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Development of fast testing procedures for determining the leachability of soils contaminated by heavy metals

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Abstract

In the Netherlands, the use of mildly contaminated soils, both treated and untreated, in civil and public works is regulated by the Dutch Act for Building Materials ("Bouwstoffenbesluit") [1]. By law, the leachability of inorganic contaminants and heavy metals, as determined by the column test (NEN 7343), is not permitted to exceed certain values. Drawbacks of the prescribed test procedure are the relatively high costs and the long time needed for testing. The latter adds to the logistics of soil handling and utilization. Therefore, faster and less costly testing procedures for determining the leachability of soils are required.

Over the last 3 years, an extensive study has been performed on the leachability of Cd, Cu, Zn, Pb, Hg, Ni, As and cyanide in treated and untreated soils and some dredged sediments. In this study and earlier studies 230 samples of untreated and treated soils and dredged sediments were analysed for specific soil parameters, contaminant contents and leachability. The analytical data were accumulated, statistically interpreted and evaluated. Some of these soil samples were selected for geochemical speciation modelling [2], some other soil samples were selected for studies on the effects of thermal treatment on leaching behaviour [3]; the results are reported in separate papers in these conference proceedings.

The study resulted in: (1) a deeper level of understanding of the chemical and physical processes governing the leachability of contaminants from (re-usable) soils; (2) the description of a less time-consuming procedure for assessing the leachability of contaminants from (re-usable) soils. The latter result can reduce the need for temporary storage and consequently can lower the handling and logistic costs considerably.

1. INTRODUCTION

In the Netherlands, the use of secondary materials like mildly contaminated soils, both treated and untreated, is regulated by the Dutch Act for Building Materials ("Bouwstoffenbesluit")[1]. About 1.5 to 2 million tonnes of mildly contaminated soil per year are re-used in the Netherlands and an additional 1.5 to 2 million tonnes of contaminated soils are treated by various processes (mainly wet processes, e.g. classification and flotation, and thermal processes). After treatment, the soil should be suitable for re-use. If a soil is to be re-used, the leachability of inorganic contaminants and heavy metals from that soil, as determined by the column test (NEN 7343), is not permitted to exceed certain values, prescribed by the Dutch Act for Building Materials [1].

Drawbacks of the prescribed test procedures are the relatively high costs and long time required for testing (about 5 weeks). The latter slows down soil handling and delays utilization, resulting in the need for costly temporary storage. Therefore, attemps are being made to develop faster and less costly testing procedures for determining the leachability of soils. Also more insight is required into methods for optimizing soil treatment with respect to leachability.

The objectives of the study described in this paper are:

1. the development of fast testing procedures for determining the leachability of inorganic contaminants from treated and untreated contaminated soils. These testing procedures need to be evaluated against the testing procedures prescribed in the Dutch regulations;
2. the assessment of the effects of soil treatment processes on the leachability of the soil.

Clearly, the second objective overlaps with topics of related papers in these conference proceedings on speciation modelling [2] and leaching behaviour of thermally treated soils [3]. Therefore, the second objective will not be discussed extensively in this paper.

A large number of samples of treated and untreated contaminated soils and dredged sediments have been investigated using laboratory analyses (determination of leaching characteristics with the standard column leaching test and fast leaching tests and leaching-related parameters). The laboratory data gathered have been combined with leachability data from earlier studies [4,5,6] and have been statistically analysed.

2. TESTED SOILS AND DREDGED SEDIMENTS

2.1. Selection of soils to be studied

For this study, samples of suitable treated and untreated contaminated soils and dredged materials were provided by the Dutch Centre for Soil Treatment (SCG), the Dutch Development Programme for Treatment Processes for Contaminated Sediments (POSW) and several soil treatment contractors and soil distribution centres.

About 250 soils to be treated via the Centre for Soil treatment were screened on certain intake parameters. According to a prescreening in 1993, the most frequently appearing inorganic contaminants were: Cu, Zn, Pb, Cd, Hg and cyanide. Initially the study was focused on sets of treated and untreated soils and dredgings which were contaminated with the above-mentioned contaminants (in dredgings Ni was analysed instead of cyanide). The soils studied were selected on the basis of their contaminant levels. Later on, in order to fill in the leachability bandwidth to be studied, an effort was made to obtain soils with elevated leaching levels (around the legal limits for leachability). From then on, non-treatable soils as well as soils re-usable without any treatment were added to the study; soils were selected on the basis of the CEN (TC 292) two-step batch leaching test and arsenic was added to the set of parameters. An overview of the origin and number of samples included in this study is given in table 1.

Table 1
Origin and number of samples included in the full research programme (one set is a combination of untreated and treated soil samples)

	Technology	Number	
Soil			
untreated/treated	wet	19 sets	(n = 38)
untreated/treated	thermal	7 sets	(n = 14)
re-usable soil		14 samples	(n = 14)
non-treatable soil		6 samples	(n = 6)
Dredged sediments			
untreated/treated	wet	2 sets	(n = 6[1])
untreated/treated	biological	1 sets	(n = 2)
untreated/ripened		2 sets	(n = 4)
ripened sediments		2 samples	(n = 2)

remark:
[1] two outputs (coarse and fine granular) and one input (3 samples per set).

2.2 Additional data from other studies

The data set obtained from selected soil samples and laboratory testing was expanded using data from other studies on leaching characteristics of different types of soils. These soils were:

- natural soils [4];
- contaminated soils from the Rotterdam [5] and Amsterdam [6] region.
- the leaching database of the Centre for Soil Treatment (SCG).

In this way a database containing data on contaminant concentration and leachability of a total of ca. 230 samples of soil and dredged material was compiled.

3. LABORATORY TESTING

The following parameters were determined for 86 samples investigated:

- contaminant concentrations: Cu, Zn, Pb, Cd and Hg. Cyanide was also determined in soils treated by wet techniques (classification, flotation, etc.). Ni was determined in dredged sediments; As was determined mainly in non-treatable and re-usable soils;
- leachability of the above-mentioned contaminants using the column test (NEN 7343). Leachability was determined only for contaminants with soil concentrations

above Dutch background levels;
- leachability of the above-mentioned contaminants using the CEN TC 292 two-step batch test;
- availability of the above-mentioned contaminants for leaching (NEN 7341). Availability was determined only for contaminants with soil concentrations above Dutch background levels;
- analysis of specific soil parameters: lutum, fraction < 63 μm, organic matter, cation exchange capacity (CEC), chloride, sulphate, carbonate, phosphate, iron, sulphide, pH and electric conductivity.

4. STATISTICAL EVALUATION AND RESULTS

The data gathered were stored in a database-structure and were statistically analysed with SPSS 7.0 for Windows™, using non-parametric statistics. Correlations between soil parameters and leachabilities (determined by several different leaching tests) have been calculated using the Spearman rank correlation coefficient. The use of non-parametric statistics was preferred, because of the possible existence of outliers and a large number of leachabilities below the detection limits.

Most striking observations are:
- at near neutral soil pH-values and soil concentrations below Dutch intervention levels, heavy metal leachabilities are below Dutch legal re-use standards in more than 95% of the cases;
- at pH < 5, the pH of the soil is strongly correlated with the percentage of the total amount of heavy metal leached (relative leachability of heavy metals);
- leachability of cyanide nearly always exceeds the Dutch legal maximum limits for re-use;
- there is no correlation between typical soil-characterizing parameters (e.g. calcite, organic matter) and leachability;
- wet soil treatment (classification/flotation) and, in particular, thermal soil treatment generally lower the leachability.
- leachability and contaminant soil concentration are not correlated within the relevant concentration bandwidth (background level to intervention level)[7].Figure 1 shows the leachability (column test) versus contaminant concentration for copper.
- the results of the column-test (NEN 7343) and the 2-step batch test (CEN TC 292) show a positive correlation. This is illustrated in figure 2 which shows the correlation between these two leaching tests for copper, which has the highest correlation. However, this correlation has a very large bandwidth, which reduces the prediction precision of the CEN-test in relation to the column-test L/S=10 (which is the reference test in the Netherlands).
- a short column test L/S=1 is also positively correlated with the complete column test L/S= 10 (NEN 7343) (illustrated for copper in figure 3). Although the bandwidth is somewhat smaller, extrapolation from L/S= 1 to L/S= 10 introduces an additional inaccuracy in the predictability, because part of the leaching occurs in the interval between L/S=1 and L/S=10.

Remark: the leachability or emission limit (U1) for the re-use of contaminated materials, as stated in the Dutch Building Material Act [1], differs according to the application depth of the contaminated material. Throughout this paper the value of U1 is related to an application depth of 0.7 m.

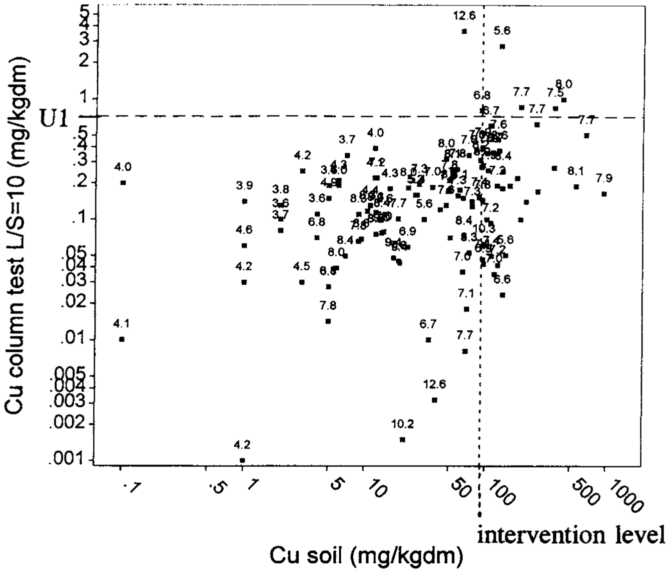

Figure 1. Cu-leachability (column test L/S=10) versus Cu soil concentration (labels are pH-values).

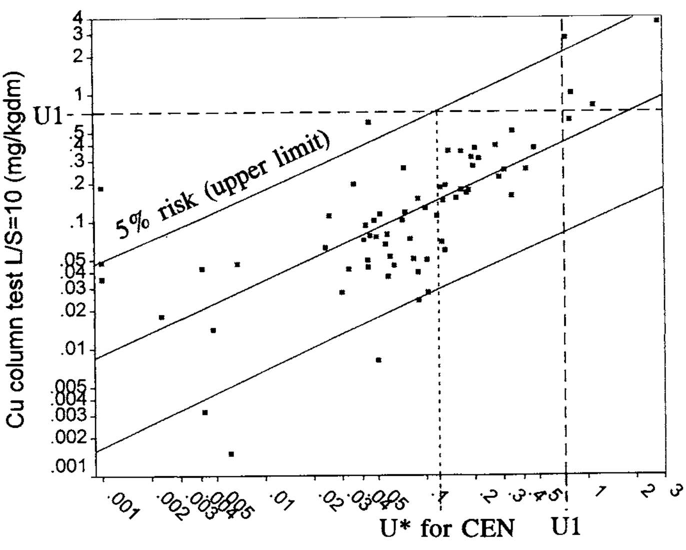

Figure 2. Column test (L/S=10) leachability versus two-step CEN (TC 292) batch test leachability for copper.

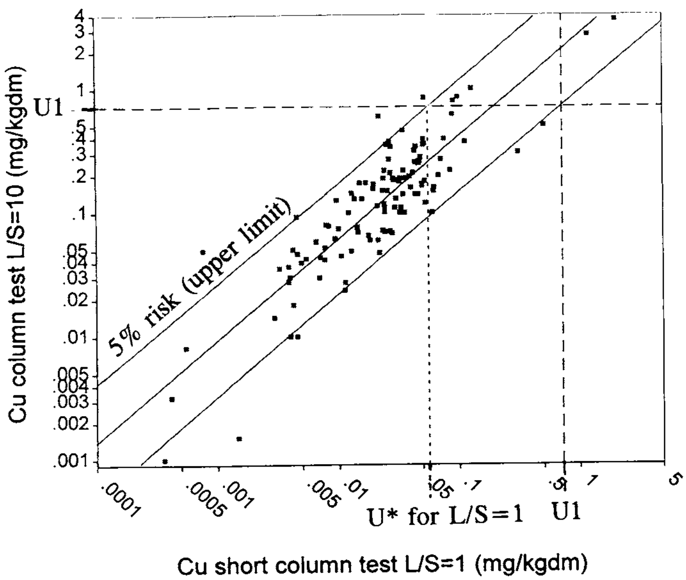

Figure 3. Column test (L/S=10) leachability versus short column test (L/S=1) leachability for copper.

5. FAST PROCEDURES FOR DETERMINING LEACHABILITY

Because the legal maximum leaching limits in the Netherlands are based on the column test L/S=10 (NEN 7343), a fast procedure can only be used as a tool for predicting the results of the column L/S=10 test.

Available options are:

1. a faster test;
2. an empirical model, based on the relation between soil parameters and leachability;
3. testing at maximum contaminant concentration and pH bandwidth.

5.1 Faster leaching test

For this option, two tests are taken into consideration:

- the CEN TC 292 test;
- the column test L/S = 1.

Both tests save about 3 weeks, as compared with the full (L/S= 10) column test.

There is a risk that a fast leaching test result will be below the legal limit whereas the mandatory test result will exceed the legal limit (U1). This risk can be set at an acceptable level: e.g. 5 percent. The results of the fast test should consequently be compared to newly adjusted, derived limits (U*) with a 95% reliability that the result of the mandatory test will not exceed the legal limit (U1). This is illustrated in figures 2 and 3, by the 5% risk (upper limit) line of the linear regression (data are log-transformed to achieve normality). Table 2 gives the derived leaching limits, based on the available dataset, and the percentage of results below the derived leaching limit.

Table 2
Derived leaching limits (U*) for the CEN and column L/S=1 test, below which the result of the mandatory column test with 95% reliability will not exceed Dutch leachability limits (U1).

metal	U1 (mg/kg)	results < U1 (%)	U* CEN (mg/kg)	results < U* CEN (%)	U* L/S=1 (mg/kg)	results < U* L/S=1 (%)
As	0.88	92	0.025	41	0.006	44
Cd	0.032	94	0.00017	79	0.0012	69
Cu	0.72	94	0.12	60	0.056	83
Hg	0.018	90	0.00035	57	0.00032	62
Pb	1.9	99	0.23	86	0.04	90
Ni	1.1	93	0.017	68	0.018	73
Zn	3.8	94	0.7	71	0.38	82

Remark: U1 (and derived U*) are based on emission limits as stated in the Dutch Building Material Act [1] for an application depth of 0.7 m.

The percentages below U* are comparable for both datasets. Note that the percentage below U* is lower than the percentage below U1. This is due to the chosen risk limit of 5%. Soils with a fast test leachability above U* have a risk higher than 5% of exceeding U1 (Dutch leaching limit for re-use) and should be tested with the mandatory column test (L/S=10).

Sufficient data have now been gathered to justify the conclusion that both fast tests can be used for Cd, Pb, Zn and Cu. With regard to As, Hg and Ni and other metals not included in this study, more data are necessary to obtain a more precise prediction.

There is no statistical preference for the one or the other studied option for this fast leaching test. The CEN-test uses the same L/S=10-ratio as the full L/S=10 column test, which means that time-dependent leaching behaviour can be compared more easily. Further the CEN-test is probably better compatible with (developing) European regulations. However, if the result of the fast test does not comply with the derived limits, a full mandatory column test L/S=10 (NEN 7343) should be carried out. The advantage of the short column L/S=1 test is that it is the first step of the full column test and can simply be prolonged.

5.2. Empirical model

The anticipated relation between contaminant soil concentrations and leachability could not be confirmed by statistically significant correlations. A combination of concentrations and soil specific parameters (calcite, organic matter etc.) did not yield significant correlations with leachability either. Therefore it is concluded that an empirical leaching model that uses the soil characterization parameters analysed in this study is not feasible.

5.3. Maximum levels

An examination of the overall database (230 soils including earlier studies [4,5,6], containing As, Cd, Cr, Cu, Hg, Ni, Pb, Zn and cyanide) reveals that only 0 (Cr) to 9 (As) % of all observations on heavy metals and arsenic exceed the Dutch legal leachability limits (U1). This percentage can even be reduced when soils comply with two preconditions:

- the contaminant concentration is below Dutch intervention level;
- the pH of the soil is above 5.

For all soils that complied with these two conditions this percentage decreased to 5% or less. This implies that over 95% of all soils (that comply with the above-mentioned preconditions), meet the standards that Dutch legislation has set for re-use in civil and public works. This finding is in agreement with the results of other recent studies, e.g. a study on the leaching behaviour of zinc in contaminated Meuse sediments [8]. It would seem therefore that leaching tests on soils that comply with mentioned preconditions may be skipped, since in 95% of the cases their leachability will meet Dutch legal standards.

Exceptions to this rule may be:

- thermally treated soils with respect to Mo, Sb, Se; these metals may have a higher leachability in thermally treated soils [3];
- ripened dredged sediments which may have elevated leaching of sulphate, chloride and bromide [9]. However, leachability of these anions is strongly correlated with their concentration, which probably gives a good indication of the result of the column L/S=10 leaching test (NEN 7343).
- soils with cyanide concentrations above background level.

6. CONCLUSIONS

In 95% of the 230 cases the soil samples contaminated with the following heavy metals: As, Cd, Cr, Cu, Hg, Pb, Ni and Zn, with contaminant levels below Dutch intervention levels and pH above 5, showed leaching levels below the legally set Dutch limits for re-usable contaminated soils. It would seem therefore that treated and untreated soils, with heavy metal concentrations below intervention level and pH above 5, in fact comply with the Dutch Building Material Act [1]. However, soils containing other contaminants or soils which do not meet the preconditions for pH and metal concentration must still be tested for leachability.

A fast leachability test can be used to obtain a prescreening. This fast test could be: (1) the 2-step CEN TC 292 batch test or (2) the short column L/S=1 test. The results of these fast tests can be translated into the mandatory column L/S=10 (NEN 7343) test by stating an adjusted leachability level below which the mandatory column test with 95% confidence level will not exceed the legal limits. There is no statistical preference for the one or the other fast leaching test. From the practical and regulatory point of view, both tests have their own advantages and disadvantages.

Apparently, in most cases, a fast leachability determination procedure is possible. There will then be no need for extended temporary storage while awaiting test results. This will result in significant cost reductions for handling and logistics.

7. RECOMMENDATIONS

A leaching protocol for treated and untreated contaminated soils and dredged materials needs to be developed within the framework of a quality assurance system. In this protocol guidelines must be given for situations where no leachability tests are required. This protocol can be based on the results of this study.
The precision of the observed relations and derived reliabilities can be further improved by continued systematic gathering of data of soils and dredged materials, that become available in the future. Special attention should be given to possible critical parameters: oxy-anions in thermally treated soils and sulphate, chloride and bromide in (ripened) dredged materials and cyanide.
The data currently available are reliable enough for applying a fast leaching determination procedure, as described in this paper, to treated and untreated soils contaminated with Cu, Pb, Cd and Zn.

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