds with the silanol groups, it could make it possible to interfere with the pozzolanic reaction, since it could bind on the same location to the silica as the calcium ions do (directly on the silica) (Cairns *et al.*, 2006). Lastly, Cairns *et al.* (2006) saw that copper nitrate hydroxide had larger particles than the copper chloride hydroxide, where the particles only followed the silicate particles and not the whole substrate. This could indicate that some of the copper ions did not bind to the hydroxyl groups on the silanol (Cairns *et al.*, 2006).

Chen *et al.* (2009) have also studied the C-S-H's potential binding capacity to heavy metals. Depending on the level of hydration and curing time, the C-S-H will have different structure and density, where it becomes denser and more complex with increasing curing time, hence achieving a larger surface area (Taylor, 1997; Chen *et al.*, 2009). The curing and reaction process when creating C-S-H is highly complex, especially when in reaction with heavy metals. C-S-H encapsulates heavy metal ions by having calcium ions bonding to the negatively charged surface (due to the hydroxyl groups), creating a second layer, and where the cations in the heavy metals can create a third layer by bonding to the calcium ions and the hydroxyl groups (Chen *et al.*, 2009). Hong and Glasser (2002) confirmed that low valence metals adsorb well to C-S-H, by bonding to the silanol groups. It was also stated that the calcium ions on the surface can be exchanged by two-worthy heavy metal ions, which at high pH form hydroxides that replaces the calcium hydroxides in the making of C-S-H (Chen *et al.*, 2009). This was shown were copper and lead increased the hydration due to faster reaction of the calcium hydroxides. Further, zinc was found to reduce the hydration process, this since it reacted with the calcium and formed calcium zincate, which acted as a coating around silica particles and prevented further reactions with calcium ions. At later hydration stages, when pH has increased due to the C-S-H formation, the calcium zincate was dissolved thus the calcium could react with the silica and hence the solidification went faster. Also, heavy metals decrease the pH and can affect the structure and decrease the binding properties of the C-S-H, by lowering the binding potential for the calcium hydroxide and making it less solid (Chen *et al.*, 2009).

As mentioned in 2.1.2., the electro potential is an important aspect when evaluating how heavy metals bind to clay particles. The adsorption force between the clay particles is referred to as the zeta potential, where a lower zeta potential corresponds to a higher repulsive force and vice versa. Hence zeta potential is a measure of the surface potential on a particle (this can be seen in Figure 2). By adding positively charged metal ions, the repulsive forces are balanced out, this was shown by Yukselen and Kaya (2003) where the zeta potential increased when adding heavy metal ions to a basic kaolin clay, thus the adsorption increases. However, in general, a lower pH results in a more positive zeta potential and at higher pH it becomes more negative, it can also fluctuate depending on the concentration of ions. For example, at some concentrations the zeta potential was strongly positive while at others just slightly negative. This results that in basic conditions the surface potential of the kaolin clay is strongly negative, thus creating strong bonding with positively ions such as heavy metals. (Yukselen & Kaya, 2003)

Trussel and Spencer (1994) studied how heavy metals interfered the stabilisation process of cement, which also is based on C-S-H production. They found that lead precipitates on the surface of the calcium silicates and causing an impermeable layer, preventing the calcium silicates to react with water. It was also shown that the strength of fly ash mixed with lime decreased noticeably when having low concentrations of lead. The effects were the same for zinc, in accordance with Chen *et al.* (2009). Further, it was shown that zinc, as well as other metals, reacted with the calcium and formed ettringite, that is a calcium aluminium sulphate mineral, that could lower the permeability of the cement due to cracking but also preventing it from reacting with silica (Poon *et al.*, 1985; 1986). Tashiro *et al.* (1977) did also show how heavy metals disturb the cement process when stabilising using OPC. The results were in-line with Trussel and Spencer (1994) but slight positively effects were obtained at low concentrations of heavy metals. According to Tashiro *et al.* (1977) the heavy metals create double salts between the metals and the calcium ions, preventing them to react with water, resulting in softer cement. Tashiro and Oba (1980) also stated that heavy metal chlorides such as copper chloride, reacted with the silica in the cement, the effect of this was however unknown.

2.4.1. Heavy metal adsorption on kaolin clay

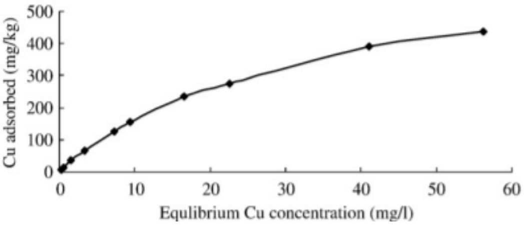


Figure 8. Adsorbed copper on kaolin clay vs. increase in concentration. (Wang et al., 2009)

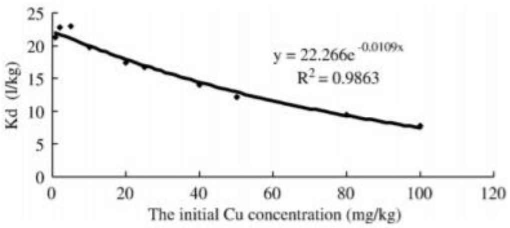


Figure 9. How the distribution coefficient (kd) varies with concentration. Wang et al., 2009)

Wang *et al.* (2009) have studied the uptake of copper in pure kaolin clay and factors that affect the uptake. The study was conducted similar to Beitlich (1938), by using Freundlich isotherms. The result showed on an almost linear relationship between the adsorbed copper and increase in concentration, see Figure 8. The distribution coefficient, k_d [1/kg], is used to evaluate the ratio between the adsorbate and the initial concentration in the added solution, hence it describes how well the material bonds. Where a 1:1 relationship indicates that all the adsorbate has adsorbed, and a lower k_d-value states higher adsorption. A decreasing k_d-value with increasing concentration indicates that the adsorption increases with increase in concentration. The adsorption showed an exponential behaviour, as can be seen in Figure 9, which indicates that kaolin clay adsorb copper equally well at almost all concentrations. Lastly, it was also shown that more copper was adsorbed at higher pH and at higher concentrations, with a behaviour similar to Beitlich (1938), see Figure 10. (Wang et *al*., 2009).

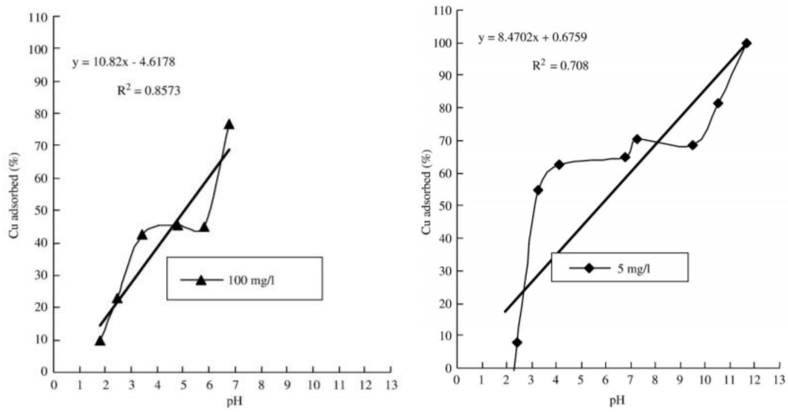


Figure 10. pH dependence of adsorption of copper. Note the difference in inclination of the slope for 100 mg/l. (Wang et al., 2009)

The reason for this increase in adsorption potential is due to, as mentioned by Mitchell & Soga (2005), Yuksulen & Kaya (2003) and Borrmann *et al.* (2005), that the zeta potential decreases at higher pH, leading to that the edges of kaolinite are more negatively charged and thus have higher cation exchange. Therefore, the copper ions bind more likely to the hydroxyl groups in the kaolinite rather than the alumina and silicates. It should be noted that Frost (1998) and Ma and Eggleton (1999) also shown tendencies that some of the lead and zinc ions bonded to the hydroxyl groups in the kaolinite. Further, Miranda-Trevino and Coles (2003) speculated that the van der Waals forces could be affected by having substitution of hydrogen ions for heavy metal ions and having swelling as a result of weaker bonded clay minerals. This was visualised by studying the metal ion uptake and the displacement of hydrogen ions, were an increase of adsorption resulted in an increase in displacement (Miranda-Trevino & Coles, 2003).

To conclude, kaolin has two locations where the copper can bind, referred to active and passive sites. The silica and alumina, on the surface that causes a slight negative charge which is pH-independent and is therefore active sites, where the positive copper ions can bind to. On the edges, where the hydroxyl groups are located, metals can bind via hydrogen bonds by exchanging hydrogen ions for metal ions. However, the charge on these hydroxyl groups is pH-dependent and are negatively charged only at high pH's, thus being a passive binding site. Therefore, at high pH, most of the metals bind onto the hydroxyl groups and not to the silicates and alumina, the active sites, which is what the mentioned studies above have shown. (Matlok *et al.*, 2015)

Kaolin has, due to its composition, smaller surface area than other clay minerals and this will have an impact on the amount of metal ions that can adsorb to it, as well as having a longer reaction time (Otunola & Ololade, 2020). As mentioned earlier, adsorption between calcium hydroxide and silica increased with temperature but the ideal temperature for adsorption of metals to clay has found to be around 22-29^o C (Potgieter *et al.*, 2006). Coles and Young (2002) stated that kaolinite had fully adsorbed metal ions after 6 h of storage, though this depends on the mixing process. The metal uptake can be improved by having more organic matter, so called "organoclays", this due to the chelating groups in the organic matter (that is two or more functional groups that binds with a metal ion in a special ring formation) that binds stronger than to the clay mineral (He *et al.*, 2014; Richey 1975). Further, Unuabonah *et al.* (2009) found that organically modified kaolin clay adsorbed eight times more than normal kaolin clay. Using acids to increase the CEC of the clay was found to improving the metal adsorption significantly, where a stronger acid increases the CEC more (Shawabkeh *et al.* 2007). It should be noted that having too strong acids can destroy the clay's structure and lower its adsorption, thus causing new problems with having more chemicals in the soil (España *et al.*, 2019). So, combining different methods to improve the metal adsorption in the clay could improve the remediation effectiveness dramatically (Otunola & Ololade, 2020).

2.4.2. The influence of soil type and clay composition

The above mentioned applies to kaolin clay, this since that it is the main clay that will be tested in this study. However, it is rare that the soil consists of one pure clay mineral and this could influence the reaction processes. A clay found in field often consist of a mixture of minerals, such as illite, kaolinite, smectite etc. According to Ito and Wagai (2017), illite and smectite are often found to be the dominant clay mineral in over 70% of all clays in the world. Illite and smectite are both a 2:1 mineral, and with a similar composition as could be seen below in Figure 11. Both kaolinite and illite are nonswelling clays, whereas smectite can swell greatly when in contact with water (Anderson, n.d.). Compared to kaolinite, both illite and smectite are more complex and have neutral charge with significantly higher CEC. This more neutral charge results in less attraction between the minerals and positive metal ions, but due to the higher CEC more ions can bind which increases the concentration of heavy metals on the surface of the minerals. The CEC relates to the surface area where smectite has largest surface area and kaolinite the smallest, this results in less bonding can take place on kaolinite, but since kaolinite has larger negatively charge, it attracts positively ions better.

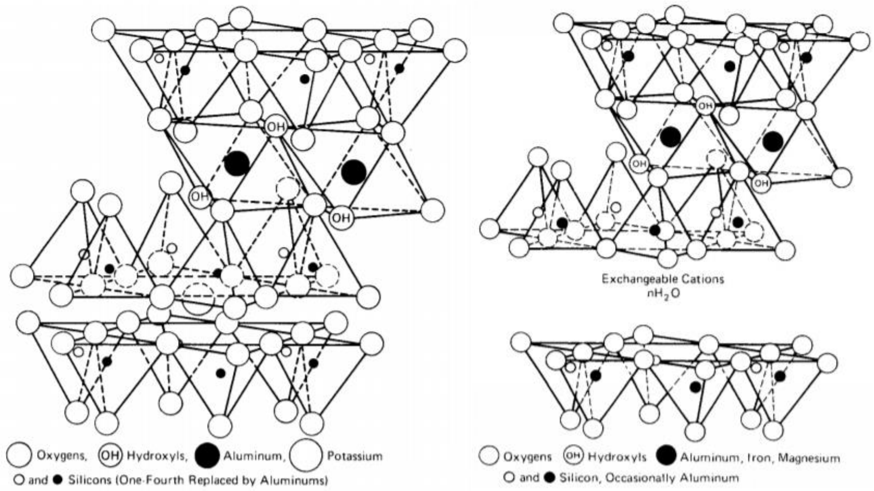


Figure 11. Ion composition of illite (left) and smectite (right). Note the small differences, the potassium ions and that the aluminium could be changed for iron or magnesium. (Mitchell & Soga, 2005)

Due to the low content of pozzolans (silicates and aluminates) in illite and smectite, more binding additive could be needed. However, Turan *et al.* (2011) and Mhamdi *et al.* (2013) have proven that illite and smectite behaves similar to kaolinite in terms of adsorption to heavy metals, indicating that they are equally good at binding to heavy metal ions. Smectite, for example, encapsulates the metal ions better due to its expanding properties. It should be noted that the stronger hydrogen bonds between the kaolinite minerals results in stronger bonding between the cations, compared to both illite and smectite that bonds with weak van der Waals bindings (Mitchell & Soga, 2005). The fact that heavy metals bind to hydroxyl groups at high pH, more heavy metals could be able to bind to kaolin since it has more available hydroxyl groups than both illite and smectite. Therefore, there is a risk that the metal ions will bind more to the active sites (the alumina and silicate) on the illite and smectite and thus preventing them to react with the binders (Mitchell & Soga, 2005; Matlok *et al.*, 2015). There is a discussion that smectite itself can act as a treatment for binding heavy metals, due to its high CEC and large surface area, and thus no need for cement or an additive (Niu *et al.*, 2019). Having only bentonite as a stabiliser is not favourable in terms of strength, this since it has low pozzolanic activity which results in less C-S-H production (Niu *et al.*, 2019).

The reaction between calcium hydroxide and the different clay minerals will differ, this due to the mineralogy and the pozzolanic content. According to Morris and Zpik (2009) the structure and adsorption potential of smectite are strongly dependent on calcium ions, where the zeta potential is more negative and the structure more complex in presence of calcium ions. Ouhadi *et al.* (2021) stated that the precipitation of lead ions increased at low-medium pH (2-9) and very high pH (> 13), compared to kaolinite where more lead was adsorbed at only higher pH (>11) (Muththalib & Badouet, 2019; Atanassova & Okazaki, 1997; Cuske *et al.*, 2013). This due to the high content of calcium carbonate in smectite that increases the buffering capacity greatly, in combination of having high concentrations of hydroxide ions from the lead hydroxide in the contaminated soil (Young *et al.*, 2012). Lastly, illite have a high content of calcium oxide (7%), meaning that less additives are needed for it to react with silica (Uddin, 2017).

Having other soil types, such as sand, silt and gravel affects the stabilisation process, since the pozzolanic content in the different soil types differs. Sand and silt are made of pozzolanic minerals which consists of silica and aluminates, similar to kaolinite. However, they do not bind to heavy metals as strong as kaolinite, and other clays, do, due to lack of charge and cations (Lines-Kelly, 1993). The result of this could be that more metal ions bind to the C-S-H and not to the soil itself. One of the most critical differences is that water could have greater influence, this due to the high permeability and greater groundwater flow in sandy soils than clayey. Thus, having excess water are likely to disturb the curing process of the C-S-H, as shown by Horpibulsuk *et al.* (2013), where flushing a stabilised soil resulted in lower strength. Other soils such as gravel can bind well to heavy metals but can also bind poor to heavy metals (Sizirici *et al.*, 2018). Indicating that all metal ions will be bonded to the C-S-H and, again, having potentially high water flows can also cause problems with the curing, resulting in a weak stabilised soil and thus potential risk for spreading.

2.5. The effect of having several types of contaminants

There are often different types of contaminants in the soil and rarely just one type of heavy metal, this due to industries often pollute different contaminants depending on their internal processes. This could influence the C-S-H-production since its reaction depends on the chemicals in the soil. Few, probably no, studies have been made investigating the effects of having more than one type of contaminant when stabilising soil. What can be said is that the adsorption of PFAAs (perfluoroalkyl acids) onto clay decreases with increase in pH, indicating that treating clay with heavy metals in alkaline environments can increase the leachate of PFAAs, which is toxic to the environment (Zhao *et al.*, 2014). Also, Wainipee *et al.* (2013) showed that oil changed the adsorption potential at clay significantly, this when studying the adsorption potential between arsenic and clay. Further, the oil acts as a coating on the clay particles, reducing their effective surface area and thus preventing binding to the metal ions. Cation bridging, is also prevented by oil coating (Wainipee *et al.*, 2013). This could result in greater leaching of heavy metals and could affect the pozzolanic reaction through preventing calcium hydroxide to react with the silica on the kaolinite's surface, hence a weaker cement and lesser remediation performance.

2.6. Potential environmental risks and impacts of enhanced stabilisation

Having an enhanced stabilisation based on reaction between silica and calcium hydroxide has proven, according to the studies above, to be successful in terms of strength, stiffness and somewhat leachability. However, it is crucial that this mixture does not imply potential risks or contribute to an increase in emissions to the environment. Bajpai *et al.* (2020) have compared the environmental impact of fly ash stabilised soil versus cement stabilised soil. Several studies have shown a decrease in CO_2 emissions, some as high as 80% but some as low as 9%, depending on outer factors such as materials, transportation and extraction process (McLellan *et al.*, 2011; Stengel *et al.*, 2009; Turner and Collins, 2013; Yang *et al.*, 2013).

Bajpai *et al.* (2020) investigated three risk targets; eco system, human health and resources, and it was shown that when using stabilisation with fly ash and silica fume the environmental impact was almost half compared to when using cement. Also, when considering transportation processes the impact was 42% lower than ordinary cement, hence using enhanced stabilisation proves a significant reduction in environmental impact (Bajpai *et al.*, 2020). However, the enhanced stabilisation mix could pose a risk for toxicity, depending on its content. Usage of fly ash in combination with sodium hydroxide and sodium silicate increased the freshwater toxicity by 18% and human toxicity by 30%, this mixture is however not commonly used.

Using calcium hydroxide (slaked lime) pose little to no risk to the environment due to its low bioavailability but requires relatively high amount of energy to produce (Lewis *et al.*, 2016; Pacyna *et al.*, 2009). Producing one tonne of calcium oxide (quicklime) produces 0.75 tonne of CO_2, compared to OPC that produces 0.95 tonnes CO_2 per tonne cement (Pacyna *et al.*, 2009; Davidovits, 2015). Slaked lime is produced by hydrating quicklime with water and mixing it, hence it requires more energy than quicklime (Pacyna *et al.*, 2009). However, the amount of calcium hydroxide needed for stabilisation is roughly 5-10% of the soil weight (depending on the soil and water content), compared to using a cement with a water to cement ratio of 0.3, where 75% cement is needed (Horpibulsuk *et al.*, 2012; Wang *et al.*, 2018). Hence, the total environmental impact of using calcium hydroxide as a binder is much less than using cement.

2.7. Summary of other literature

Li *et al.* (2019) have studied the performance of stabilisation with magnesium oxide (MgO), which is a substitute for calcium hydroxide. This has proven to have similar solidification properties but with a reduction in strength, that could potentially weaken the soil from its initial strength. Further, Li *et al.* (2019) have summarised the effects of stabilising heavy metals with both OPC and other additives. Table 1 below summaries the relevant studies for this study, where different binders have been used. However, the main objective was the encapsulation of the solidifier, thus having as low leachability as possible. Note that few have studied the impact on the strength and stiffness in combination with heavy metals.

Table 1. Summary of relevant studies on stabilisation method's effect on the soil. The soil differs from clayey to sandy. (Li et al., 2019)

Reference	Binder	Conclusion
Wu *et al.* (2018)	MgO	Reduced the leachability of Pb.
Wang *et al.* (2018)	MgO	Reduced strength compared to OPC but more solidified soil.
Zhou *et al.* (2016)	OPC	OPC adsorbed heavy metals well.
Jiang *et al.* (2014)	OPC	It was found that Zn affected the hydration of C-S-H and caused a weaker cement in terms of strength and stiffness.
Du *et al.* (2014)	OPC	Zn affected the strength of the stabilised soil, by disturbing the hydration.
Hale *et al.* (2012)	OPC & CaO	The binders encapsulate heavy metals good, but OPC slightly better.
Cubkucuoglu and Ouki (2012)	MgO	Not efficient for solidifying soil containing heavy metals.
Yin *et al.* (2006)	OPC & OPC with rice husk	The strength increased at low metal concentrations but decreased greatly at high concentrations. Adding risk husk improved the encapsulation, though unclear if it improved the strength.
Boardman (1999)	CaO	Similar results as Yin *et al.* (2006), weaker strength with soil stabilised with CaO.

Based on the studies mentioned in Table 1 and in Chapter 2, there is a lack of studies on how heavy metals affect the strength, and especially the stiffness, of stabilising soil with enhanced additives.

3. Remediation in Practice

Landfills are a growing problem, both in terms of sustainability, administration and practicality, where it requires a vast amount of space and posing a risk for not achieving future environmental goals (SVT, 2020; Volchko *et al.*, 2016). Based on data from 2017, the most used remediation method in Europe is still “dig and dump”, or other methods that requires excavation, where *in-situ* methods are used to a small extent depending on the country, see Figure 12 (EEA, 2017).

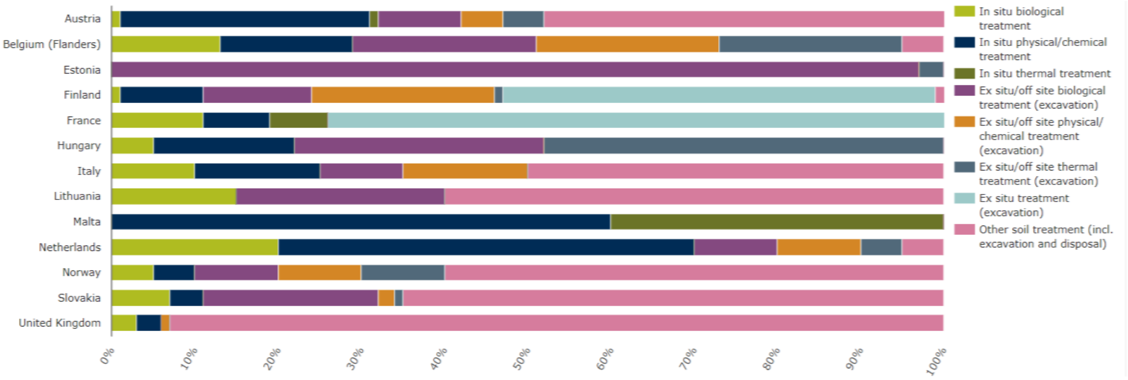


Figure 12. Most frequently applied remediation techniques for contaminated soil, based on data from 2017 (EEA, 2017).

This is also the case in Sweden, where “dig and dump” is a common remediation method (Volchko *et al.*, 2016). Once the soil is dug up it is heavily regulated by Swedish environmental law, making it complex legally dealing with soil masses. To simplify, it can be said that having soil that must be dug up is classified as waste, if it should not be used within a reasonable time, however if the soil should be used it is not classified as waste (Naturvårdsverket. 2020a). Further, the soil can be classified as either “clean” or as a byproduct, where the following regulations apply for clean soil: i) the soil is not contaminated ii) the soil will be used at the current construction and iii) the soil does not pose a risk for the environment or human health (SGI, 2019a). For the soil to be classified as a byproduct it must: i) have been made during fabrication ii) be used directly without any further processing and iii) be further used without posing risk to the environment or human health (SGI, 2019a). Having concentrations over the so called MRR concentration, that is the concentration at which the contaminant poses no risk, classifies the soil as waste and working with this soil must be registered to the municipality (Länsstyrelsen Västra Götalands Län, 2018). Soil that does not change its physical, chemical or biological properties due to different circumstances are defined as inert waste, such as gravel and rock (Avfall Sverige, 2018).

The prioritization of how the soil should be used is as follows: firstly, it should be reused, secondly, the material should be recycled and lastly it should be used as a landfill or as fill (SGI, 2019b). Therefore, based on this the remediation methods have a significant influence on how the soil is classified, thus a better *in-situ* remediation method is the most sustainable. Due to high availability for landfills, low treating costs and low interest for new remediation techniques, contaminated soil is often remediated by “dig & dump” (Naturvårdsverket, 2013; SGI, 2012).

This is an old method commonly used if it is unfeasible to remediate the soil *in-situ*, due to high concentrations of contaminants or that the soil should be excavated anyway for future constructions. However, this requires vast amounts of land and transports for disposal to landfills, often located far outside large cities. Sweden is placed at the bottom when it comes to recycling materials and this due to ineffective regulations regarding remediation and storing soil on site (Byggvärlden, 2019).

As mentioned above, remediating the soil with “dig and dump” is, due to the regulations, not as user-friendly as it might seem. This since the flow of the masses is complex, as can be seen in Figure 13, and many decisions must be made and must be reported to the local authorities. Meaning that it requires much administrative work, especially when compared to *in-situ* processes. (SGI, 2019c)

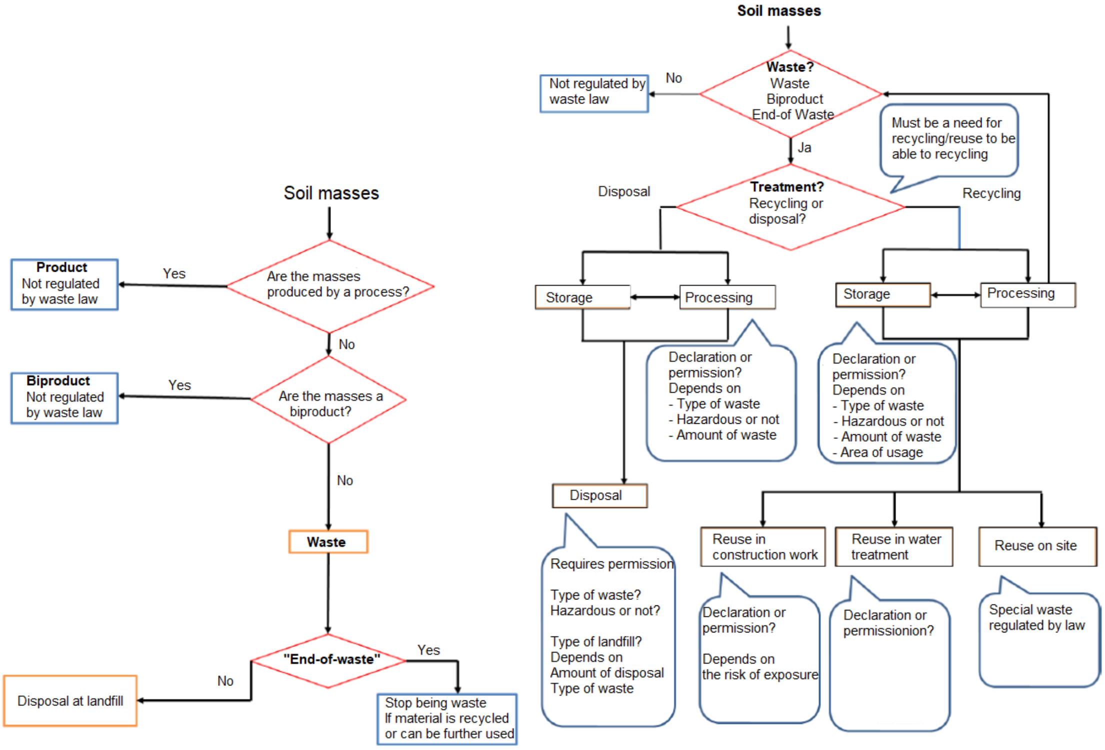


Figure 13. Flow scheme of soil masses (left) and management of contaminated soil (right) based on SGI (2019c).

3.1. Regulations and guideline values

The Swedish EPA (Naturvårdsverket) is the governing authority that decides whether the soil should be remediated or not. By classifying the future land use as sensitive (KM) or less sensitive (MKM), where residential areas are more sensitive and industrial areas are less sensitive, the soil must meet a specified guideline value for a given contaminant. Note that no remediation is needed if the concentrations are lower than the MRR concentration. However, there are no regulations or guideline values for the leachability for the soil, that is how well the contaminant can spread, which could cause ineffectiveness during construction since the soil is remediated, independent on the leachability. The Swedish EPA states that these guideline values are set to a level that does not pose a risk for humans or the environment, but a concentration over the guideline values does not necessarily need to pose a risk. Further, it is stated that they are based on "normal-dense" soil, thus not applicable in clayey or sandy soils with different permeability, and that collaboration effects are not considered. This means that the guideline values can be inappropriate for many soil types and site conditions. The leachability is only regulated in landfills, where the soil must meet specific requirements if it is inert waste or hazardous waste. Since contaminants binds more strongly to fine-grained soils and that the permeability of a soil is decreased with an increase in fine-grained material, these soils will have a lower leachability than coarse-grained soils. Thus, neglecting the leachability could lead to more remediation since the exposure only depends on the concentration of the contaminant, and if it is lower than KM or MKM. Site-specific guideline values can be used if there are specific risk objects that poses a risk, or as mentioned earlier, that the site conditions are special where the general guideline values do not apply. However, the land use (KM/MKM) still decides if remediation is needed. (Naturvårdsverket, 2010; Naturvårdsverket, 2020b).

Using site-specific guideline values is one way to prevent unnecessary remediation, but it is still based on the concentration. Clay binds, as mentioned in the previous chapter, strong to heavy metals, meaning that the leachability will be low, also due to clay's low permeability, which could make it ineffective to remediate even though high concentrations are observed.

3.1.1 Development of guideline values and guidance

How the Swedish EPA has developed and worked with regulations and guidance for contaminated soil is essential to understand what progress that has been done, and how. Having stricter guideline values is one way to regulate, and increase focus to more sites, so that more remediation is potentially needed, thus having fewer contaminated sites. This is done by comparing the guideline values for a selection of heavy metals, from 1996 with the latest from 2016, as seen in Table 2 below.

Table 2. Comparison between guideline values (GV) based on 1996 and 2016 for heavy metals (Naturvårdsverket 1996, Naturvårdsverket 2016).

Contaminant	**GV 1996 (KM/MKM) [mg/kg]**	**GV 2016 (KM/MKM) [mg/kg]**	**Difference [%]**
As	15/40	10/25	-33/-38
Cd	0.4/12	0.8/12	+100/0
Co	30/250	15/35	-50/-86
Cr (VI)	5/20	2/10	-60/-50
Cu	100/200	80/200	-20/0
Hg	1/7	0.25/2.5	-75/-64
Ni	35/200	40/120	+14/-40
Pb	80/300	50/400	-38/+33
Zn	350/700	250/500	-29/-29
Average	**-**	**-**	**-21/-30**

As shown, most of the guideline values have decreased with an average of 21% for KM and 30% for MKM, to a lower acceptable concentration. Some of the concentrations for sensitive land use; nickel and cadmium, as well as lead, for less sensitive land use, have however increased, whereas some are unchanged. The concentrations for cobalt and mercury have decreased significantly, this probably due to their toxicity (Naturvårdsverket, 2006).

The fact that the concentration for KM has decreased while the concentration for MKM has increased for some metals is questionable. This since European Food Safety Authority (EFSA) in 2010 lowered the limit for uptake of lead, meaning that it is more toxic than what stated before (EFSA, 2010). Further, it has been investigated that having 150 mg/kg is a more relevant limit, due to accumulation of copper in plants, showing that the present MKM concentration for copper should be decreased from 1996’s levels (Anastassiadou *et al.*, 2020).

An evaluation of the Swedish EPA “handbook” of remediating contaminated areas, from 2009, has been made in 2018 where several aspects were discussed on how they could be improved, based on answers of a survey. The majority of the asked participants were authorities (49%), consultants (24%) and contractors (17%) of a total of circa 100 respondents. 85% used the handbook for guideline values, 73% for risk assessment for contaminated sites and 57% used it for choosing remediation method. Further, it was stated that the handbook is complicated to understand with vague defined formulations. Also, more concrete advice should be given, there is a lack of practical guidance and that the guidance does not apply to remediation projects in collaboration with construction projects. Consequently, stated environmental goals are difficult to reach and especially regarding sustainability where there is a clear lack of advice and little guidance due to little usage of *in-situ* methods. Entrepreneurs found the handbook less useful where 43% of the participants answered that it was not useful, since it is difficult to relate it to construction projects. It was also a broad variety of answers regarding how the handbook is interpreted and applied, where several had different perceptions based on their experience. For example, contractors seem to have stricter rules than authorities and municipalities do. Regarding guideline values, it was found that several participants thought that they are used as limits and that use of site-specific values often was declined, due to lack of knowledge/experience. Most of the participants thought that it was difficult to implement new remediation techniques and that this handbook does not cover the relevant guidance. To conclude, 52% found that they have been in situations where there was a lack of guidance regarding remediation, thus a great improvement can be done. (Naturvårdsverket, 2018)

3.1.2. Standards and guidance for sustainable remediation

What remediation method to use in Sweden, is guided and regulated by ISO 18504, an international standard for sustainable remediation, and Report 5978 from the Swedish EPA, *How to choose remediation method* (Åtgärdsportalen, 2020). ISO 18504 applies for remediation of soil and groundwater and is a short and concise “handbook” for describing the key aspects for achieving sustainability, based on the environmental, economic and social dimensions. Indicators for goals and values (often CO_2-eq), and how they are conducted are also described, to be used for comparison between different remediation alternatives. It also considers the decision-making process, assessments and the role of sustainable remediation, all to stress the importance of sustainability and to guide when choosing remediation method (ISO, 2017; Åtgärdsportalen, 2020).

Report 5978 thoroughly describes different remediation goals and how they should be implemented, as well as their importance in the decision-making process. The general goals should describe the main goal of the remediation, for example what land use the area will have afterwards, and if these goals can/should be based on national, regional or local environmental goals/policies. It is important to have measurable goals, since they are used and are based on the risk assessment. Hence, a combination of ISO 18504 and report 5978, gives a comprehensive guidance on how to choose a sustainable remediation method. (Naturvårdsverket, 2009)

3.2. Life cycle analysis of remediation methods

The Swedish EPA (2008) have conducted a rigorous life cycle analysis (LCA) on different remediation methods as well as the economic costs. It was shown that “dig and dump” emitted two times more CO_2 than excavating and treating on site and roughly three times more than *in-situ* methods. The reason for this is due to the extensive usage of transports that are needed when disposing soil on a landfill. Compared to treating on site where it is only the machines that are excavating the soil that pollutes the most and for *in-situ* it is often the electricity and use of material that emits CO_2. However, it is important to have the correct soil conditions when using *in-situ* remediation, this since the efficiency decreases with less favourable soil conditions and it can be difficult to achieve low concentrations for sensitive land uses. Despite this, it was shown that the choice of remediation method had a larger influence on the environment than the choice of remediation level, that is *e.g.* less sensitive land use or sensitive land use. Hence, having a detailed field investigation before using *in-situ* remediation is of most importance, to prevent unsuccessful remediation. The economic costs of the emissions from the different remediation methods were in-line with the emissions of the remediation methods, where “dig and dump” was three times more expensive than *in-situ* methods. Depending on the type of pollutant, the cost will differ depending on the geographical location. A large city will have higher costs for particle matter, due to larger exposure than for a smaller city, whereas the cost for emitting CO_2 is independent on the location since it affects globally and not regionally. Because of this, “dig and dump” will in general have higher costs due to the higher cost for CO_2 emissions. Regarding total economic costs for the remediation itself, and not its emissions, it was shown that “dig and dump” was 50% more expensive than both remediation on site and *in-situ* methods, where *in-situ* was found to be cheapest. Hence, *in-situ* methods are favourable in all aspects. (Naturvårdsverket, 2008)

3.3. Reuse of material

Having a circular economy in the society, to re-cycle and re-use as much as possible of the material, is something that is achieved more every year, and is expected to be the norm in 2030 according to the Swedish environment institute (IVL, 2019). Having material in landfills or at other locations where the material is waste, are at the bottom of the scale since it cannot be re-used, indicating that this type of material-use must be improved to be re-used or recycled (IVL, 2019). This is crucial since the construction waste generated by 28 EU-countries on its own contributes to 33% of all waste produced in the EU (Delvoie *et al.*, 2019). Delvoie *et al.* (2019) have studied the drivers and barriers for recycling waste in Belgium, France, Germany, Luxemburg and Netherlands, and have studied the total construction waste as well as sand and aggregates. Excavated soil and stones contributes to over 50% of the total sand and aggregates waste, making it the most important parameter in that category (Delvoie *et al.*, 2019). However, due to the lack of a clear definition of what types of soil and waste that are included in the different categories, whether if it is excavated soil, soil treated on site or others, it is difficult to summarise all waste without making misconceptions (Delvoie *et al.*, 2019).

According to Magnusson *et al.* (2015), the cost of handling soil waste can account for up to 30% of the total cost of a construction project, making it not only a question about the environment but also economics. For the soil to be reusable, it must fulfil the geotechnical and hydrogeological properties but also the rules of contaminated soil, as explained above, which could be problematic. It was shown that if almost half of the excavated soil were reused (in this case 700 000 m^3), it would save up to 4000 tonnes of CO_2 and almost 1.8 million US dollars (Eras *et al.*, 2013). In other projects as much as 85% of both the environmental and economic costs were reduced, due to material reuse (Chittoori *et al.*, 2012). Further, Magnusson *et al.* (2015) proposed that soil masses can be exchanged between projects if the excavated soil is not suitable at the actual project but in a project nearby. This is also the case when landfills are getting full. Moreover, this has been done in England where excavated soil was placed temporarily on site and transported to other projects, which resulted in a 30% cost saving and 100 tonnes saved CO_2 (CL: AIRE, 2013).

Excavated soil can also be reused as construction materials, such as clay in cement mortar, as stated by (Priyadharshini *et al.*, 2018). Instead of using river sand, that is a depleted material, excavated clay was used and mixed with binders such as lime and calcium, to improve cementitious properties. The environmental or economic impacts were not investigated but could be assumed to be improved. Further, it was shown that low plastic soil, the excavated, required less additives than the more plastic soils, *i.e.* a more efficient material to use, and that it showed to have the same properties as cement mortar based on fine sand. (Priyadharshini *et al.*, 2018).

Vaccari *et al.* (2012) have investigated the possibility to reuse slag from soil washing treatment. In Italy, where the authors lives, the slug landfill can contribute up to half of the costs in a soil treating plant (Piepoli *et al.*, 2010). It was shown that the slug could be used for stabilising soil as reinforcement, brick production and for cement. Making waste from soil treatment even more recyclable (Vaccari *et al.*, 2012).

As mentioned earlier, Sweden has many landfills and to ensure that these are more easily recycled, the government has withdrawn the previous waste tax on landfills (Naturvårdsverket, 2015). Also, by increasing the tax on depositing waste, it is more expensive to use landfills but less expensive to treat the material (Regeringen, 2020). These economic effects have generated no positive trend with less material deposited on landfills, this since the price has been halved over the last 20 years and is still significantly low (Aktuell Hållbarhet, 2016). Moreover, some environmental “experts” claim that it is better to store contaminated soil masses until there are better remediation techniques, if they pose no risk (Aktuell Hållbarhet, 2016).

Further, due to little knowledge of the number and size of depleted landfills, it is difficult to estimate the potential amount of recycled material. Also, most of the regional landfills are too small for being profitable, which in combination with the last mentioned and technical challenges, makes it difficult to recycle landfills. Larger landfills with more homogenous material could be more efficiently remediated, if the material achieved is of sufficient quality and not more costly than new material. Lastly, according to the Swedish EPA, there are no potential benefits for promoting landfill recycling due to the vast lack of knowledge. Also, it is stated that even if all landfills were to be recycled, it would have little to no impact on the usage of “new” material. (Naturvårdsverket, 2015)

3.4. Methods to improve sustainability

3.4.1. SCORE

To ensure that the environmental goals regarding remediation are fulfilled, that is a non-toxic environment and a good built environment, a project called SAFIRE (Sustainability Assessments For Improved Remediation Efficiency) has been conducted (Naturvårdsverket 2020c; Volchko *et al.*, 2016). By the name, the project aims for increasing the sustainable efficiency of remediation, this since as mentioned earlier, that it is often costly and difficult in several ways to implement sustainable remediation (Volchko *et al.*, 2016). Rosén *et al.* (2015) have developed SCORE, a user-friendly method that consider multiple parameters (economic, environmental and social) to achieve a fully sustainable method. By weighing different parameters to each other, depending on situation, the result will show how much impact the different remediation alternatives have to different parameters (Volchko *et al.*, 2016). Volchko *et al.* (2016) have investigated a known contaminated site in Sweden (Teckomatorp) and evaluated the remediation alternatives with the SCORE-method. This to evaluate how the social, economic and environmental parameters differs between remediation alternatives and if more sustainable remediation alternatives are recommended instead of the conventional "dig and dump". Figure 14 shows how the SCORE-method works and the required input parameters.

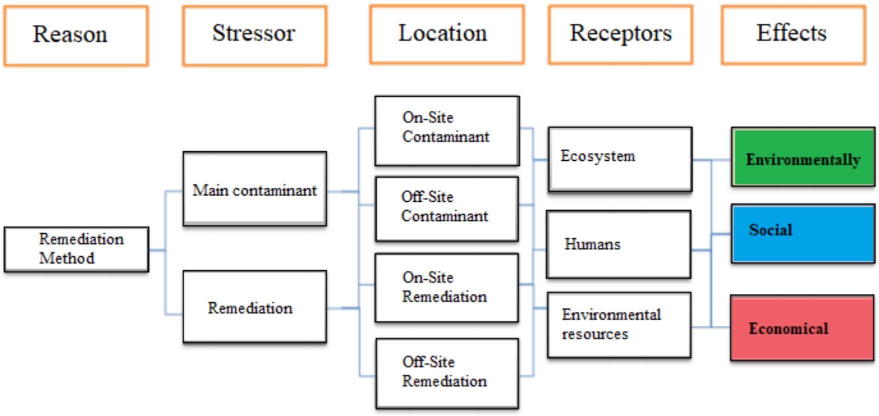


Figure 14. SCORE method based on Rosén et al. (2015) as described of Volchko et al. (2016).

The study discussed remediation alternatives that included *in-situ* methods, *ex-situ* or disposal at landfill. Due to the contaminants, *in-situ* methods were scoring low in environmental effects, but also economic since the house prices would benefit more from having the soil excavated than isolated. Also, the fuel for treating the soil *ex-situ* by thermal induction, proved to be crucial if it was more environmentally sustainable than disposal at landfill. However, this depends on how the different parameters are weighted to each other, and which one is the most important, and this is easily achieved in SCORE. (Volchko *et al.*, 2016)

3.4.2. Gentle remediation options

Gentle Remediation Options (GRO) refers to a type of remediation methods that have little impact on the environment and economic, hence methods such as enhanced stabilisation as mentioned in 2.2.1 (Cundy *et al.,* 2013). The reason behind GROs is to make the risk management strategies easier for stakeholders and is a result of collaboration of researchers in the EU by developing guidelines and tools for decisionmakers, to facilitate work with sustainable remediation (CORDIS, 2016). The essence with GROs is that the soil should still be useable while remediating and that the soil "remediates" itself, not that it is dug up and disposed, but to utilise sustainable chemical and biological processes (Cundy *et al.*, 2013). This has led to discussion since remediation during construction is an expensive process since time is money, compared to remediating an old site on the countryside (Cundy *et al.,* 2013). Further, Cundy *et al.* (2013) states that having a generic reference value for concentration of the contaminant is not a sustainable reference guide. This since it does not consider site specific conditions such as source and pathways, hence the actual risk for having a certain concentration of a contaminant in the soil (NICOLE, 2012; Bardos *et al.*, 2011). On the other hand, having generic target values are simpler to implement since the soil often is excavated but it is the actual risk of spreading (leachability) that is relevant to if the site poses a risk or not (Cundy *et al.,* 2013). It is also important to stress that sustainable remediation can pose a risk to the site on its own using chemicals and other resources (CL: AIRE, 2011). The applicability of GRO can be evaluated by the following questions in Table 3 below.

Table 3. Questions to be answered if GRO are suitable or not. If Yes, GRO are advantageous, if No, GRO are not advantageous. (Cundy et al., 2015)

Question (All questions answered "Yes")	Suitability
Is time of essence, that is, if a fast remediation is needed?	No, processes are too slow.
Is the "performance" of the remediation based on soil concentration values?	No, this is not beneficial for GRO and conventional methods such as "dig and dump" are more suitable.
Are there buildings or other permanent usage, covering the soil on the site?	No, soil cannot be remediated using GRO, only by methods such as electrokinetics or else that does not require access to the soil.
Is the soil currently used by biological activity?	Yes, this is suitable for GRO.
Is it a large area not currently in use?	Yes, this is very suitable and beneficial for GRO since other methods are likely to be more costly.
Are contaminants placed shallow in the ground?	Yes
Is the future land use of recreational type?	Yes, soil can be treated without damaging potential vegetation etc.

3.5. Sustainable remediation today

3.5.1. Interest of sustainable remediation

In 2014, about 2.5 million sites were stated to be contaminated in Europe, with a remediation cost of over five billion euros, based on 2004 EU market value (Van Liedekerke *et al.*, 2014; Ernst and Young, 2006). How different countries are dealing with remediation, their development and their future approaches, were studied by Bardos (2014) and Rizzo *et al.* (2016). There is a vast interest at stakeholders to improving the sustainability and have a good definition of what it means (Bardos *et al.*, 2013). Also, as mentioned in the previous chapter, it is important to consider all necessary aspects such as economic and social factors and not only the environmental risk. This by using networks such as: Sustainable Remediation Forum (SURF), Contaminated Land: Applications in Real Environments (CL:AIRE) and Network for Industrially Co-ordinated Sustainable Land Management in Europe (NICOLE) (Bardos, 2014). These networks and forums are working thoroughly with improving the management around sustainability, this through frameworks and reference guides (Bados, 2014). Compared to the past, when only the human health and the environment were considered and little decision making were done, today there are several management networks and regulations to improve the decision making where SCORE is a good example (Bardos, 2014).

The understanding of sustainable remediation is widely spread across nations and stakeholders. This understanding takes place in definitions, principles and decision management, where the driving factor is to prevent risks for human health and the environment in total. Further, the final goal is to interpret a sustainable remediation method, where the regulations and policies are the divergent factor between the countries, that could interfere with the end-result. Also, how much care that should be taken for the stakeholders' opinions are different between countries, where some think it is an important aspect while others want a stricter management top-down. (Rizzo *et al.*, 2016)

Hou *et al.* (2016) have investigated how the trend regarding studies about sustainable remediation have elapsed over the past decades. This was done by analysing the progress of number of publications on the subject and how different countries manage sustainable remediation USA, UK, China, India, Japan, South Korea (referred to as "Asia"), France, Germany, Israel, Ireland, Italy, Netherlands, Sweden, Belgium, Czech Republic, Denmark, Canada, Australia (referred to as "developed countries") and lastly Brazil, Mexico, Nigeria and South Africa (referred to as "developing countries") (Hou *et al.*, 2016). This was done by surveys about the effectiveness in adopting sustainable remediation strategies, the importance to adopt sustainable remediation strategies and if there are barriers preventing sustainable remediation, on a scale of 1 to 5. As shown below in Figure 15, there is a clear trend that USA, UK and the developed countries are in the lead and more aware of sustainable remediation. Also, it is noticeable that in

Asia, the stakeholders think they are better than they are, which could have negative effects on future development. (Hou *et al.*, 2016)

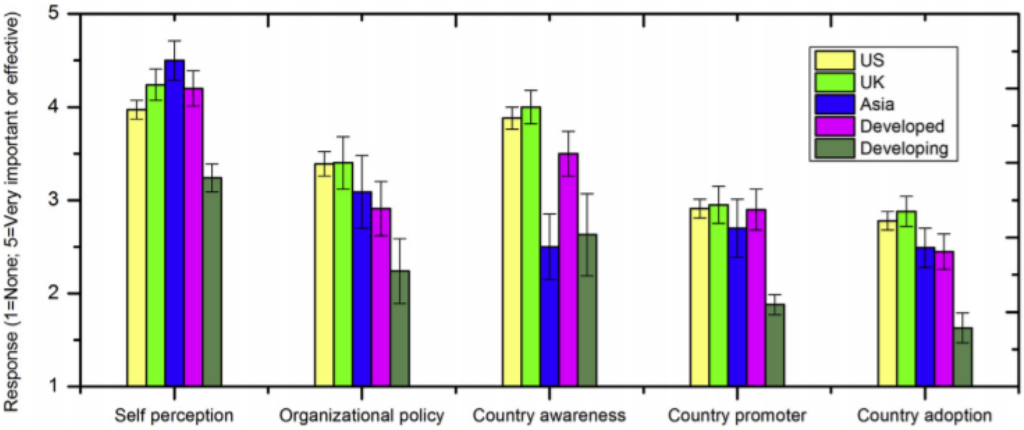


Figure 15. Results based on the survey of Hou et al. (2016).

The number of studies conducted on a subject often corresponds to its relevance. As can be seen in Figure 16 the studies regarding sustainable remediation presented on Google Scholar have increased significantly over the last decade. Showing that more research has been done, and most likely an increase in awareness and adoption. However, it was shown that even though there is a vast interest and knowledge regarding remediation sustainability, it was found that the "professionals" have little impact on the society. Therefore, it is important to have instruments rather than campaigns that enhances the understanding of the impacts of sustainable remediation for the public. It is also vital to have promoting forces that pushes the development in the right direction and have less barriers, allowing the development to take place. (Hou *et al.*, 2016)

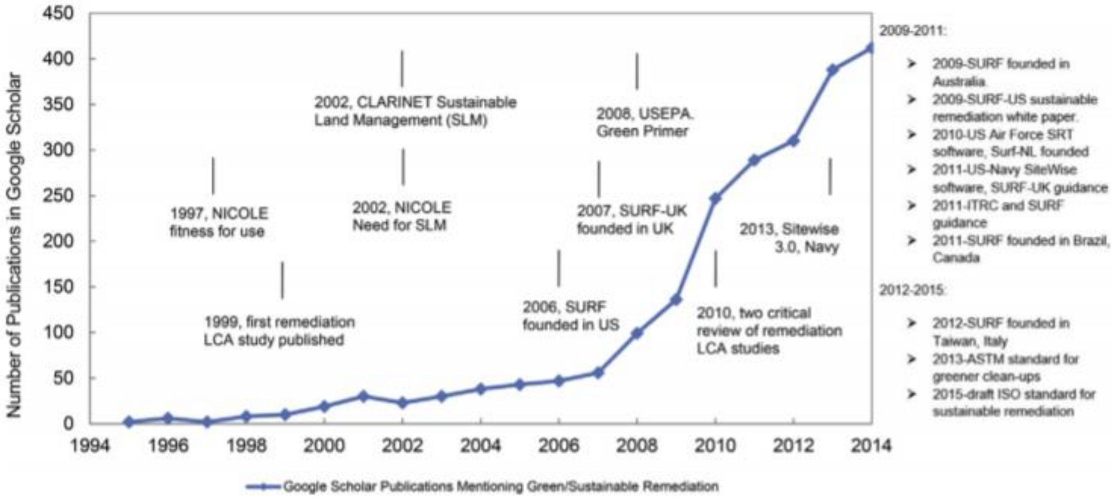


Figure 16. Number of studies regarding sustainable remediation presented on Google Scholar over the last two decades. Note the dates for when different networks and managements were established. (Hou et al., 2016)

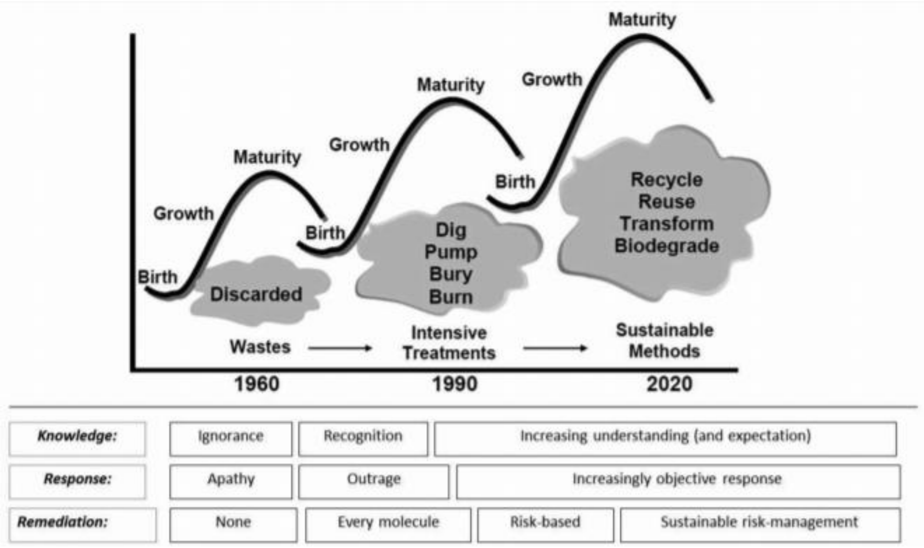
Favara *et al.* (2019) have too studied the progress of achieving sustainable remediation over the last decade. There have been a great development in different frameworks, literature and guidelines, in-line with Hou *et al.* (2016). However, the challenge today is to implement a broader view on the sustainable remediation, including economic and social factors, in accordance with Rosén *et al.* (2015) and their SCORE-method. Also, Favara *et al.* (2019) states that many stakeholders and remediation practitioners have a false and incorrect perception about sustainable remediation, leading to worrying end results. This is somewhat proven by Hou *et al.* (2016) where the self-perception can be misleading to the end-result, and create a complicated situation, when comparing to the country's level of awareness.

3.5.2. Development of remediation techniques

Gong *et al.* (2018) have conducted a comprehensive study regarding the development of heavy metal remediation over the last decade, discussing improving techniques and how the decision-making process has developed. As mentioned earlier there are several different options when choosing remediation method and this depends on many factors such as soil properties, land use, economy and site conditions. However, there are many sustainable remediation techniques that are beneficial for all aspects (economically, social and environmentally). In summary, this can be said about the development and what should be done:

- More focus has been put into the contaminant's leachability rather than the total concentration, since this is the part that can pose a risk for the surroundings. It is also more economic and practically easier. It should be noted that the leachability could be difficult to monitor due to potential reactions in the soil. Also, this could be difficult to control since the environmental impacts of the remediation is sometimes "forgotten" where the soil should just be dug up.
- Stabilisation processes have been successful since it encapsulates the contaminants which is beneficial in almost all aspects. However, the long-term remediation could be questionable since there is little knowledge whether the processes are irreversible or not. The lack of knowledge is a result of having bad monitoring data over longer time periods.
- Having different types of contaminants in the soil can lead to new potential reactions between the remediated contaminants and the other chemicals in the soil. This was not well considered in the past which can have future consequences.
- Due to impossibilities or difficulties to excavate soil, better remediation methods are needed to remediate the soil while keeping the land above undisturbed from its daily usage.

Below, the progress of sustainable remediation is summarised in a figure, with the aspects described above (see Figure 17), where different countries are on different locations on the "timeline" today (Smith, 2019).



Knowledge:	Ignorance	Recognition	Increasing understanding (and expectation)	
Response:	Apathy	Outrage	Increasingly objective response	
Remediation:	None	Every molecule	Risk-based	Sustainable risk-management

Figure 17. The progress of sustainable remediation summarised by Smith (2019) note that the last cycle is "recycled", thus no birth of material.

Still, as shown in Figure 12 in Chapter 3, there is a large spread between countries in what remediation method that is used, indicating that some countries are ahead in the development. It is also interesting to see that it is either almost 100% remediation by excavation, as for UK, or it is the other way around, almost 100% *in-situ* technologies as for Malta and the Netherlands (EEA, 2017). This is most likely due to regulations and how implemented new sustainable technologies are in the construction sector, as will be mentioned below.

3.5.3. Construction Companies' Sustainability Goals and Progress

How construction companies are dealing with sustainability has improved significantly over the last decade. This is evident when comparing studies done on sustainability management in the construction sector from 2005 to 2020, by Myers (2005), Afzal *et al.* (2017) and Goel *et al.*, (2019). See Figure 18 for how the studies on sustainable management in the construction sector have developed over the last two decades, with the same trend as Hou *et al.* (2016) in Figure 16. Previously, it was only the economic aspects that were considered when budgeting on new projects, but today almost all economic costs have a corresponding environmental cost. For example, by having simplified models based on guidelines from Swedish Environmental Institute, an easy overview is achieved when comparing

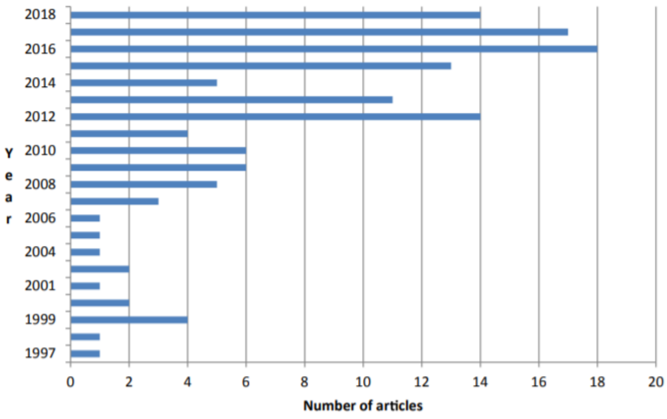


Figure 18. Number of articles concerning sustainable management in the construction sector over the last two decades (Goel et al., 2019).

different alternatives. It can then be visualised what parameters that emits the most and take the right decisions.

Regarding environmental goals, most of the large construction companies in Sweden aims for being carbon neutral in 2045. Where Skanska is one of the most sustainable companies in the construction sector in Sweden (Skanska, 2019). When comparing to construction giants in Europe (Vinci), USA (Bechtel) and China (China State Construction), sustainability management is not as prominent. This by comparing their sustainability goals, where firstly, they were difficult to find, and secondly, they were diffuse and not as defined as the ones in Sweden. For example, Bechtel did not have any clearly defined goals about *e.g.* carbon neutrality or emissions, and Vinci have a few statements almost in-line with Swedish companies (Bechtel, 2021; Vinci, n.d.). This shows the current divergence in construction companies' sustainability management and how they want to be recognised. So, with this in consideration, there are still room for improvement.

Afzal *et al.* (2017) have investigated this further by comparing the "sustainability performance" for the largest construction companies in the world according to Engineering New Record. This by reviewing their approaches to sustainability, their sustainability reports and their general sustainability performance. In summary, it was six categories that were compared which gave one point each, thus six points maximum. It was evident that Asia was significantly worser than Europe and America, where most of the European construction companies had the best results, as can be seen in Figure 19. Of the top 10 largest companies in the world (6 Chinese, 1 American, 1 Brazilian, 1 French and 1 Spanish), only one scored almost max with 5/6 points, whereas seven scored 0/6. In total, no company scored 6/6 and Europe had roughly five times the average score than China, and more than twice than America (USA and Brazil), with an average at 63% of the companies scoring one point in each category. This shows that most, almost all, of the largest construction companies have poor to non-existing sustainability performance, indicating vast room for improvements.

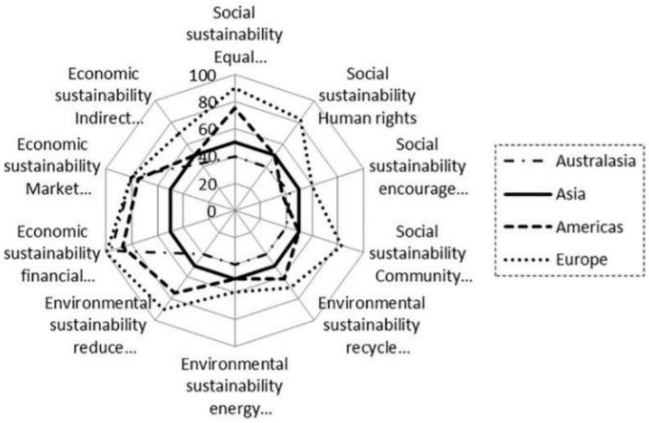


Figure 19. Results based on Afzal et al. (2017) when comparing the sustainability performance at construction companies.

As an example, Skanska's work with sustainability is extensive, where all projects are classified in five different levels, based on energy, climate, material usage and disposal and water usage. The lowest level represents the environmental regulations and responsibilities, and the highest level represents a carbon neutral project, meaning that most of Skanska have stricter regulated projects than the authorities require. The sustainability management also includes the actual site office, which has its own classification, all to improve the sustainability. However, due to Swedish regulations during state-funded construction, where the offer with the lowest price must be chosen, this could be problematic since there is no valuation of the sustainability of the projects but only valuation in terms of money. Also, there are no demand or request to utilize sustainable remediation techniques, but only that the concentrations are below the guideline values. Indicating that the usage of new *in-situ* methods is not promoted or valuated by the authorities, the effect is almost the opposite, if it results in higher costs. Further, having a more sustainable management than the regulation requires is optional, making it difficult to implement in some scenarios. This is a good example, showing that a company can have a comprehensive sustainability management but is somewhat restrained by the governing authorities, regulations and laws.

3.5.4. Barriers to achieving sustainable construction projects

Opoku *et al.* (2019) have investigated potential barriers for achieving higher sustainability in construction projects, based on professionals, consultants and contractors in Finland, UK and Ghana. The result showed that there were three types of barriers, either it is due to knowledge: lack of technology/demand/regulations/public awareness, due to financial: over all costs, risk of investment and general higher costs, or it is due to other aspects such as attitude of professionals, measuring tools for building sustainability, resistance to change etc. In poorer countries the lack of technology/demand/knowledge is more common barriers whereas in richer countries the barriers are sometimes more due to financial, but mainly due to regulations and measuring tools to ensure sustainability.

Sourani and Sohail (2011) have also studied barriers, and then by public clients aiming for more sustainable construction projects in the UK. This was done by interviewing sustainable professionals and experts. It was found that the main barriers were due to regulations, lack of awareness/understanding, insufficient guiding/tools, diffuse and misinterpretations of definitions of sustainability, and financial barriers such as funding and long-term consequences. The main responsibility for solving this problem and removing these barriers were held at the government (changing regulations and making sustainability more economic beneficial) and professionals (educating the users and supplying tools/guidance to make it more easily implemented).

In Sweden, the Swedish Geotechnical Institute (SGI) conducts surveys about the need for knowledge and factors that prevents use of more efficient remediation. These surveys were answered by authorities, construction companies, consultants and problem owners and the results are summarised in Table 4. (SGI, 2020)

Table 4. Results from surveys based on barriers and development in more sustainable remediation, based on consultants, construction companies, authorities and other stakeholders. (SGI, 2020)

Year	Result
2010	Great lack of other remediation methods than “dig and dump”. - Lack of technology and tests. - Lack of knowledge and education. - Lack of guidance, risk assessment tools.
2011	More focus about ecological aspects - 25% have used ecotoxicological methods or are aware of their existence. - The use of resources and environmental impact should be more considered. - Still a large lack of knowledge regarding remediation techniques and risk assessment methods.
2013	Contaminated sediment was in focus. - Need more knowledge about contaminated areas in general but especially regarding sediments. - Lack of guidance adapted for the Swedish environment. - A need for alternative more sustainable remediation methods.
2014	How contaminated areas were dealt with in the planning process. - Lack of knowledge and communication about contaminants in the area can result in delays and high costs for the project. - Lack of knowledge results in lack of time and hence a bad remediation is often chosen (“dig and dump”).
2018	The spread of contaminants from different areas and the stress on recipients. - Still a great need for more knowledge. - Need for more user-friendly guidance as well as more advance guidance. - Need for more complex models that accounts for more exposure paths. - More involvement of water treatment sector is needed since contaminants spread with water.

Still in 2018, there is a need for several essential things such as knowledge and guidance, that prevents the development of sustainable remediation.

3.5.5. Remediation projects in Sweden

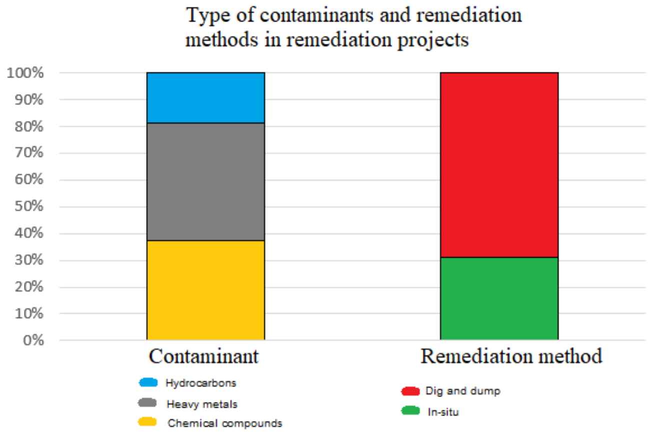


Figure 20. Type of contaminant and remediation method used for remediating old industries in Sweden, from 2010 and 2017. (SGU, 2020a)

Many of the present and finished remediation projects in Sweden due to contamination from industries operated by the government, are managed by SGU (Sweden's Geological Investigation) together with the Swedish EPA (SGU, 2019). Figure 20 summarises the most common contaminants and remediation methods for sites that have been remediated between 2010 and 2017. It is a great lack of data since many of the current project are not finished or that the choice of remediation method has not yet been decided. The data is based on 16 of a total of 61 remediation projects from 2010 to 2017 from large industries (SGU, 2020a). Here it is clearly visualised that few sustainable remediation methods were used, representing only 31%, where 69% of the remediation were done by excavating the soil. Also, *in-situ* methods were only used when treating soil contaminated with hydrocarbons and chemical compounds, hence the largest part of the contaminants, the metals, were all removed by "dig and dump". However, new technologies will be used/tested in almost a third of the planned remediation projects, where half of these are concerning remediation of heavy metals (Naturvårdsverket, 2020d). What remediation methods that should be used for the other projects are not yet decided, hence no data can be gathered.

The data above can be compared with 226 finished remediation projects between 1994 and 2005, where 62% of the contaminants were heavy metals and the other 35% where oil. 88% of these projects were remediated by "dig and dump" and 10% of the projects were remediated *in-situ*. Thus indicating little progress of over almost two decades, compared to the data in Figure 20. (Naturvårdsverket, 2006b)

To achieve better knowledge about sustainable remediation, the Swedish EPA has in 2019, together with SGU, SGI and municipalities in Sweden, started a research project "Collaboration for innovation". The goal with this is to create a handbook, to help for example construction companies to choose a more sustainable method, but also identify problems when implementing new techniques. This is done by testing the applicability of new remediation techniques and spread the information and knowledge about working with these methods. All to decrease the usage of "dig and dump" and increase the usage of *in-situ* methods. (SGU, 2020b; EBH-portalen, 2020)

3.5.6. Contaminants in and around Stockholm and Gothenburg

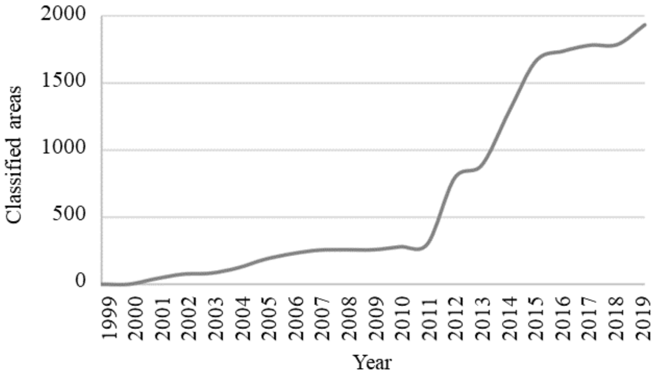


Figure 21. Risk-classified areas in Stockholm (Länsstyrelsen Stockholm, 2019).

Stockholm and Gothenburg are the two largest cities in Sweden, posing a large urbanization with many big construction projects as a result. The gathering of data on the number of contaminated sites has evolved rapidly the last decade, similarly to the general progress of sustainable management, as mentioned and displayed in Figure 16 and Figure 18. This progress is presented in Figure 21 and since there is a lack of knowledge about how many risk-classified areas there are, the number of sites will probably continue to increase from today’s 1933 contaminated areas in Stockholm alone. In Gothenburg there are 545 risk-classified areas, which of many are in the inner-city, the same for Stockholm. Thus, indicating that potential residential areas are planned to be built on contaminated soil, which requires vast amount of remediation, see Figure 22. (Länsstyrelsen Stockholm, 2019; Länsstyrelsen Göteborg, 2020)

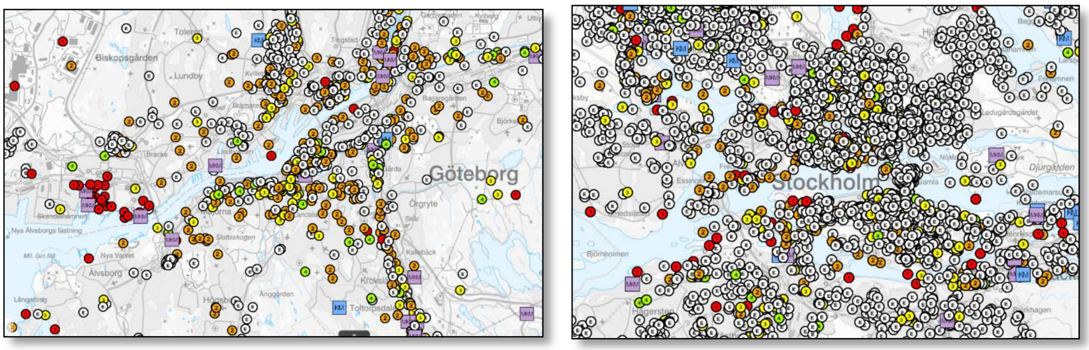


Figure 22. Classified contaminated areas (marked “1-4”, coloured, 1 is highest risk) and potential contaminated areas (marked “E”, white), in Gothenburg (left) and Stockholm (right) (EBH-kartan, 2020).

4. Methodology

This chapter describes the methods and strategies that were utilized in this study.

The study is divided into two parts. The first part concerns how to improve sustainable remediation methods in clay, conducted by laboratory testing and a literature study. The second part concerns how construction companies are dealing with remediation, if it is user-friendly or if remediation is difficult to improve due to regulations. This was conducted by a literature study of how remediation could be more sustainable and if laws affect future progress. Also, by a questionnaire to investigate site-managers' and governing authorities' opinions about remediation and if it could be improved.

4.1. Experimental methods

Two mixtures of binders were tested: 10% SiO_2 with 5% $Ca(OH)_2$ and 10% SiO_2 with 10% $Ca(OH)_2$. The two binder-clay mixtures were examined with and without copper contamination. 5% $Cu(NO_3)_2$ was added in the contaminated samples. All mass percentages are calculated on the dry mass of the clay. Further, the curing time was 7 days and 14 days.

4.1.1. Material

Kaolin samples were reconstituted by mixing of distilled water with pure kaolinite powder from Acros Organics (art. nr. 211740025, CAS nr. 1332-58-7). It has the following properties, as stated in Table 5.

Table 5. Properties of the kaolin clay.

Particle Size [µm]	**Specific Gravity**	**Plastic Limit [%]**	**Liquid Limit [%]**	**Plasticity Index [%]**
1-18	2.61	24.5	52.5	28

Natural clay was used for eight consolidated samples, it was obtained from Kärra test site, north of Gothenburg, further referred to as "Gothenburg clay". Deposits in the area typically consist of illite and smectite. Clay from 4 m and 10 m were mixed, their properties are presented in Table 6.

Table 6. Properties of the Gothenburg clay.

Depth [m]	**Particle Size [µm]**	**Wt [%]**	**Organic content [%]**	**Bulk Density [g/cm^3]**	**C_u [kPa]**	**Sensitivity**	**LL**	**PL**
4	1-18	88.8	4.5	1.53	10.4	11.5	80	25
10	1-63	70	6.3	1.61	23	9.7	-	-

The particle size distributions for the two clays are shown in Figure 23.

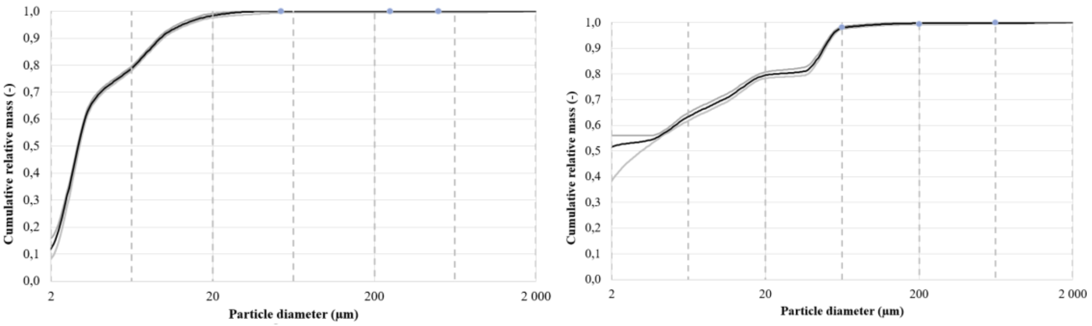


Figure 23. Particle size distribution of kaolin clay (left) and Gothenburg clay (right).

The choice of concentration of the copper(II) nitrate was chosen according to Table 7 and is based on the literature study. Based on this, the clay was contaminated using 50 g copper(II) nitrate for 1000 g dry soil, (*i.e.* concentration of 5%). At depleted industries, the concentration of pure copper can be as high as 23 g per kg soil, with an average of 1.5 g/kg. One third of copper(II) nitrate is pure copper, hence 50 g corresponds to about 16 g copper, thus a realistic concentration that can be found in contaminated soil. Further, 50 g copper(II) nitrate per one kg dry soil corresponds to roughly 23 g pure copper per kg wet soil, and thus represent a maximum concentration that can be found in field. (Norrman, 2020)

The copper(II) nitrate used was from Scharlau (art. nr. CO0098, CAS nr. 10031-43-3).

Copper(II) nitrate is more stable than copper sulphate why it was used, this since it is more inert and thus avoiding unwanted reactions to happen and to interfere with the pozzolanic reaction (Du *et al.,* 2014; Wang *et al.,* 2020). Kaolin clay was used since it is more applicable in academic reports since it is available for everyone. Further, it consists mainly of silicate and alumina which is pozzolanic, making it suitable for this study (Mitchell & Soga, 2005). Natural clay was also used to since it is applicable in field. Calcium hydroxide, $Ca(OH)_2$, from VMR Chemicals (art. nr. 22355.298, CAS nr. 1305-62-0), was used as binder and silica(IV) oxide, SiO_2, from Alfa Aesar (art. nr. 12727, CAS nr. 7631-86-9) was used as pozzolanic material. These additives were used since to achieve as pure pozzolanic material as possible, to prevent unwanted reactions and to get a purer pozzolanic reaction.

Table 7. Summary of spiking properties, based on the literature study.

Chemical used	Spiking concentration	Sample preparation	Reference
Copper (II) sulphate	200mg/kg dry weight	Consolidated to near saturated condition.	*Pazos et al. (2008)*
Copper (II) sulphate	10-25% of dry weight	Soaked and stored for 30 days, wrapped in plastic.	*Karkush et al. (2013)*
Copper (II) sulphate	100mg/kg	Stored 24 hours.	*Torres & Varennes (1998)*
Copper (II) nitrate	1000-5000 ppm	Not specified.	*Muththalib & Baudet (2019)*
Not specified	100mg/l	Not specified.	*Mu'azu et al. (2015)*
Copper (II) nitrate	100mL of 5000mg/l for 1kg soil	4 weeks.	*Wuana et al. (2010)*
Copper (II) nitrate	600mg/L (says to be high)	Not specified.	*Sipos (2010)*
Copper (II) nitrate	125-1000mg/kg	Added over 5 days, stored in plastic container.	*Schwertfeger & Hendershot (2013)*
Copper (II) nitrate	0.1-10% of dry weight	Cured 2 hours, stored in plastic sealed PVC tube for 1/2/4/14 weeks,	*Saeed et al. (2014)*
No chemical used	No chemical used	Stored 24 hours, stored for 7-90 days in vinyl bag.	*Horpibulsuk et al. (2013)*

Horpibulsuk *et al. (2013)* and Latifi *et al.* (2018) (presented in section 2.2-2.4), showed that the optimum content for fly ash, as a binder and CCR, as an additive, is 15% and 10% of dry soil, respectively. Since the used additives were more concentrated than the ones mentioned above, lower concentrations were used. Fly ash consists of roughly 50% silica, so half of the content was used and CCR consists of 80-90% calcium hydroxide, so the content stated in the literature was used. Hence 10% of silica fume and 5% and 10% of calcium hydroxide was used.

Storing specimen under pressure have proven to yield different result than storing without (Taiyab *et al.*, 2012). In general, soil *in-situ* have an overburden pressure, storing samples without an applicated pressure might cause misleading results. Taiyab *et al.* (2012) stored clay samples with different contents of cement (0-7%) and different overburden pressures (0-50 kPa) for 28 days. It was shown that the overburden pressure increases the strength by 25% and where the increase was more noticeable for the higher cement content. However, due to the vast increase of strength when mixing the soil with cement, storing the samples under pressure could seem irrelevant (Taiyab *et al.*, 2012).

Further, having an overburden pressure could affect the curing process since the pozzolanic reaction could go faster. This due to an increased flow in the soil, hence less time required for the chemicals to react and better contact between the grains due to compression.

4.1.2. Sample preparation

For tests on Kaolin specimens, Kaolinite powder and deionized, deaired water were mixed to an 80% water content, the slurry was then mixed for ten minutes under vacuum to achieve a homogenised mixture. Further, the slurry was carefully sealed and left to hydrate for 24 hours in a temperature-controlled humid storage room. The same procedure was followed to prepare the contaminated samples, but the copper(II) nitrate was dissolved in a 1:1 ratio of deionized deaired water, increasing the water content to 82%. The actual water content after mixing and after each test was measured according to ISO 16586:2003. After hydration, the additives were added and mixed for 10 minutes to achieve a homogeneous mixture. Later, the mixture was put into teflon tubes of 38 mm diameter and 76 mm height and oedometer rings of 20x50 mm. A thin layer of silicon grease was applied on the tube walls to help in easy extraction. All samples were weighted before testing, to evaluate potential changes in density after curing.

Furthermore, the soil was retained in the moulds by the use of porous discs or filter papers at the ends, to prevent material loss before the hardening process. They were covered with a wet cloth and put into a box with shallow water, as can be seen in Figure 24. These samples will further be referred to as “boxed” samples. Also, since the reaction consumes water from the soil, it can potentially dry it out which results in a brittle cement, which was why 100 mL of deionized deaired water was added per sample every week. To be able to evaluate effect of curing age, samples were stored in this state for 7 and 14 days in constant room conditions of 70% relative humidity and 7°C temperature, representative to the conditions in the ground. Compared to other studies, where the samples are almost always cured wrapped in plastic film at room temperature, the curing conditions do not correspond to the soil environment in reality. Furthermore, to evaluate the influence of overburden pressure and consolidation, eight samples were consolidated to an effective stress of 50 kPa (corresponding to an average effective stress at shallow depths of 5 to 10 m) and cured for 7 and 14 days, see Figure 27. In addition, as shown in Figure 26, four samples were also stored fully submerged, without load application, to evaluate the effect of hydration process.

In more detail, both the hydrated pure and contaminated kaolin slurries had good workability. When mixing the uncontaminated clay with calcium hydroxide and silicon oxide, its viscosity increased considerably with a noticeable lower water content and less workability. The uncontaminated clay had also noticeable lumps in the slurry, especially for the samples with 10% calcium hydroxide. Further, when mixing the contaminated clay, the initial change in stiffness and strength were not as significant and it took a longer time to achieve a stiffer clay.

Two reference samples, one contaminated and one uncontaminated, were stored in 40 mm tubes and were consolidated to an overburden pressure of 50 kPa before testing.

Figure 24. Storage of uncontaminated (left) and contaminated (right) samples, normally covered in the wet cloth in the picture.

The extraction of samples was done by placing the tubes in a modified caulk gun that pressed the soil out of the tubes by using a 38 mm metal disk. The extraction process is shown in Figure 25 and had little impact on the samples, compared to pushing by hand.

Figure 25. Extraction process of a sample, note the metal disk pushing out the sample in the picture to the right.

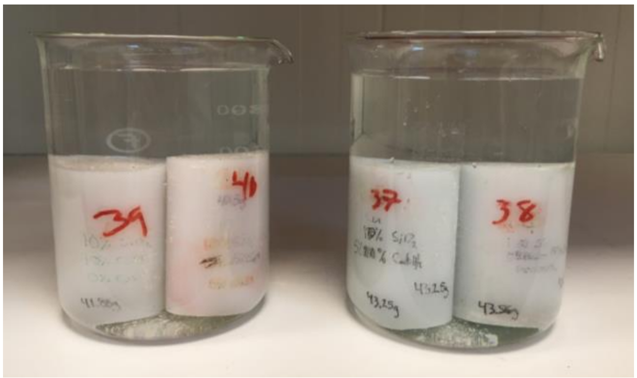


Figure 26. Evaluating the influence of water content during curing by submerging samples fully.

Figure 27. Samples prepared by having a weight of 6 kg, corresponding to 50 kPa at an estimate soil depth of 3-4 meters. Note that the samples were not wrapped in plastic.

For investigating the applicability in field, a natural clay from Angered, located northwest of Gothenburg in Sweden, was used. This clay contained several soil types and is a common soil type found in the Gothenburg region. This clay was prepared similar to the kaolin clay but was remoulded by mixing for 10 minutes to ensure sufficient mixing quality with the additives. Moreover, based on results from the kaolin clay, all samples were cured in the consolidation-rig under 50 kPa for 7 and 14 days. All samples had the same concentration of the additives, 10% $Ca(OH)_2$ and 10% SiO_2. The two reference tests were prepared similar to the kaolin reference tests.

The pH was measured with a soil pH tester (Hanna HI981030) before mixing, right after mixing, after 7 days and after 40 days. This according to the literature, where the pH starts to decrease when the reaction has stabilised. Since the copper(II) nitrate originates from nitric acid, which is a strong acid, it could reduce the pH significantly leading to weaker binding of the additives.

Table 8 summarises the type of samples, the number of samples and the used storing method for all tested samples. For full details of the tested samples, see Table 9.

Table 8. Summary of tested samples and their storing method.

Clay	Storing method	#Samples
Kaolin	Boxed	30
Kaolin	Submerged	4
Kaolin	Consolidated	8
Gothenburg	Consolidated	8

*Table 9. Layout of the tested specimens. If contaminated (Y, cont.), the concentration of copper(II) nitrate was always constant at 5% of the dry clay. Note that the oedometer samples were tested from the "Curing started" date to the "Tested" date. * = these samples were submerged, ** = these samples were consolidated to 50 kPa.*

Sample	Cont. (Y/N) [%]	SiO_2 content [%]	$Ca(OH)_2$ content [%]	Tested after [days]	Curing started	Test method	Tested
1	No	10	5	10	12/3	UCS	22/3
2	No	10	5	7	12/3	UCS	19/3
3	No	10	10	7	12/3	UCS	19/3
4	No	10	10	21	12/3	UCS	1/4
5	Yes	10	5	10	12/3	UCS	22/3
6	Yes	10	5	7	12/3	UCS	19/3
7	Yes	10	10	21	12/3	UCS	1/4
8	Yes	10	10	7	12/3	UCS	19/3
9	No	10	10	14	23/3	UCS	6/4
10	No	10	10	7	23/3	UCS	30/3
11	No	10	10	14	23/3	UCS	6/4
12	No	10	10	14	23/3	UCS	6/4
13	Yes	10	10	14	23/3	UCS	6/4
14	Yes	10	10	14	23/3	UCS	6/4
15	Yes	10	10	14	23/3	UCS	6/4
16	No	10	10	7	23/3	UCS	30/3
17	No	10	10	7	1/4	UCS/BET	8/4
18	No	10	10	7	1/4	UCS/BET	8/4
19	Yes	10	10	7	1/4	UCS/BET	8/4
20	Yes	10	10	7	1/4	UCS/BET	8/4
21	No	10	10	14	8/4	UCS/BET	22/4
22	No	10	10	14	8/4	UCS	22/4
23	Yes	10	10	14	8/4	UCS	22/4
24	Yes	10	10	14	8/4	UCS/BET	22/4

25	Yes	10	5	7	15/4	UCS	22/4
26	Yes	10	5	7	15/4	UCS	22/4
27	Yes	10	10	7	15/4	UCS	22/4
28	No	10	5	7	15/4	UCS/BET	22/4
29	No	10	5	7	15/4	UCS	22/4
30	No	10	10	7	15/4	UCS	22/4
31**	Yes	10	10	7	22/4	UCS	29/4
32**	Yes	10	10	7	22/4	UCS	29/4
33**	Yes	10	10	7	22/4	UCS	29/4
34**	No	10	10	7	22/4	UCS	29/4
35**	No	10	10	7	22/4	UCS	29/4
36**	No	10	10	7	22/4	UCS	29/4
37*	Yes	10	10	7	3/5	UCS	10/5
38*	Yes	10	10	7	3/5	UCS	10/5
39*	No	10	10	7	3/5	UCS	10/5
40*	No	10	10	7	3/5	UCS	10/5
41**	Yes	10	10	14	3/5	UCS	17/5
42**	Yes	10	10	14	3/5	UCS	17/5
43**	No	10	10	14	3/5	UCS	17/5
44**	No	10	10	14	3/5	UCS	17/5
Gothenburg Clay 45**	Yes	0	0	7	10/5	UCS	17/5
Gothenburg Clay 46**	No	0	0	7	10/5	UCS	17/5
Gothenburg Clay 47**	Yes	10	10	7	20/5	UCS	27/5
Gothenburg Clay 48**	Yes	10	10	14	20/5	UCS	3/6
Gothenburg Clay 49**	No	10	10	14	20/5	UCS	3/6
Gothenburg Clay 50**	No	10	10	14	20/5	UCS	3/6
Gothenburg Clay 51**	Yes	10	10	14	20/5	UCS	3/6
Gothenburg Clay 52**	No	10	10	7	20/5	UCS	27/5
Ref.**	No	0	0	-	24/3	Fall cone	15/4

Ref. 2*	Yes	0	0	-	24/3	Fall cone	15/4
Gothenburg ref.*	No	0	0	-	13/5	Fall cone	27/5
Gothenburg ref. 2*	Yes	0	0	-	13/5	Fall cone	27/5
Oedo	Yes	10	10		23/3	BET	15/4
Oedo	No	10	10		23/3	BET	15/4
Oedo 7	No	10	10		15/4	Oedo	6/5
Oedo 8	Yes	10	10		15/4	Oedo	6/5
Oedo 9	No	10	5		15/4	Oedo	6/5
Oedo 12	Yes	10	5		15/4	Oedo	6/5

4.1.2.1 Chemical Reactions

When adding the calcium hydroxide, the contaminated clay became blue-green and stayed in this color throughout, see Figure 28. This is due to a so-called precipitation double replacement reaction shown below (Equation 3), where the copper nitrate switched its nitrate group with the calcium's hydroxyl group, due to the solubility of calcium nitrate is higher than copper hydroxide. Thus, copper hydroxide is precipitated and the clay's color turns blue-green. It should be noted that copper hydroxide would also be created if copper sulphate was used instead of copper nitrate, this since it is less soluble than calcium nitrate. In fact, all copper compounds that are less soluble than the same compound bonded with calcium, will react and create copper hydroxide. However, if the solubility is equal for both compounds, no reaction will occur and the compounds will only be mixed, though this is rare to occur.

$$Ca(OH)_2\,(s) + Cu(NO_3)_2\,(aq) \rightarrow Ca(NO_3)_2\,(aq) + Cu(OH)_2\,(s) \qquad (3)$$

Figure 28. Start of mixing with calcium hydroxide (left), finished mixing after 10 minutes (right). Note the how the colour differs at the locations where the clay is in contact with the calcium hydroxide (marked in red), indicating production of copper hydroxide.

Since the green colour is a result of the copper hydroxide, it proved that some of the calcium hydroxide initially reacted with the copper nitrate. Though, the clay got stiffer throughout the mixing process, the calcium hydroxide most likely also reacted with the silicon oxide and formed C-S-H.

The reaction was similar when mixing the Gothenburg clay, where the calcium and copper created blue dots in the clay, see Figure 29. However, this was not as evident as for the kaolin clay as seen above.

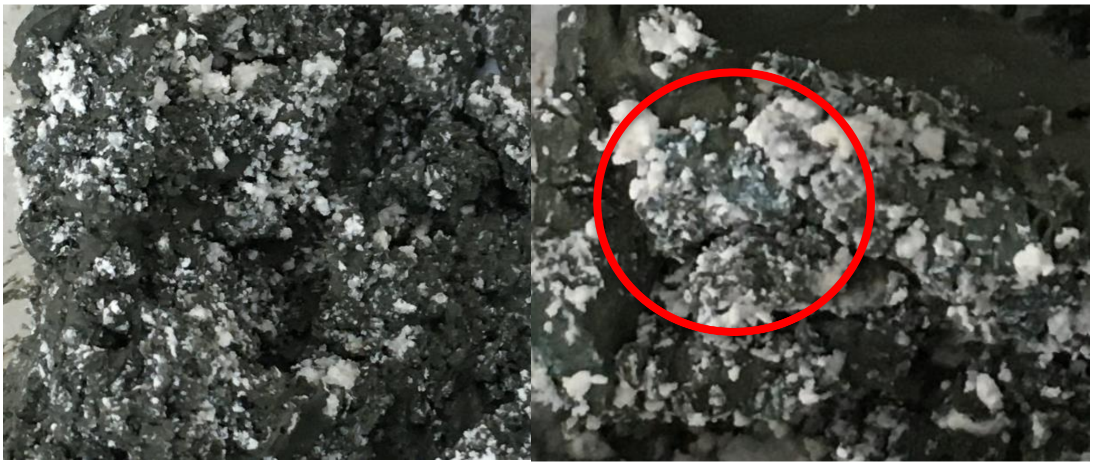


Figure 29. Mixing of uncontaminated Gothenburg clay with calcium (left) and contaminated clay (right). Note the small light-blue colour at the marked area, where the copper was oxidised.

It should be noted that the molar ratio between the reaction of calcium hydroxide and copper nitrate is 1:1, but due to the different concentrations, the actual molar ratio when mixing was 5:1 or 2.5:1. Hence, there was an abundance of calcium hydroxide and thus possible for the excess amount to react with the added silica. The calculations below (Equation 4-7) show that the contaminated soil needs 18.5% more calcium hydroxide than the non-contaminated, this due to the reaction with copper. This is based on a concentration of 10% calcium hydroxide and 5% copper nitrate.

$$n_{Ca(OH)_2} = \frac{m}{M} = \frac{7}{74} = 0.095\, mol \qquad (4)$$

$$n_{Cu(NO_3)_2} = \frac{m}{M} = \frac{3.5}{241} = 0.0145\, mol \qquad (5)$$

Hence the copper nitrate is the limiting mass of substance and since the molar ratio is 1:1 (Equation 5), 0.0145 mol of the 0.095 mol $Ca(OH)_2$ can react and create copper hydroxide and calcium nitrate. Hence the mass $Ca(OH)_2$ left to react with silica is:

$$m_{Ca(OH)_2}(reacted) = 0.0145 * 74 = 1.07g \rightarrow 7 - 1.07 = 5.93g$$

So, 5.93 g of the 7 g added $Ca(OH)_2$ can react with the silica, where the molar ratio also is 1:1. So, the higher concentration of contaminant there is, the more binder is needed. Moreover, the amount of calcium silicate produced from the reaction below, will be:

$$Ca(OH)_2 + SiO_2 \rightarrow CaSiO_3 + H_2O$$

$$n_{Ca(OH)_2} = \frac{m}{M} = \frac{5.93}{74} = 0.08\,mol \quad (6)$$

$$n_{SiO_2} = \frac{m}{M} = \frac{17.5}{60} = 0.29\,mol \quad (7)$$

Where the 0.08 mol $Ca(OH)_2$ is the limiting factor, hence 0.015 mol less C-S-H will be created, since it was 0.095 mol $Ca(OH)_2$ from the start, corresponding to 18.5% less C-S-H, compared to the uncontaminated clay.

The reaction for C-S-H production from $CaSiO_3$ is described by Equation 8:

$$CaSiO_3 + H_2O \rightarrow CaO * SiO_2 * H_2O \quad (8)$$

For this reaction, the molar ratio between calcium silicate and water is 1:1, indicating that 0.08 mol water should be added to achieve a balanced equation, hence ca 1.5g water. However, there are several different variants of calcium silicates, thus affecting the molar ratio between the calcium silicates and water (Baltakys, 2009). Further, the molar ratio between the silica and calcium hydroxide can range from 0.6 to over 2, indicating that it is not important to have a specific molar ratio to create C-S-H (Baltakys, 2009). Also, due to the pozzolanic material in the clay and the hydration of C-S-H, more water could be needed.

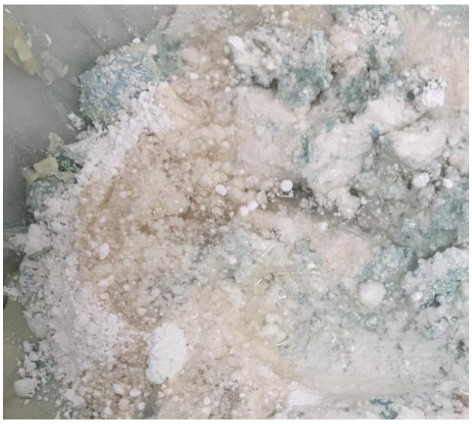


Figure 30. Reaction of Ca(OH)2 and SiO2, making the slurry brown at some locations with high concentration Ca(OH)2.

Once the silica was added, before mixing the slurry, the colour went from blue-green (the copper hydroxide) to beige-brown, at the locations where there was calcium hydroxide, see Figure 30. The slurry became instantly stiffer at these locations, thus it could indicate that small amounts of $CaSiO_3$ was created.

4.1.2.2. Safety and environmental protection

Risk assessments were conducted for all included chemicals and adequate protection equipment were used for the experimental work. All contaminated residuals were treated as chemical waste and were treated carefully to prevent contamination.

4.1.3. Test methods and testing

4.1.3.1. UCS Test

Uniaxial compression strength (UCS) tests were performed to evaluate the uniaxial compressive strength, according to ASTM D2166. With a strain rate of 1 mm/min (1.25 %/min) and a maximum strain of 20%, the tests were finished earlier if noticeable failure had taken place before 20% strain (ASTM, 2021). As mentioned, and seen in Table 8, multiple samples with the same properties were tested several times to reduce the impact of the sample quality and repeatability of the results. The UCS test set-up is presented in Figure 31. The test was started once the load cell almost had established contact with the metal ball.

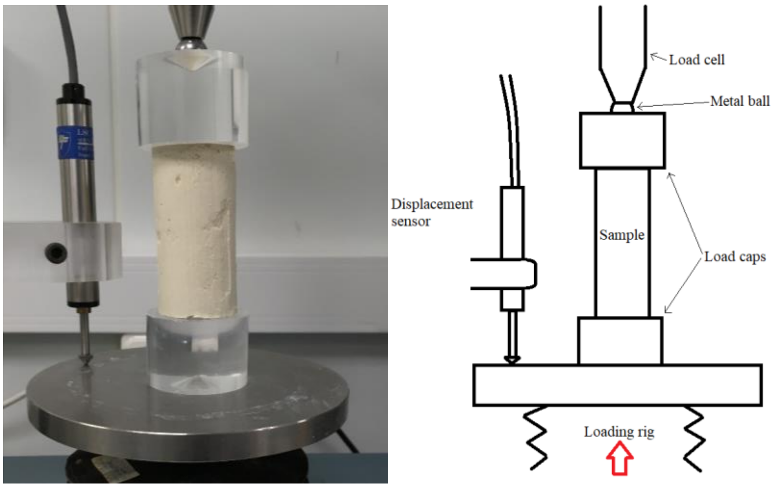


Figure 31. Setup of an UCS test (left) and schematic figure of the setup (right), note that the red arrow describes the direction of movement.

The stiffness of the samples was evaluated by Young's modulus from the UCS test.

Young's Modulus (E'_{50}) was derived from the stress-strain relationship by taking the secant of the slope E'_{50}, as shown below in Figure 32, according to Equation 9. Note that the E' is derived by taking the tangent of the slope, while the secant modulus (E'_{50}) is the secant between the origin and half of the peak strength $\frac{q_f}{2}$.

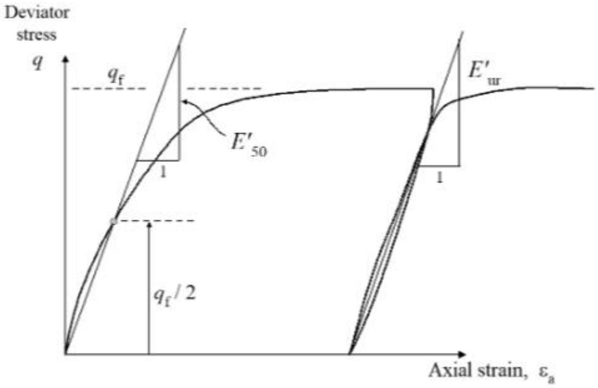


Figure 32. Evaluation of Young's Modulus by the tangent (E'ur to the right) and the secant modulus (E'50 to the left) (Knappett & Craig, 2019).

$$E'_{50} = \frac{q_f - \frac{q_f}{2}}{\varepsilon_f - \frac{\varepsilon_f}{2}} \tag{9}$$

Where q_f is the peak strength and ε_f is the maximum strain at peak strength.

While testing, the load and displacement were measured and the stress was calculated by dividing the measured force with the corrected area, that was calculated according to ASTM D2166 (Equation 11) (ASTM, 2021). Due to the fast testing and undrained conditions, this method is applicable.

$$\sigma = \frac{Q}{A} \tag{10}$$

Where Q is the measured load, and A is the corrected area as described below:

$$A = \frac{A_0}{(1 - \frac{\varepsilon}{100})} \tag{11}$$

Where A_0 is the initial aera before starting the test and ε is the strain at a given load.

4.1.3.2. Oedometer test

Oedometer tests were performed on four samples with different additive concentrations, to evaluate the increase in stiffness, (*i.e.* 5% and 10% calcium hydroxide, and 10% silica, equal for all samples), in which two of them were contaminated and the other two were uncontaminated. The displacements were monitored and recorded every ten seconds, to determine the increase in strain. The loading steps were applied in a window of 8 hours and the samples (consolidation was completed very fast) and then the sample was left under constant effective stress of 50 kPa for a week, when the procedures were repeated. The total duration of the test was 3 weeks.

It is important to state that the curing can go faster with increasing overburden stress, hence the stiffness of the samples will most likely be higher than to the ones stored without pressure for seven days. See Figure 33 for the set-up of the oedometer tests.

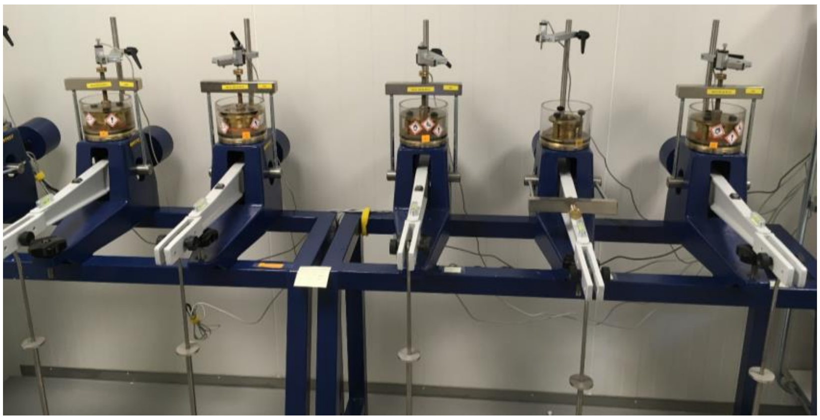


Figure 33. Setup of oedometer testing for determine the increase in stiffness.

The stiffness was evaluated by calculating the compressibility indices, κ and λ, for the different samples, described by Figure 34.

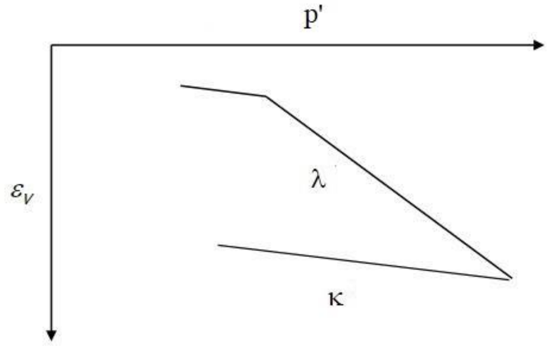


Figure 34. Evaluation of κ and λ based on oedometer data, note that the κ-values were evaluated from the unloading/reloading cycle of the oedometer test and the λ-values from the first loading cycle.

Where *p'* is the mean effective stress as by Equation 12 and ε_v is the volumetric strain, calculated from the measured displacement data.

$$p' = \frac{\sigma'_{min}+\sigma'_{max}}{2} \tag{12}$$

κ and λ are, as shown, the slope of the curve, as described by Equation 13 and 14 (Knappett & Craig, 2019):

$$\kappa = \frac{\Delta\varepsilon_v/100}{\ln(\Delta p')} \tag{13}$$

$$\lambda = \frac{\Delta\varepsilon_v/100}{\ln(\Delta p')} \tag{14}$$

A lower value of κ and λ corresponds to a stiffer soil, since a greater stress is required for the same level of strain. Whereas a softer soil requires a lower stress, a lower stress-strain coefficient, hence higher κ and λ.

The rate of consolidation was also evaluated by calculating the coefficient of consolidation, c_v, according to both the Casagrande method (Equation 15) and the Taylor method (Equation 16). Where a higher c_v corresponds to a faster consolidation process and thus a softer soil. (Knappett & Craig, 2019)

$$c_v = \frac{0.196 * d^2}{t_{50}} \tag{15}$$

$$c_v = \frac{0.848 * d^2}{t_{90}} \tag{16}$$

Where d^2 is half of the sample height in meter and t_{50} and t_{90} is the time at the point that corresponds to 50% and 90% of the final consolidation respectively, in minutes. 0.196 and 0.848 is empirically based constants for U50 and U90, that is 50% and 90% of the final displacement, respectively.

The coefficient of volume compressibility, m_v, can also be calculated based on the oedometer data. This is a coefficient that explains the relationship between increase in stress and change in volume, (1/kPa). It was calculated according to Equation 17. (Knappett & Craig, 2019)

$$m_v = \frac{\Delta V/V}{\Delta\sigma} \tag{17}$$

Where V is the initial volume of the sample, ΔV is the change in volume and $\Delta\sigma$ is the difference in stress between the lowest and highest load increment.

Based on m_v and c_v, the hydraulic conductivity can be calculated, according to Equation 18 below. Also, since m_v describes the strain-stress relationship, the oedometer modulus E_{OED} can be calculated, see Equation 19 (Knappett & Craig, 2019). This is related to Young's modulus, as explained by Equation 20 (Atkinson, 2007). Note that since the clay might not behave as a soil due to the cementation, this hydraulic conductivity should only be used as a comparative parameter.

$$k = c_v * \gamma_w * m_v \tag{18}$$

$$E_{OED} = \frac{1}{m_v} \tag{19}$$

$$E' = E_{OED} * \frac{(1-2v^2)}{(1-v)} \tag{20}$$

Where γ_w is the unit weight of water = 9.81 kN/m^3 and ν is Poisson's ratio = 0.2.

4.1.3.3 Bender Element Test

For evaluating the small-strain stiffness of the samples, Bender Element Test were conducted. This is a test where the bender element on the base cap generates a bending motion, which produces a shear wave. The shear wave propagates through the soil sample and is received at the bender element in the top cap, which detect the propagated vibration and generate a voltage output that can be measured. See Figure 35 for the layout of the setup and a schematic figure of the test.

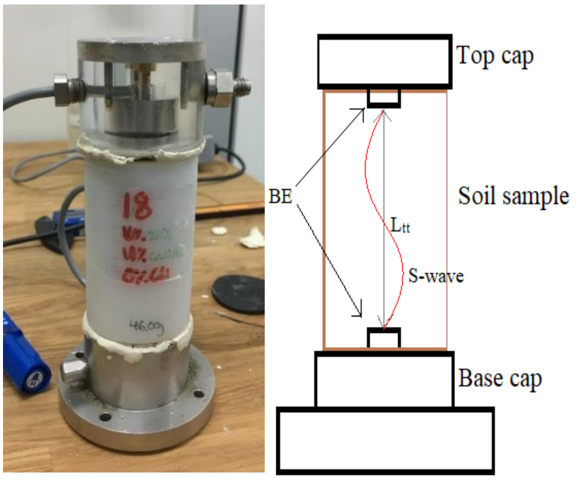


Figure 35. Set up of the Bender Element test on a 38x76mm sample (left) and a schematic figure of the test (right). Note the added kaolin clay at the top and bottom. The length of the Bender Element (BE) is 3 mm.

Two samples for different curing times of 7-40 days and with 10% calcium hydroxide and 10% silica were tested. Kaolin clay was used as fill-up material between the caps and the sample to ensure there was sufficient connection between the samples and the Bender Element. Kaolin was also added to the small area where the conductors have penetrated the sample, to keep the sample as intact as possible before the UCS test.

Since the transmitted voltage was sent as a shear wave through the sample, the time between the transmitted peak and the receiving peak can be measured and the shear wave velocity can be calculated. Thus, the shear modulus can be evaluated by Equation 21 below, describing the small-strain stiffness (Leong *et al.*, 2005).

$$G = \rho * v_s^2 \tag{21}$$

Where ρ is the soil density and v_s is the shear wave velocity obtained from the Bender Element test, according to Equation 22. L_{tt} is the distance between the Bender Elements (Leong *et al.*, 2005).

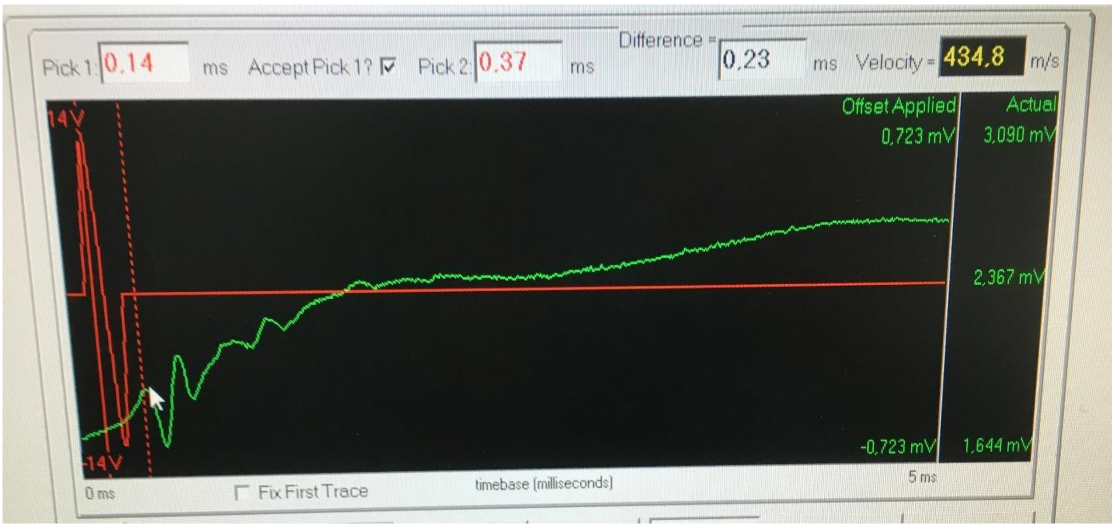
$$v_s = \frac{L_{tt}}{t} \tag{22}$$

Since the stiffness of the soil influences the choice of frequency, where a less stiff soil requires a lower frequency and vice versa, three different frequencies were tested, namely 2.5, 5 and 10 kHz (Rahman *et al.*, 2016). According to the literature, there is an optimum frequency depending on the soil type and its properties. This is explained by the R_d-value, which is the height of the sample (d) relative to the wavelength (λ) of the shear wave, explained by Equation 23 (Jovicic et al, 1996). For kaolin clay, and kaolin clay mixed with cement, the R_d-value should be 10.35-20.5, whereas some say it should be lower than 1 or between 1 and 10 (Rahman *et al.*, 2016; Xiao *et al.*, 2018). Since the

Bender Element test was conducted on samples with heights of 76 mm, 40 mm and 20 mm, the R_d-value will differ between the tests with the same frequency.

$$R_d = \frac{d}{\lambda} = \frac{df}{v_s} \tag{23}$$

The transmitted voltage was 14V and the time was measured from the first peak of the transmitted wave to the first noticeable peak of the receiving wave. This was done even though there could be a greater peak afterwards, as visualized in Figure 36 below.



Pick 1: 0,14 ms Accept Pick 1? ☑ Pick 2: 0,37 ms Difference = 0,23 ms Velocity = 434,8 m/s

☐ Fix First Trace

Figure 36. Evaluation of shear wave velocity from Bender Element test, the red line corresponds to the transmitted voltage and the green corresponds to the received voltage. Note that the shear wave velocity is calculated from the first noticeable peak in the received voltage as displayed.

4.1.3.4 Fall cone test

The reference samples (without binder) even after consolidation to 50 kPa, were very soft, in which extrusion of the samples without disturbance was not possible, resulting in poor UCS results. Therefore, the fall cone testis more appropriate to evaluate the undrained shear strength of the samples.

The fall cone test was performed, according to ISO 17892-6:2017, using 60° angle tip and 100 g weight for the soft samples and 30° tip angle and 80 g weight for the stiffer soils. The results from the fall cone test are comparable with the UCS test by calculating the undrained shear strength (c_{ufc}) of the clay, according to Equation 24. (ISO 17892-6:2017)

$$c_{ufc} = c * g * \frac{m}{i^2} \tag{24}$$

Where c is a constant depending on the tip angle, c = 0.8 for 30° and c = 0.27 for 60°, g is the gravitational constant = 9.81 m/s^2, m is the mass of the cone and i is the average cone penetration. The different set-ups are shown in Figure 37.

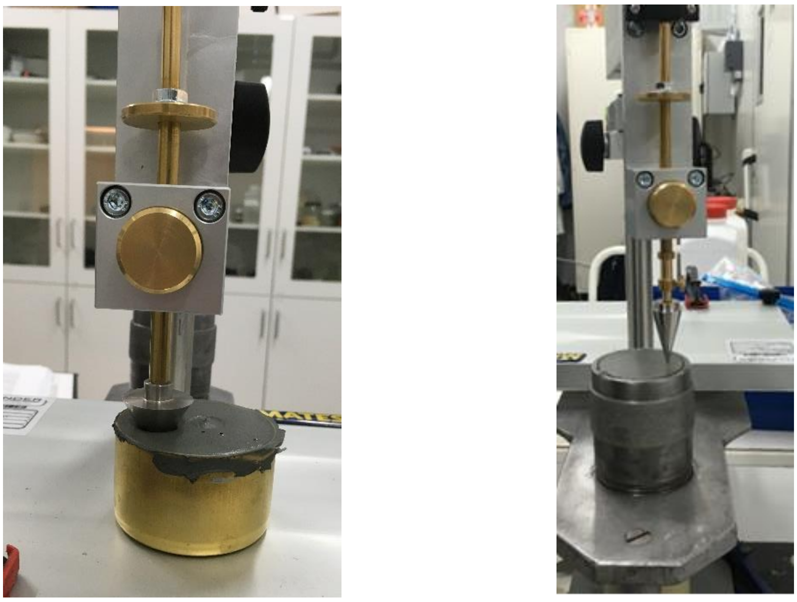


Figure 37. Fall cone test of Gothenburg clay, remoulded sample with a 30° tip to the left and an undisturbed sample with a 60° tip to the right.

4.1.3.5. SEM-Analysis

The consolidated samples of the kaolin clay and Gothenburg clay were investigated by scanning electron microscope (SEM), this to evaluate potential differences in structure and investigate how the copper interacts with the C-S-H production.

SEM is a method for imaging and phase identification that has been used extensively for quantifying the 2D morphological topography of clays. This is achieved by scanning the surface with a focused electron beam. The FEI Quanta 200 ESEM instrument at the Chalmers Materials Analysis Laboratory (CMAL) was used for image acquisition and mapping of elements. Slices were collected from the test samples and subsequently were freeze dried prior to scanning in order to preserve microstructure intact. After freeze drying, additional fractures were made to reveal the internal surfaces. The measurements were acquired in high vacuum mode to achieve the highest resolution. For imaging with the back-scatter detector, a 15 kV beam was used. Information on the chemical analysis of the sample constituents was obtained by the Energy Dispersive X-ray (EDX) mode of the instrument for elemental mapping.

4.2 Practical application of sustainable remediation

This was conducted with help from Skanska, which is one of the largest construction companies in the Nordic. By conducting a questionnaire, based on 39 site managers at different projects, asking what they thought of working with remediation, and how efficient the regulations are, an overview of Sweden as whole was obtained. The following questions were asked:

- Have you ever done remediation that was not necessary, based on regulations and guidelines?
- Do you feel that working with remediation is complicated, due to regulations and laws?
- Is the work with remediation sustainable (environmentally, economically and social), or could it be improved to be more efficient?
- What is your general opinion on the regulations and laws regarding remediation? Is it good/bad, complicated etc.?

Also, Svevia, a state-owned company working with the maintenance of infrastructure and remediation, was also asked these questions.

Since the problems with landfill and disposal of waste lies somewhat in responsibility of the municipality, the city of Gothenburg was also interviewed. This to understand their point of the problem and to compare this with the result from the questionnaire. The following questions were asked:

- What, and how, are You doing to make remediation more sustainable, *i.e.* to use more sustainable methods?
- What is your general perception on how construction companies are dealing with remediation and how sustainable it is today?
- Do You experience complaints or other comments from users, that remediation is difficult to implement?
- What do You think is needed to achieve a more sustainable remediation?
- Do You experience any problems with remediation, *e.g.* that companies are taking shortcuts and “cheats”, or that the sustainable management is low?

The Swedish EPA was also given these questions (with one additional question) to understand what the governing agency thinks about sustainable remediation and present problems.

5. Results

5.1. Laboratory testing

The results from the different test methods are presented in section 5.1.1-5.1.3., where the results for the Gothenburg clay is presented in section 5.1.4. The samples were of good quality and the consolidated samples yielded more accurate results, why they are used when comparing the kaolin clay to the Gothenburg clay.

As presented in Figure 38 and as described in Table 10, were the samples of similar quality.

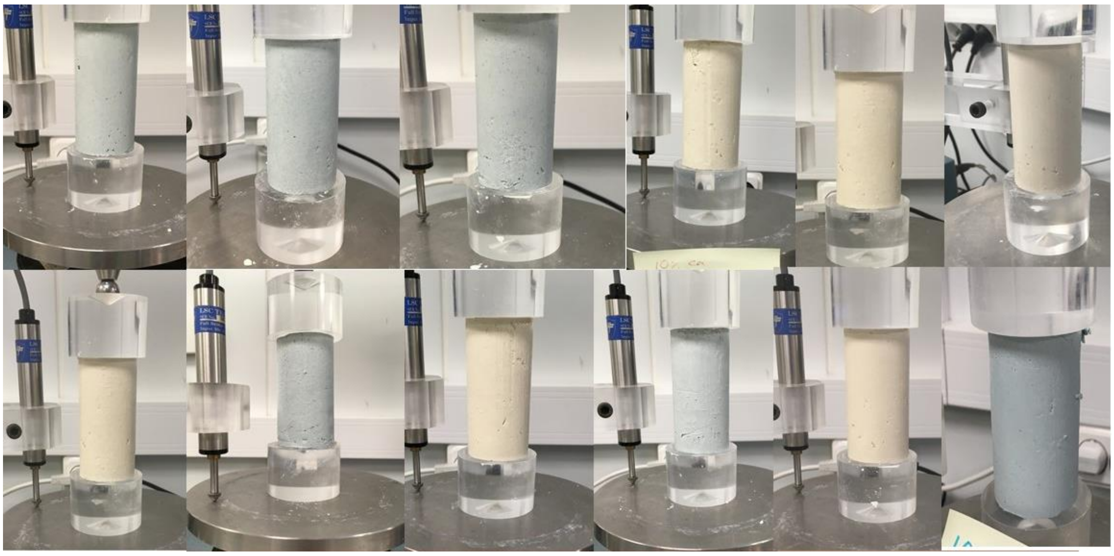


Figure 38. A selection of samples, showing the quality consistency of the samples, all samples had similar surface quality. It should be noted that the concentration of additives and curing time did not affect the visual appearance of the samples.

The different failures for different curing times are presented in Figure 39, all with 10% calcium hydroxide and silica.

The samples become more brittle with time and fails immediately after the sample has start to crack, compared to seven days where it starts to crack but does not fall apart as fast. Note the similar behaviour between the uncontaminated and contaminated kaolin samples, compared to the significantly different behaviour of the uncontaminated and contaminated Gothenburg clay.

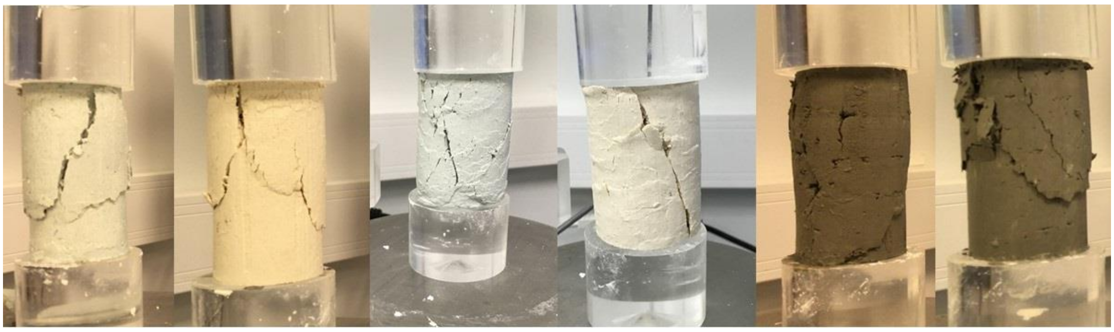


Figure 39. Different types of failure after 14 days contaminated/uncontaminated (left), 7 days contaminated/uncontaminated (middle) and 7 days Gothenburg clay contaminated/uncontaminated (far right). Note the buckling failure of the contaminated Gothenburg clay sample, compared with the highly brittle failure of the uncontaminated samples.

Previous studies have reported the great effect of water content on the mechanical behaviour of soil (Ahmadi-Nagadeh and Toker, 2019; Ahmadi-Naghadeh, 2016). The variation of water content and density of the samples before and after curing are summarised in Table 10, full details are presented in Appendix A. It can be seen that the densities decrease slightly, whereas the water content has decreased noticeably from 80% to 45%, as a result of the hardening process of the binders. The data is based on 30 partially submerged "boxed" samples, 14 consolidated samples and four submerged samples. The samples have similar water content and densities, indicating on similar properties.

Table 10. Summary of sample properties, based on 48 samples as described in Chapter 4.

Sample	**Water Content after curing [%] (Max, Min)**	**Density Before curing [kg/m^3] (Max, Min)**	**Change in Density after curing [%] (Max, Min)**
"Boxed"	44 ± 2 (49, 43)	1.46 ± 0.02 (1.49, 1.43)	0.7 ± 1 (3.4, -0.7)
Consolidated	42 ± 1 (44, 39)	1.44 ± 0.03 (1.47, 1.41)	6.2 ± 3 (10, 1.8)
Submerged	44 ± 0 (44, 44)	1.47 ± 0.02 (1.48, 1.45)	-0.1 ± 1 (1, -1)

Another important factor for the hardening process and for sufficient binding between the additives is having a high pH environment. The pH evolution through the mixing and curing process is presented in Table 11.

Table 11. Change in pH over time for mixtures with 10% SiO_2 and 10% $Ca(OH)_2$.

Mixture	pH of slurry	pH after mixing	pH after 7 days	pH after 40 days
Uncontaminated kaolin	6	12.6	12.8	12.1
Contaminated kaolin	4.3	12.3	12.2	10.6
Uncontaminated Gothenburg clay	8.1	12.9	12.8	-
Contaminated Gothenburg clay	6.6	12.5	12.2	-

As seen, there is a significant increase in pH instantly after mixing, due to the basic reactions of the calcium hydroxide. Note that the pH is still high after 40 days, at 10.6 for the contaminated samples and 12.1 for the uncontaminated.

The reference strength for the tested kaolin and Gothenburg clay is presented in Table 12, based on the fall cone test. Note that two tests were done for the Gothenburg clay, both from the undisturbed sample and from the remoulded sample. Here, there is a significant increase in strength for the contaminated kaolin clay, whereas the Gothenburg clay has similar strength uncontaminated and contaminated.

Table 12. Reference peak strength for the used clays, consolidated to 50 kPa. Note that the undisturbed Gothenburg clay was extracted at a depth of 4 m.

Clay	Uncontaminated Peak strength [kPa]	Contaminated Peak strength [kPa]
Kaolin	3.2	3.8
Gothenburg clay	17.5	17.3
Gothenburg clay undisturbed	8.7	-

5.1.1. UCS Test

The strength is represented by the peak axial stress and the stiffness is represented by the Young's Modulus. Figure 40 and Table 13 in this chapter represent the samples that were consolidated to 50 kPa, this since they yielded the most accurate results and are also more applicable in field. The influence of hydration and curing processes will be presented in 5.1.1.1.

Table 13 and Figure 40 below presents the results of the six samples that were stored under 50 kPa of overburden pressure and fully submerged in water, all to represent the reality. As can be seen, the stiffness and strength are noticeably higher for the contaminated samples. Since the hardening process takes time, there is a significant increase in strength between seven and 14 days of curing. As mentioned in the methodology, the aim was not to evaluate at what curing age yields the highest strength, but to evaluate the influence of a heavy metal.

Table 13. UCS results of consolidated samples.

Sample	Peak Axial Stress [kPa]	Young's Modulus [kPa]
Contaminated 7 Days	39 ± 4	328 ± 45
Uncontaminated 7 Days	31 ± 3	149 ± 10
Contaminated 14 Days	81 ± 7	1833 ± 4
Uncontaminated 14 Days	52 ± 3	1046 ± 53

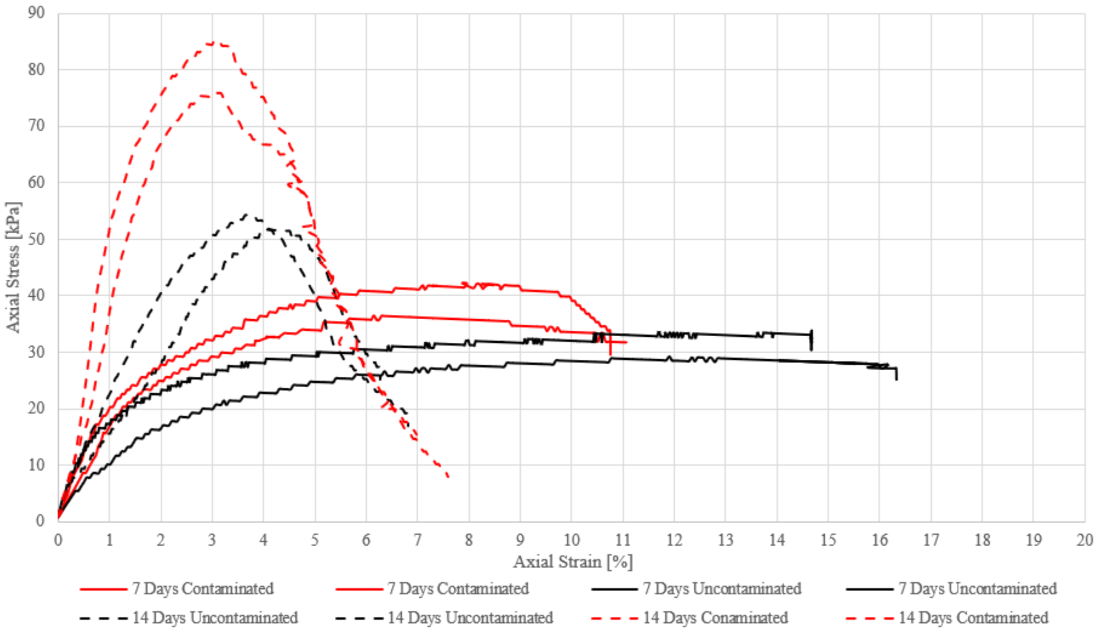


Figure 40. Results of consolidated kaolin samples all prepared with 10% $Ca(OH)_2$ and 10% SiO_2.

5.1.1.1. Effect of saturation and storing

Table 14, Table 15 and Figure 41, summarises all results for the samples that were stored partially submerged in water (in the box) and wrapped in plastic film. The contaminated samples have noticeably lower peak strength and stiffness. However, there is a large scatter as seen on the standard deviation (the ±). This will be discussed further later.

Table 14. Stiffness properties in terms of Young's Modulus.

Young's Modulus [kPa]	Max	Min	Average ± stdav
7 Days Uncontaminated	11455	1299	5718 ± 4193
7 Days Contaminated	2335	429	1863 ± 730
14 Days Uncontaminated	5235	4568	4811 ± 368
14 Days Contaminated	4375	2095	3144 ± 1045

Table 15. Peak strength using 10% $Ca(OH)_2$ and 10% SiO_2 for all samples.

Peak Axial Stress [kPa]	Max	Min	Average ± stdav
7 Days Uncontaminated	72	31	55 ± 17
7 Days Contaminated	38	22	33 ± 7
14 Days Uncontaminated	87	43	62 ± 21
14 Days Contaminated	59	31	46 ± 13

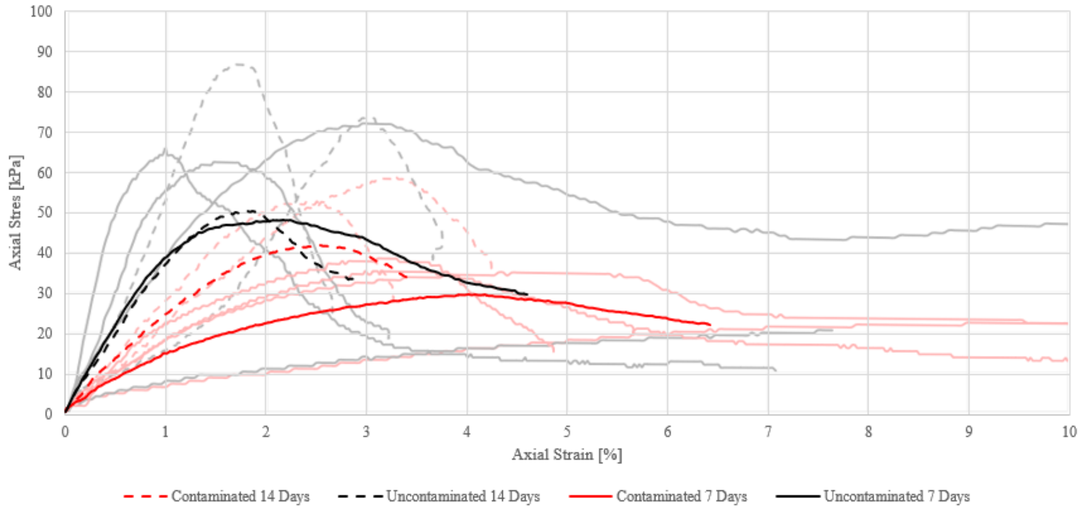


Figure 41. The average results of the "boxed" samples, based on 12 tested samples shown in the background in faded colours.

For the 7 days submerged samples, the strength and stiffness were significantly lower than for the consolidated and partially submerged samples, as shown in Table 16. The peak strength was almost identical for both samples, but the uncontaminated samples were a bit stiffer, as can be seen by the higher stiffness.

Table 16. UCS results for the submerged samples, cured for 7 days. Note the similar peak strength.

Sample	Peak Axial Stress [kPa]	Young's Modulus [kPa]
Contaminated	19 ± 3	91 ± 16
Uncontaminated	19 ± 1	117 ± 0.2

5.1.1.2. Effect of binder concentration

Different concentrations of $Ca(OH)_2$ were tested by alternating the concentration between 5% and 10% $Ca(OH)_2$ while keeping the concentration SiO_2 constant at 10%. The results are presented in Figure 42 and Table 17. It can be seen that the failure behaviour is similar but with a noticeably lower peak strength for the samples with 5% $Ca(OH)_2$. All samples were cured consolidated to 50 kPa.

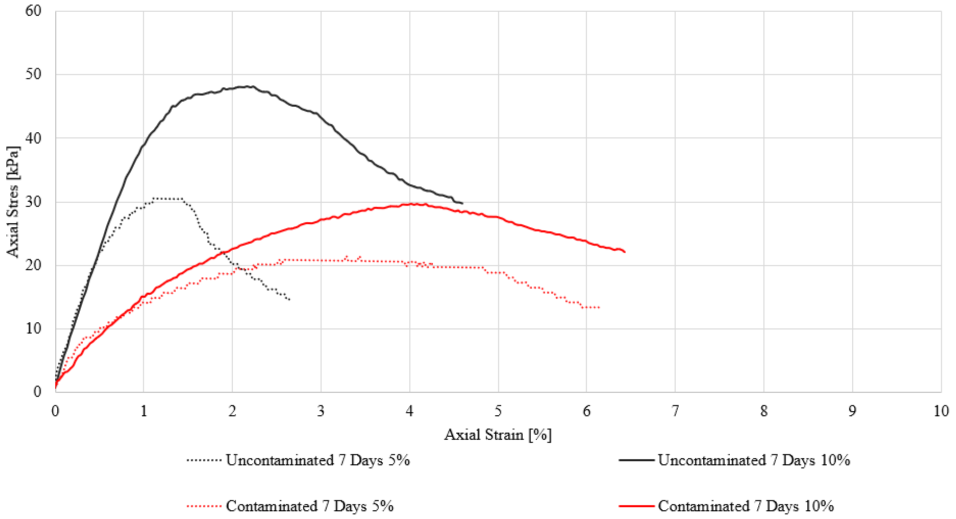


Figure 42. Stress-strain behaviour of lower (5%) and higher (10%) concentrations of $Ca(OH)_2$, all with 10% SiO_2.

The stiffness is similar between the two concentrations, where the samples with lower concentration have higher Young's Modulus. It is evident that the uncontaminated samples reach a lower peak axial strain at peak axial stress, and have a more brittle failure, compared to the contaminated samples that have a more ductile failure behaviour.

Table 17. Peak axial strength and Young's modulus for different concentrations of $Ca(OH)_2$. Note the similar stiffnesses for the samples.

Sample	Peak Axial Stress [kPa]	Young's Modulus [kPa]
Uncontaminated 5% $Ca(OH)_2$	30.5	1794
Uncontaminated 10% $Ca(OH)_2$	48	1590
Contaminated 5% $Ca(OH)_2$	21	514
Contaminated 10% $Ca(OH)_2$	30	484

5.1.2. Bender Element Test

The results from the Bender Element tests are summarised in Figure 43 and the contaminated and uncontaminated results are shown in Figure 44 and Figure 45 respectively. It is evident that the contaminated clay is softer since it takes a longer time for the shear wave to transmit through the sample. This can be seen where the first noticeable peak for uncontaminated samples occurs much sooner than for the contaminated samples. It should be stated that all Bender Element tests are based on the partially submerged samples and not the consolidated samples. However, the samples were chosen randomly so the mentioned scatter is considered.

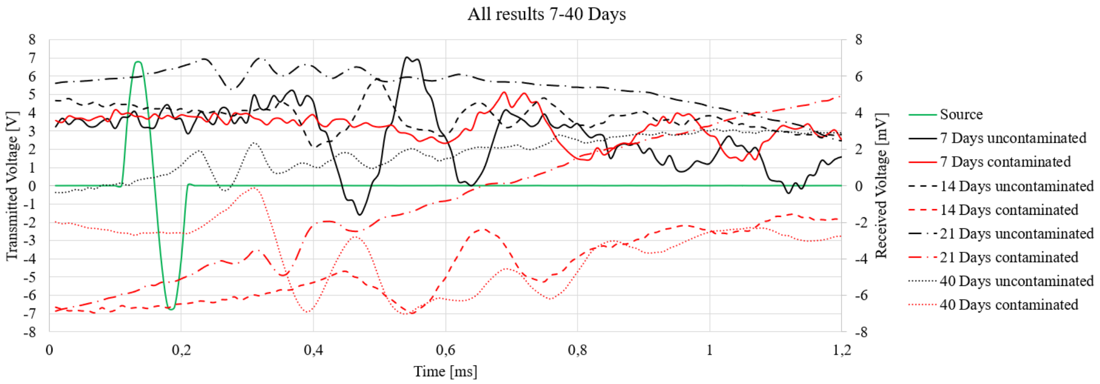


Figure 43. Results for Bender Element test 7-40 Days, 2.5 kHz was used for soft samples, 5 kHz for medium stiff samples and 10 kHz for stiff samples.

When studying the graphs in more detail, there is just a small difference between the receiving time for the 21-day and 40-day sample, indicating similar stiffness. This is visualised in Figure 44 and Figure 45.

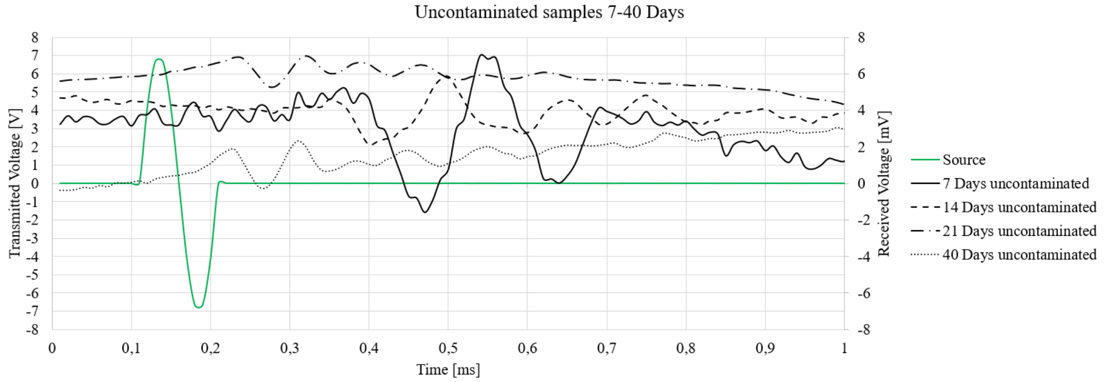


Figure 44. Results for Bender Element test 7-40 Days on uncontaminated samples. 2.5 kHz was used for soft samples, 5 kHz for medium stiff samples and 10 kHz for stiff samples.

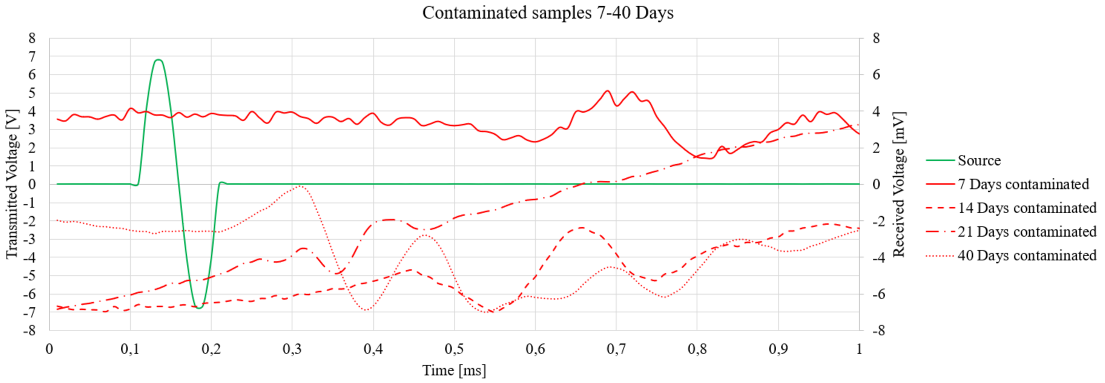


Figure 45. Results for Bender Element test 7-40 Days on contaminated samples. 2.5 kHz was used for soft samples, 5 kHz for medium stiff samples and 10 kHz for stiff samples. Note the small difference between 21 days and 40 days.

For 21 and 40 days of curing, 40 mm oedometer samples were used, hence the shorter receiving time due to shorter transmitting distance. It should be noted that a longer receiving time results in a vastly lower shear modulus since it is related quadratic to the shear wave velocity and not proportional. As seen in Figure 46, there is an increase of stiffness with time, but where the stiffness of the contaminated sample is higher at 14 days than at 21 days and 40 days.

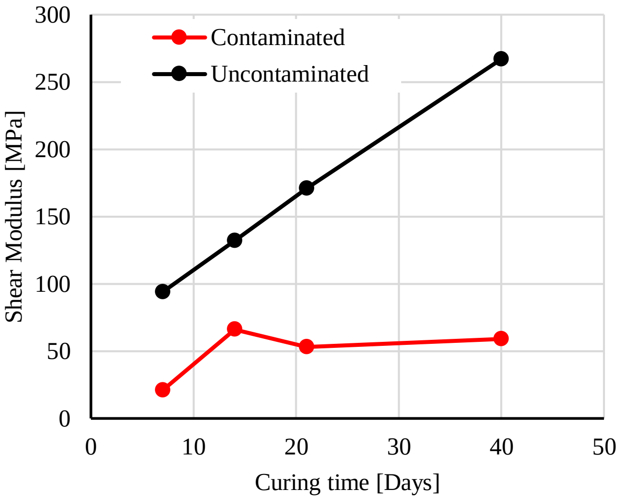


Figure 46. Shear modulus vs. curing time based on Bender Element test. Note that all samples have 10% calcium hydroxide and 10% silica.

The R_d-values for the different samples heights and frequencies are presented in Table 18, to validate the choice of frequencies. Note that the values will differ with the type of mixture and curing time, which is why only the lowest and highest values are presented. The shorter and stiffer the samples are, the lower will the R_d-value be. As seen, most of the samples have roughly the same values, the 40 mm samples were cured for 40 days hence low values due to their stiffness. All of the values were in-line with the literature, which indicates that the chosen frequencies were appropriate.

Table 18. Minimum and maximum R_d-values for the different types of samples and frequencies. Note that all types of mixtures (5% & 10% Calcium) and curing times (7-40 days) were included, hence the spread.

Sample Height [mm]	**40**	**76**
Frequency [Hz]	2 500-10 000	2 500-10 000
Shear Wave Velocity [m/s]	189-425	120-300
R_d [-]	0.2-2.1	0.6-6

5.1.3. Oedometer Test

The results from the oedometer tests are presented in Table 19, Table 20 and is evaluated from Figure 47 and Figure 48. Showing on a higher stiffness in form of compressibility indices for the uncontaminated samples but lower oedometer modulus. However, the contaminated samples showed on smaller displacement, where the contaminated sample with 10% $Ca(OH)_2$ and has the smallest displacement, which can be seen in Figure 47. It should be noted that all samples had 10% SiO_2.

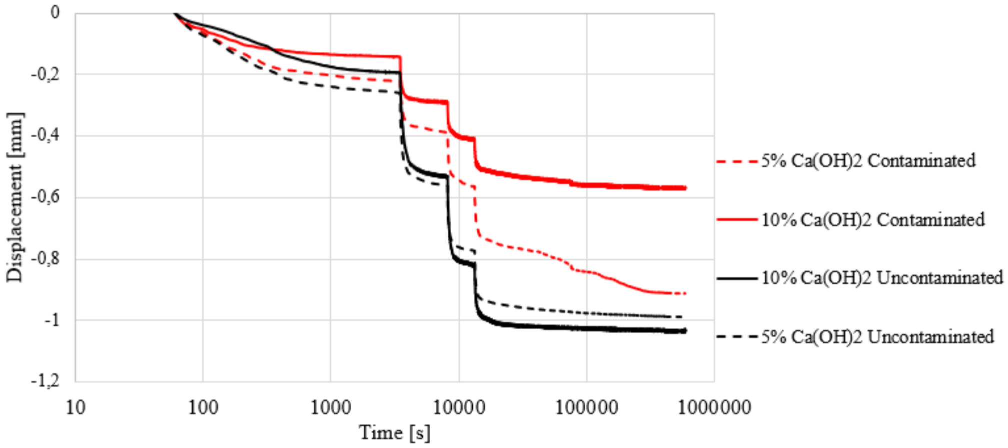


Figure 47. Oedometer results showing displacements vs. time with a total loading of 50 kPa. Note the larger displacements for the uncontaminated samples.

The contaminated sample with 10% $Ca(OH)_2$ is the stiffest sample according to the compressibility indices and the uncontaminated sample with 5% calcium hydroxide was the softest. A lower κ-value and λ-value corresponds to a stiffer soil. The κ-values are based on the unloading and reloading of the samples, see Figure 48, hence different curing times compared to the λ-values.

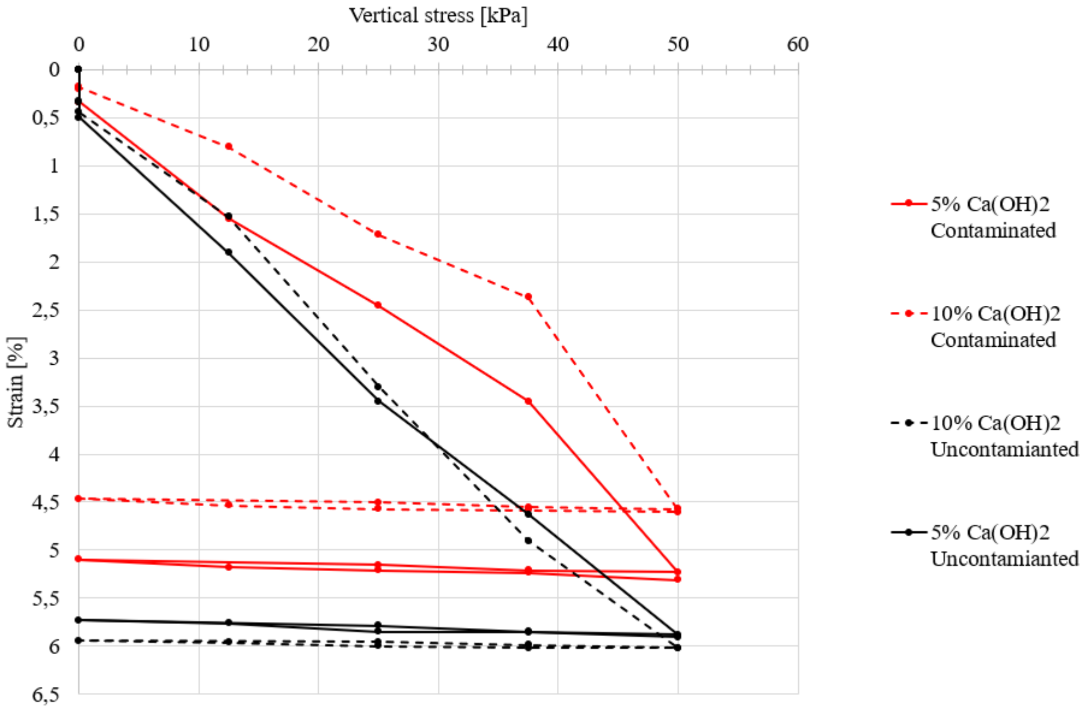


Figure 48. Loading and unloading/reloading of the different samples. Note that the samples gained stiffness throughout the test, since they were tested over three weeks.

Here it is evident that the dashed-lines when unloading/reloading are flatter than the solid lines, and that the solid lines are steeper during the initial loading phase, indicating on higher stiffness when having 10% $Ca(OH)_2$. Further, the dashed red line is flatter than the dashed black line, indicating that the contaminated soil is stiffer. Note that the contaminated sample with 5% $Ca(OH)_2$ is stiffer than both the uncontaminated samples at early stages but is softer afterwards. This can be seen in Table 19, showing the compression indices.

Table 19. λ- and κ-values based on oedometer tests.

Mixture (All have 10% SiO_2)	κ [-] (Unloading/reloading)	λ [-]
5% $Ca(OH)_2$, 0% $Cu(NO_3)_2$	$4.4*10^{-3}$	0.015
10% $Ca(OH)_2$, 0% $Cu(NO_3)_2$	$4.3*10^{-3}$	0.014
5% $Ca(OH)_2$, 5% $Cu(NO_3)_2$	$5.9*10^{-3}$	0.012
10% $Ca(OH)_2$, 5% $Cu(NO_3)_2$	$3.7*10^{-3}$	0.010

The coefficient of consolidation and volume compressibility and hydraulic conductivity are presented below in Table 20. Due to different behaviour at final loading step, as seen in Figure 47, the Casagrande method provided better values since it is based on t_{50} and not t_{90}. For this, the uncontaminated samples have a higher coefficient of consolidation which corresponds to a faster consolidation, hence a softer soil. The pattern is the same for the hydraulic permeability, showing higher values for the uncontaminated samples, indicating on a less dense soil. Lastly, the uncontaminated samples have similar Young's modulus, E', at a loading of 50 kPa, but where the contaminated sample with 10% $Ca(OH)_2$ has the highest stiffness. This corresponds well with the UCS results, showing that the contaminated samples have higher strength than the uncontaminated.

Table 20. Consolidation properties inform of coefficient of consolidation and volume compressibility and hydraulic conductivity. Note that all results are based on the Casagrande method.

Mixture (All have 10% SiO_2)	c_v [m^2/min] (Taylor)	c_v [m^2/min] (Casagrande)	m_v [1/kPa]	E' [kPa]	k [m/s]
5% $Ca(OH)_2$, 0% $Cu(NO_3)_2$	$4.5*10^{-6}$	$2.6*10^{-7}$	0.0012	5081	$1.8*10^{-7}$
10% $Ca(OH)_2$, 0% $Cu(NO_3)_2$	$4.6*10^{-6}$	$2.7*10^{-7}$	0.0012	5508	$1.9*10^{-7}$
5% $Ca(OH)_2$, 5% $Cu(NO_3)_2$	$1.9*10^{-6}$	$1.2*10^{-7}$	0.0011	2491	$7.4*10^{-8}$
10% $Ca(OH)_2$, 5% $Cu(NO_3)_2$	$4.5*10^{-6}$	$2.7*10^{-8}$	0.0009	7627	$1.5*10^{-8}$

5.1.4. Gothenburg clay

The results for the Gothenburg clay were significantly different to the kaolin samples, where the contaminated samples have vastly lower strength. The results are presented in Table 21 and Table 22 and visualised in Figure 49. Note that all samples had 10% $Ca(OH)_2$ and 10% SiO_2 and were consolidated to 50 kPa. The scatter for the uncontaminated samples was noticeably larger than for the kaolin, as seen by the standard deviation.

Table 21. Axial peak strength for Gothenburg clay, obtained from UCS test.

Sample	Peak Strength 7 Days [kPa]	Peak Strength 14 Days [kPa]
Contaminated	46	50 ± 2
Uncontaminated	59	98 ± 25

Table 22. Young's modulus (secant modulus) for Gothenburg clay, obtained from UCS test.

Sample	Young's Modulus 7 Days [kPa]	Young's Modulus 14 Days [kPa]
Contaminated	360	484 ± 78
Uncontaminated	854	2109 ± 1366

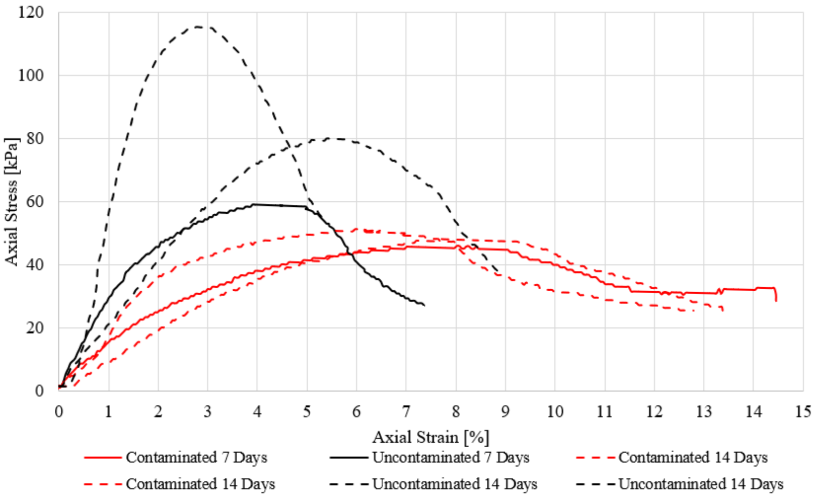


Figure 49. UCS-result of contaminated and uncontaminated Gothenburg clay.

5.1.5. SEM-Analysis

EDX provides elemental mapping on the scanned image. Thus, the spatial distribution of different elements can be resolved. In the following Figures Cu^{2+} is represented by green colour, Ca^{2+} by red and the positions that they overlap is highlighted yellow. As shown by Figure 50 and Figure 51, at all locations there is copper, there is also calcium, indicating that copper binds to calcium ions. The Gothenburg clay was difficult to analyse due to its lower conductivity that created many charging artefacts.

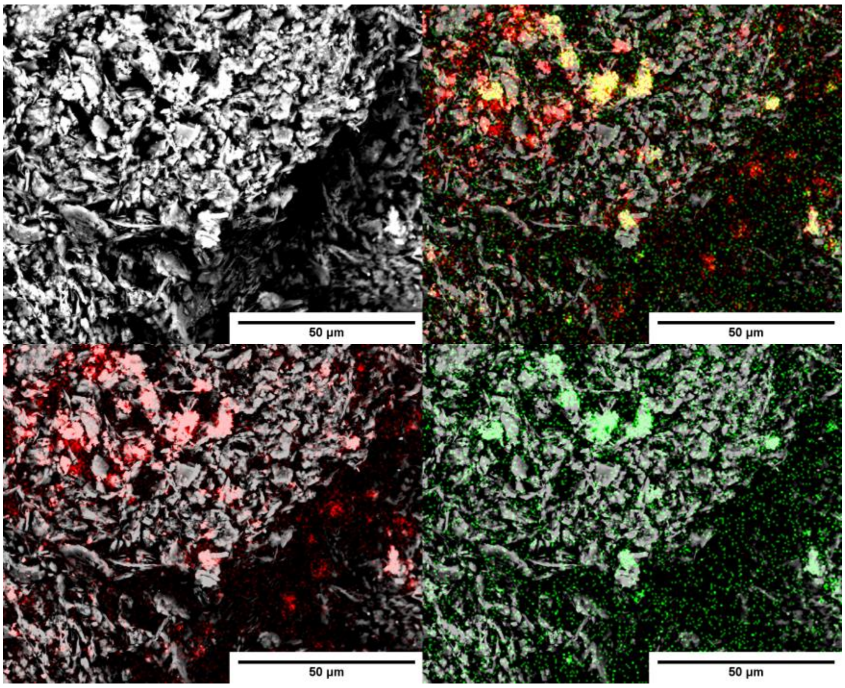


Figure 50. SEM-photo of contaminated kaolin clay with 10% SiO_2 *and 10%* $Ca(OH)_2$*. Red is calcium ions, Green is copper ions and Yellow is the overlap.*

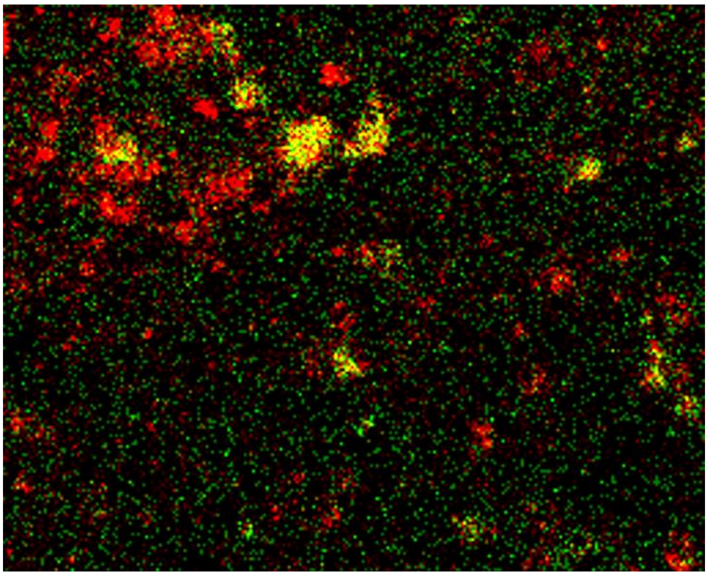


Figure 51. Same SEM-photo as above but just the mapping, here it is evident that copper and calcium follow each other. Red is calcium ions, Green is copper ions and Yellow is the overlap.

5.2. Remediation in practice

5.2.1. Answers from the questionnaire and interviews

27 of the 39 site managers answered the questionnaire, and the results are summarised in Table 23. The percentage corresponds to how many that had the same perception/answer, and it is this answer that the majority have answered, some did also comment their answers. Note the fact that no one mentioned *in-situ* methods, and that remediation for many refers to excavating the soil, this explains how the situation is in Sweden, and the lack of implementation of *in-situ* remediation.

Table 23. Answers of questionnaires for Skanska and Svevia.

<u>Asked question</u>	<u>Answer:</u> Skanska	<u>Answer:</u> Svevia
Have you ever done remediation that was not necessary, based on regulations and guidelines?	**Yes (63%)** **No (37%)** <u>Comments:</u> *Large areas are judged based on small samples, so more remediation than necessary could be done.*	*Yes, particularly in cities, where concentrations are above KM/MKM but below MRR.*
Do you experience that working with remediation is complicated, due to regulations and laws?	**Yes (72%)** **No (14%)** **Too little knowledge to answer (14%)** <u>Comments:</u> *Could be invasive plants that starts to grow in the excavated masses and thus the masses become hazardous waste. Have to hire an environmental consultant.*	*No, we have never experienced any problems with these things.*
Is the work with remediation sustainable (environmentally, economically and social), or could it be improved to be more efficient?	**Not sustainable/could be improved (91%)** **Too little knowledge to answer (9%)** <u>Comments:</u> *Could be improved, too little knowledge in the sector and unclear what happens with the material. Not sustainable since the regulations are so tough that it is difficult to dispose the soil.* *Not sustainable, the landfills have high demands and there is little help to get. Should be more guidance by having different standardized remediation options depending on the type of construction and contaminant. The reuse of material must be better, as well*	*The implementation of MRR- values resulted in that much more soil were disposed, even though if it was of good quality. It is also difficult to re-use the soil since many authorities think that the MRR-values should be applied, hence more remediation is needed.*

	as reduce the number of transports. Very ineffective.	
What is your general opinion on the regulations and laws regarding remediation? Is it good/bad, complicated etc.?	**Complicated/unclear/not good (80%)** **Too little knowledge to answer (20%)** <u>Comments:</u> *Should be clearer and more guidance, a handbook is needed. Very unclear, everyone seems to have their own perception with different values in different projects. Time consuming. Has to be less administration, since it increases the risk of something is done in the wrong way and also makes people cutting corners.*	*It is too cheap to use dispose soil at landfills, and we would like to see more usage of in-situ methods.*

The answers from the governing authorities are summarised in Table 24.

Table 24. Answers from interviews of the City of Gothenburg and the Swedish EPA.

Asked questions	Answer: City of Gothenburg	Answer: The Swedish EPA
What, and how, are You doing to make remediation more sustainable, *i.e.* to use more sustainable methods?	We are controlling that remediation and contaminated soil is reported and that the concentration of contaminants meet the guideline values from the Swedish EPA. We are also accepting/declining site-specific guideline values. However, we are not regulating or controlling remediation alternatives or have stricter guideline values than stated. Thus, we have no specific strategy for achieving a higher usage of sustainable remediation. It should be stated that much of the remediated areas in the city are going to exploited hence little usage of *in-situ* methods since the soil is excavated.	When planning for remediation, one major criterion is what alternative there is to not use dig and dump. Also, we are currently running a network to improve the knowledge of sustainable remediation methods to municipalities, “Collaboration for innovation”. This is a research project to achieve the guidance for achieving sustainable remediation.
What is your general perception on how construction companies are dealing with remediation and how sustainable it is today?	Overall, most of the construction companies are dealing good with remediation and an environmental consultant is often hired. It happens though that smaller companies are not aware of their responsibilities and present laws, resulting in potential mistakes.	Soil masses is a big concern during construction, and it must be a higher degree of circular material usage. Though it is important that this does not result in new contaminated areas. We think that *in-situ* methods can be used if the planning and collaboration were better,

		between site managers, experts and authorities. There is a slight positive trend where the use of *in-situ* methods are increasing, but there should be a much higher usage than the present.
Do You experience complaints or other comments from users, that remediation is difficult to implement?	There is not consensus between us and companies regarding remediation responsibilities and registration duties when remediating. There are misunderstandings when to report and remediation responsibilities when not doing “dig and dump’. This needs to be clarified to ensure better knowledge at companies. Questions about the relevance to dig up and dispose soil where the concentration of contaminants are exceeding the guideline values by small margins. More and more companies are using the amount of unnecessary transports as an argument for keeping contaminated soil in the ground.	Yes, we receive a lot of comments regarding the management with soil masses.
What do You think is needed to achieve a more sustainable remediation?	We have, as mentioned earlier, no responsibility to increase the usage of sustainable remediation. However, it could be argued that there are flaws in the regulations where they could include more sustainability, and could be applied in our supervisory work as well.	More knowledge and spreading of knowledge are needed. Also, instruments given by us, to help. There has to be more collaboration between authorities and the one responsible for the remediation. Lastly, good planning and foresight are key to have a sustainable remediation.
Do You experience any problems with remediation, *e.g.* that companies are taking shortcuts and “cheats”, or that the sustainable management is low?	No, not to our experience.	Not to our experience and what we have seen, but there is a potential to make the remediation more sustainable.

6. Discussion

Here are important aspects discussed, such as problems and uncertainties in the methodology, quality of the results and a summary of the sustainability aspect.

6.1. Uncertainties in the methodology

The methodology was developed taking account previous studies in the related literature. The most common practice in literature in sample preparation by compaction and curing in sealed conditions. Nevertheless, the *in-situ* conditions are not represented in this way. Therefore, three different curing procedures were tested. It is demonstrated that the preparation method affects the mechanical characteristics of the samples. A possible explanation is the hydration conditions in the sample.

The fully submerged samples showed more repeatable results, as well as the consolidated samples. This way the amount of water available for hydration might be more equal for the samples. Having an overburden pressure should minimise the amount of micropores, resulting in more similar samples.

The water content of all samples was examined and no significant difference was observed among different samples, as seen in Table 10. However, the distribution of humidity, and thus hardening, could differ depending on the preparation method.

Due to the fast hardening process of the additives, the slurry could not be mixed by a blender, this since it would be too stiff and not be broken down and mixed.
The mixing procedure is also very important to get an even distribution of the additives. By studying the colour of the contaminated clay, it was evident when a homogenised mixture was achieved, since the colour was even throughout the mixture. Mixing for a long time did also minimise the risk of having an uneven distribution.

To examine the uniformity of all samples, the bulk density, water content and pH were measured (Table 9). Several water content measurements could be taken from the same sample to ensure uniform water content distribution, this would be relevant for the "boxed" samples since the water content distribution might differ between the samples.

6.2. Evaluation of the results

6.2.1. Influence of curing method

How the curing method affected the result is summarised in Table 25. For all cases, aging increased the strength and stiffness of kaolin and natural clay.

Table 25. Peak axial stress on kaolin clay for different storing conditions, based on 7 days ageing and 10% $Ca(OH)_2$ and 10% SiO_2.

Storing Condition	**Peak Axial Stress [kPa] Uncontaminated**	**Peak Axial Stress [kPa] Contaminated**
“Boxed”	55 ± 17	33 ± 7
Submerged	19 ± 1	19 ± 3
Consolidated	31 ± 3	39 ± 4

As seen in the table above, the saturation and storing of the samples plays a significant role in the hardening process, where the results greatly differ between the three different types of storage. Firstly, the “boxed” samples, yielded the least precise results, but also different behaviour compared to the consolidated samples. The submerged samples performed similarly and had the lowest scatter in the results, however, since there only were two samples of each type, the scatter could be larger when having more samples. Further, the consolidated samples showed on a different behaviour, where the contaminated samples had higher strength and stiffness than the uncontaminated, compared to the two other storing methods that showed on the opposite behaviour. However, consolidating the samples should give more equal curing conditions and better contact between the additives, as well as the contaminant, and the clay. Therefore, and since this is the case in the field, these results are to be taken as the better ones.

The highest peak values are found from the partially submerged samples and the reason for this is most likely due to a more optimal water content and curing process. The submerged samples had very low strength and this behaviour is similar to what Horpibulsuk *et al.* (2013) showed, where soaking samples resulted in roughly 50% less strength at seven days of curing. So, by having an optimal water content with enough water for hydration but still having low enough repelling forces to not disturb the reaction process between the binders, would be ideal. Further, the water content was almost identical for all samples, only the consolidated samples had slightly lower water content due to consolidation. However, the water content can differ throughout the sample, this was noticeable for some of the partially submerged samples where they felt more dry in the centre and wetter on the edges. Here, the consolidated samples had probably a more equal water content distribution. So, having samples that were drier in the centre would result in a stiffer and stronger soil, compared to a wet clay, almost failing of its own weight. This is also the main reason for the large scatter, that samples were cured unevenly due to the amount of water in the box and the contact between the wet cloth and the samples. That is also why the consolidated samples performed more consistently and were more easily repeatable.

6.2.1.1. Effect of overburden pressure

Having an overburden pressure while curing increased the peak strength of the kaolin clay, in-line with Taiyab *et al.* (2012). The increase was however more significant at 105% for the contaminated and 63% for the uncontaminated samples, after seven days of curing.

6.2.2 Effect of contamination on consolidated samples

Overall, the results correspond well with the theory in the literature regarding metals' binding capability to clay, its reaction with calcium and the effectiveness of the pozzolanic reaction. Firstly, and foremost important in terms of remediation, all tested samples yielded a high encapsulation of copper, this due to the increase in strength and high pH, due to the additives. A summary of the results is presented in Table 26.

Table 26. Summary of the peak axial stress for the consolidated samples, note the increase in strength for the contaminated kaolin clay. Stabilised = 10% $Ca(OH)_2$ and 10% SiO_2.

Curing Age	Sample	Peak Axial Stress [kPa]	Difference
-	Kaolin Uncontaminated	3.2	-
-	Kaolin Contaminated	3.8	+19%
7 Days	Kaolin Uncontaminated & Stabilised	31	-
7 Days	Kaolin Contaminated & Stabilised	39	+25%
14 Days	Kaolin Uncontaminated & Stabilised	52	-
14 Days	Kaolin Contaminated & Stabilised	81	+56%
-	Gothenburg Uncontaminated	17.5	-
-	Gothenburg Contaminated	17.3	-1%
7 Days	Gothenburg Uncontaminated & Stabilised	60	-
7 Days	Gothenburg Contaminated & Stabilised	45	-25%
14 Days	Gothenburg Uncontaminated & Stabilised	98	-
14 Days	Gothenburg Contaminated & Stabilised	50	-49%

The kaolin and Gothenburg clay, consisting of smectite and illite, behaved significantly differently, where the kaolin had ca 20% higher strength when contaminated whereas the Gothenburg clay had similar strength when contaminated compared to the uncontaminated sample.

When stabilised, the kaolin clay had higher strength and stiffness when contaminated compared to the uncontaminated clay. It also gained stiffness faster according to the oedometer test, as can be seen in form of lower compressibility indices at early stages and higher compressibility indices after curing. The Gothenburg clay had much higher strength uncontaminated, and the contaminated samples were noticeably softer.

One possible explanation for this dissimilarity is, as mentioned in Chapter 2.4., due to the different binding potential to heavy metals for the different clays. Kaolinite has a lower CEC, thus also a smaller surface area and a slight unbalanced net charge, compared to smectite and illite that has significantly higher CEC and surface area and have a neutral charge (Mitchell & Soga, 2005). Also, in previous studies it was proven that smectite and illite had higher zeta potential when in contact with calcium ions, and in combination with the high natural content of calcium oxide in illite, it caused a higher buffering capacity and attraction to positive calcium ions (Morris & Zpik, 2009; Young *et al.*, 2012; Uddin, 2017). Therefore, it could be argued that the increase of strength on the kaolin clay is due to the compaction and solidification due to the strong bonding to copper and that most of the metal ions bind to the hydroxyl groups. This was not however evident in the SEM-analysis, where copper bonded to the calcium ions. Due to the high pH, the copper can also bind to the calcium since it can be a cation exchange of double valence ions (Gougar, 1996). Further, since there are less hydroxyl groups in illite and smectite, and that they are located further away from the edges compared to kaolinite, more of the metal ions might bind to the clay's pozzolans or the additives instead (Mitchell & Soga, 2005; Matlok *et al.*, 2015). Since the strength of the Gothenburg clay did not increase when being contaminated, it could imply that the clay binds weakly to the copper ions. Therefore, most of the metal ions might react with either the added calcium or the pozzolans in the clay and cause less amounts of C-S-H and thus interfering more with the hardening process, compared to the kaolin clay, thus making the clay softer when stabilised.

Hence, in kaolin clay there is most likely a cation exchange between mainly H^+ and Cu^{2+} as well as Ca^{2+} and Cu^{2+}, whereas in smectite and illite clay there is a cation exchange between Al^{3+}/Si^{4+} and Cu^{2+} but mainly between Ca^{2+} and Cu^{2+}. Due to the high natural calcium content in illite, a stronger bonding would be achieved for similar concentration of additives, this could be a reason why the Gothenburg clay had a greater increase in strength compared to the kaolin clay when stabilised.

Lastly, based on the SEM-photos, presented in 5.1.5., it is evident that the copper followed the calcium ions in the kaolin clay, since at locations with high copper concentration there is also high calcium concentration. This was expected to occur since the copper oxidised when adding the calcium hydroxide during the sample preparation, according to Equation 3, as seen in Figure 28. Hence, the reduction in calcium ions left to react with silica resulted in lower strength, for the Gothenburg clay. In theory it should be the same for the kaolin clay, since there should be circa 18.5% less calcium hydroxide to bind with the silica, which should result in a softer soil. However, by compacting the sample through consolidation, more copper might react with the hydroxyl groups on the kaolinite, which results in a stiffer and stronger clay. This should therefore explain the different behaviour for the two clay types.

6.3. Sustainability

The above-mentioned impacts of how contaminants in form of heavy metals affect the soil conditions are vital to know before remediating. As described by SGI, Afzael *et al.* (2017) and Opoku (2019), there is a great diversity in people's knowledge and approach to sustainable remediation. This is evident when comparing the different types of answers from the construction managers and the governing authorities. Here, there is a vast majority that thinks that remediation often is referred to as complicated and unsustainable, where the governing authorities is aware of potential flaws and that there are great room for improvement. The answers are in-line with other questionnaires from different countries, as well as previous answers from Swedish surveys, indicating that it is the same problem almost everywhere (Sourani and Sohail, 2011; Opoku, 2019; SGI, 2020). However, the solution to the problem is quite simple, since all valuable knowledge often is available at larger companies but mainly at governing authorities such as SGI and the Swedish EPA. As described by the Swedish EPA, there must be a better collaboration with better understanding for both parties, where the goal is to achieve the smallest environmental impact where both parties are in consensus with each other.

However, in countries such as Sweden where the lowest bid always wins when planning for construction, due to the law of public procurement, there must be a rearrangement where the environmental impact is equally important as the economic value. Trafikverket, which is one of the largest contractors in Sweden, has started to set a value on the environmental aspect, indicating on a change in the negotiation process (Trafikverket, 2020). This is essential for future development since it, as mentioned in 3.5.5., is a low proportion of the remediation projects that are sustainable. Further, since there are vast amounts of contaminated sites still left in Sweden today, it is not too late to make a change. As previously discussed, that having sustainable guideline values, that the remediation should be based on, is essential. This, together with increasing the knowledge in the construction sector, is probably the foremost important aspects when dealing with sustainable remediation. Due to having unsustainable references, in form of misleading guideline values, increases the risk for making unnecessary remediation, this is, as mentioned by the literature but also the answers from the surveys, today's situation. Having to do unnecessary remediation may result in argumentation with the authorities and risk for frustration, thus making remediation something problematic and complicated. However, this is a difficult situation, since having a too advanced handbook for risk assessment to evaluate the need for remediation often requires more knowledge, leading to an even more complicated situation for companies.

A need for guideline values of leachability [mg/L] is necessary to determine the actual risk of a site. When asking the Swedish EPA about why there are no guideline values for this, the answer is that this is somewhat accounted for in the general guideline values. Further, when measuring the leachability it should be related to what risk the concentration poses for the actual risk objects, this concentration is then compared to the guideline values, thus more theoretical. They also state that the usage of guideline values has some disadvantages in terms of controlling the leachability.

The third aspect of achieving higher usage of sustainable remediation methods is that the gain in environmental impact should be represented by a "fictional cost" in terms of money. It has been proven in many projects that reuse of material and the fact that carbon emissions can be converted into money, is directly related to costs making sustainable remediation more costly as a remediation alternative directly on paper, but often much cheaper when all aspects are considered (Chittoori *et al.*, 2012; CL: AIRE, 2013; Eras *et al.*, 2013; Magnusson *et al.* 2015). Having a sustainable remediation includes all aspects, where a sustainable remediation method is chosen based on sustainable guideline values, because if the remediation is not based on sustainable data, how can it then be sustainable in the end?

7. Conclusions and Recommendations of further Studies

Based on a total of 52 tested samples, three different curing and testing methods, it has been validated that heavy metals, in this case copper, have a negative effect on Gothenburg clay but positive effect on kaolin clay. Stabilisation with calcium hydroxide and silica has proven to increase the clay's strength and stiffness significantly. Where the contaminated kaolin clay has higher strength compared to Gothenburg clay which has vastly lower strength when contaminated. Further, based on questionnaires, interviews and a literature study, it is evident that there are problems regarding sustainability management regarding remediation. Therefore, both the aim and the objectives of this study have been answered and the following conclusions can be drawn:

1. The influence of heavy metal contamination in the clay affects the physical and chemical properties of the clay, by binding to the clay itself or disturbing the hardening process of the stabilisation by binding to calcium ions. This has been shown by having higher strength, stiffness and pH for the uncontaminated kaolin clay compared to the contaminated. For the Gothenburg clay the opposite was observed due to its different mineralogy. Kaolinite is positively affected by copper whereas smectite and illite (in Gothenburg clay) are greatly negatively affected by copper. Therefore, it is essential to consider the clay type when performing stabilisation.
2. Preparation and curing conditions can influence the mechanical characteristics of the specimens. The conditions closer to the *in-situ* conditions, *i.e.* fully submerged samples and K_0 stress conditions produce repeatable results.
3. The sustainability management is lacking in knowledge and usage of new technologies. However, there is a wide knowledge in the sector, but it needs to be spread and more widely available in form of better handbooks and more sustainable guideline values.
4. Based on the literature, there is a great lack of using sustainable remediation methods and it must be more beneficial to use sustainable methods prior to old "dig and dump" methods.

Since this project focused on the pure reactions between chemicals, a recommendation for further studies could be to do the opposite, hence using real-world additives, based on these chemicals. Also, study how different types of metals, as well as other contaminants, affects the C-S-H-production for a natural clay. This would give a better insight in how more common additives and soils react to contaminants. This study has proven that "real" clay is affected more negatively for heavy metals than kaolin clay, indicating that there is an important aspect to consider, also for other types of contaminants.

To conclude, this study can also be applied to ground reinforcements such as deep-mixing, since it is based on the same theory, where the consequences can be severe if contaminants are not considered. Lastly, this subject along with sustainability management challenges, are very relevant in today’s construction sector.

8. References

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9. Appendix

Appendix A

*Table 27. Layout of the different specimens to be tested, Ref. = Reference sample with just pure kaolin, Ref. 2 = Contaminated reference sample. Note that the oedometer samples were tested from the "Curing started" date and the "Tested" date. * = these samples were submerged.*

Sample	**Cont. (Y/N) [%]**	**SiO_2 content [%]**	**$Ca(OH)_2$ content [%]**	**Curing time**	**Density before**	**WC after**

1	No	10	5	10	1.47	44%
2	No	10	5	7	1.45	45%
3	No	10	10	7	1.48	44%
4	No	10	10	21	1.45	
5	Yes	10	5	10	1.48	45%
6	Yes	10	5	7	1.48	45%
7	Yes	10	10	21	1.47	46%
8	Yes	10	10	7	1.45	44%
9	No	10	10	14	1.43	44%
10	No	10	10	7	1.46	44%
11	No	10	10	14	1.45	44%
12	No	10	10	14	1.44	43%
13	Yes	10	10	14	1.46	45%
14	Yes	10	10	14	1.45	45%
15	Yes	10	10	14	1.43	49%
16	No	10	10	7	1.46	45%
17	No	10	10	7	1.46	44%
18	No	10	10	7	1.48	44%
19	Yes	10	10	7	1.46	44%
20	Yes	10	10	7	1.45	45%
21	No	10	10	14	1.47	44%
22	No	10	10	14	1.45	44%
23	Yes	10	10	14	1.43	46%
24	Yes	10	10	14	1.45	46%
25	Yes	10	5	7	1.48	45%
26	Yes	10	5	7	1.46	46%
27	Yes	10	10	7	1.46	46%
28	No	10	5	7	1.47	45%
29	No	10	5	7	1.49	45%
30	No	10	10	7	1.49	44%
31**	Yes	10	10	7	1.51	42%
32**	Yes	10	10	7	1.47	42%
33**	Yes	10	10	7	1.50	42%
34**	No	10	10	7	1.42	43%
35**	No	10	10	7	1.46	44%
36**	No	10	10	7	1.45	39%
37*	Yes	10	10	7	1.48	44%
38*	Yes	10	10	7	1.48	44%
39*	No	10	10	7	1.45	44%
40*	No	10	10	7	1.48	44%
41**	Yes	10	10	14	3.89	42%
42**	Yes	10	10	14	3.92	41%
43**	No	10	10	14	4.13	41%
44**	No	10	10	14	4.07	42%
Gothenburg Clay 45**	Yes	0	0	7		45%
Gothenburg Clay 46**	No	0	0	7		44%
Gothenburg	Yes	10	10	7		

Clay 47**					
Gothenburg Clay 48**	Yes	10	10	14	40%
Gothenburg Clay 49**	No	10	10	14	42%
Gothenburg Clay 50**	No	10	10	14	43%
Gothenburg Clay 51**	Yes	10	10	14	40%
Gothenburg Clay 52**	No	10	10	7	41%
Ref.**	No	0	0	-	-
Ref. 2**	Yes	0	0	-	-
Gothenburg ref.**	No	0	0	-	-
Gothenburg ref. 2**	Yes	0	0	-	-
Oedo	Yes	10	10		-
Oedo	No	10	10		-
Oedo 7	No	10	10		-
Oedo 8	Yes	10	10		-
Oedo 9	No	10	5		-
Oedo 12	Yes	10	5		-

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