

# Leaching characteristic of unbound recycled aggregates: Preliminary study and ongoing research

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## **Abstract**

The environmental aspect of recycling of demolition waste has been studied for years, in terms of different leaching experiments, trying to provide real estimates of the release. The main challenge is the prediction of long-term release, usually described by a leaching model. Extensive leaching experiments in laboratory followed by field site verification are decisive in the development of such models. Prior to 1998 such experiments with respect to recycled aggregates (RCA) have not been carried out in Norway.

In view of these facts the national EcoBuild program funded the three-year R&D project RESIBA (1998-2002), in order to promote the use of RCA for various applications. The present project studied technical and environmental aspects, including a preliminary study in laboratory and field site in terms of leaching characteristics of RCA. At field site the study focused on possible impact from RCA on the soil system. The achieved results verified significant influence, including increased soil-pH and increased amounts of Calcium in the soil water.

The paper will also focus on the ongoing research activities in a national R&D program (2002-2005). This program, funded and managed by the Norwegian Roads Public Administration, is looking at various recycled materials, including RCA in road construction. The working group will investigate possible environmental impact related to the use of recycled materials, including extensively leaching investigation of cementitious waste materials in laboratory, prediction of long-term release in terms of modeling and verification at field site. On this scientific basis, environmental criteria can be proposed for the studied field application.

## **1. INTRODUCTION**

Recycling of construction and demolition (C&D) waste is still at a relatively low level in Norway compared to other European countries like for example Denmark, Germany and the Netherlands. Estimated recycling for C&D waste in Norway is approximately 20 %, which is below the EU average of 25 %. One reason for this is the abundance of high quality and relatively low cost natural aggregates. Another reason is the environmental concern regarding long-term leaching behaviour combined with the difficulties in providing relevant and realistic release estimates for the waste materials.

However, several contaminant leaching experiments for a broad range of waste materials have been reported over the past decade in the literature, including municipal solid waste incinerator (MSWI) bottom ash, coal fly ash, synthetic aggregates and concrete [1-4]. The great number of leaching test methods applied in these experiments have various performances trying to provide relevant release estimates for waste management scenarios. Standard leaching test methods, which often are short-term tests, are applied for waste materials from large scale industrial processes in relation to regulatory control. However, these methods often fail to give a realistic release estimate for a given field scenario resulting in both over- and underestimation of the actual release. Owing to this fact test methodologies have been developed with the intention to reflect the actual field scenario conditions [5].

The EcoBuild project RESIBA-Recycled Aggregates for Construction and Building (1998-2002) was carried out in order to make recycled aggregates (RCA) a competitive product for a range of applications in Norway [6]. Part of the project was a preliminary study of the leaching characteristic for RCA, mainly containing crushed concrete and masonry, including both short-term laboratory leaching and field site experiment. In laboratory the standard test method EN 1744-3 was applied, and the environmental impact on the soil system from RCA used as back-filling material in a utility trench was studied under field conditions. This paper presents the main results from these preliminary investigations.

The results from RESIBA formed the basis of a four-year national R&D program funded and managed by Norwegian Roads Public Administration (NRPA). This project was initiated in August 2001 and will continue until the end of 2005. The main objective in this program is to arrange for more frequent and environmental safe use of C&D waste in road constructions. The environmental impact is studied in terms of elaborated laboratory experiments, modelling and field site verification. A brief overview of the ongoing research activities regarding environmental issues will be given in the present paper.

## **2. EXPERIMENTAL**

### **2.1 Short-term leaching test**

The standardised test method EN 1744-3 developed in CEN/TC 154 was applied in the RESIBA project [7]. This method includes the contaminant leaching of both, natural and recycled aggregates. The individual test samples, collected from the producer of recycled aggregates in Oslo (BA Gjenvinning AS), are listed in Table 1.

Table 1 Listing of test samples and grain sizes.

Sample designation	Bulk composition	Grain size (mm)	Date of sampling
CM1	Concrete and masonry	0-10	June 1998
CM2	“ “	“ “	“ “
CM3	“ “	10-20	“ “
CM4	“ “	“ “	“ “
CM5	“ “	“ “	August 2002
CM6	“ “	“ “	“ “
CM7	“ “	10-38	November 2001
CM8	“ “	“ “	“ “
C1	Concrete	0-10	June 1998
C2	“ “	10-20	“ “
LECA CM <sup>1</sup>	Concrete, masonry and LECA	< 32	October 2001
LECA Ref <sup>2</sup>	LECA	“ “	“ “

<sup>1</sup> LECA (light expanded clay aggregates) lightweight masonry concrete block casted in laboratory with a mixture of 30 % of RCA (0-10 CM) by weight as replacement for natural aggregates, size reduced.

<sup>2</sup> LECA (light expanded clay aggregates) lightweight masonry concrete block casted without RCA (natural aggregates only), size reduced, reference sample.

## 2.2 Leaching at field site

The field site was established in the soil deposits next to a utility trench (Figure 1) parallel to the freeway E18 (Skullerud) outside Oslo. This trench contained a surface water drain pipeline (impervious) with the intention to collect the water from the road surface to a sedimentation basin. RCA was used as back-filling material in the pipe zone of trench. A water permeable fibre membrane prevented contact between the solid materials (RCA and the local soil). The construction was finished in the spring of 1997, and the field site experiment started in the spring of 2000.

Sampling of soil water was carried out by inserting 4 lysimeters (ceramic filters) at sampling point R1, S1, S2 and S3 (see Figure 1). Soil water samples were taken in the spring of 2000 and in the autumn of 2001. During the preparation of lysimeters, soil was sampled at all sampling points (R1, R2 and S1-S4).

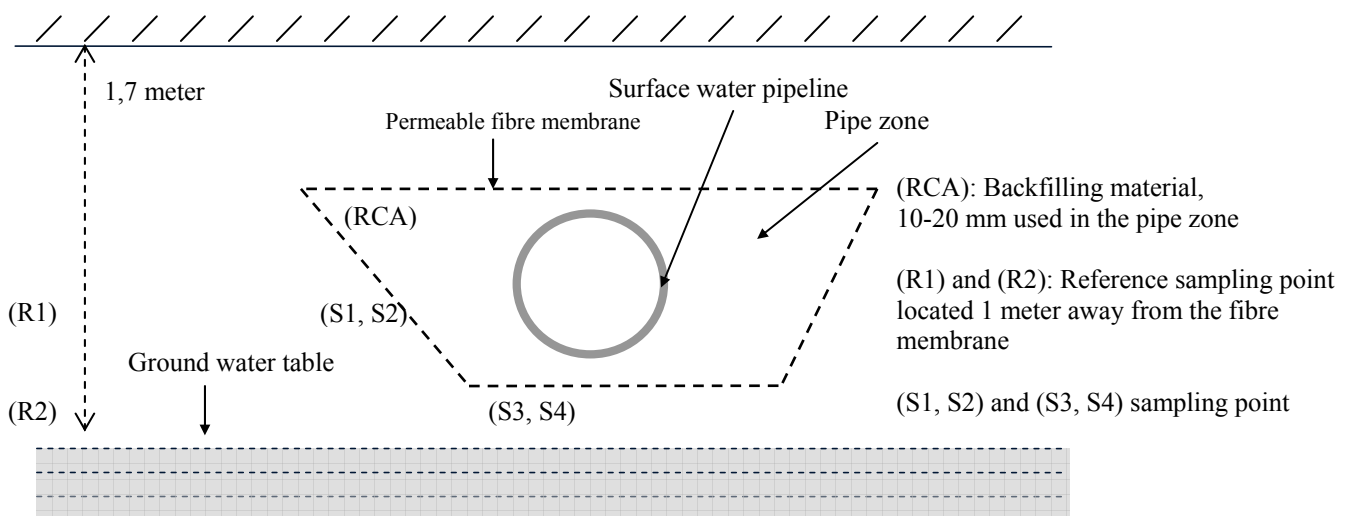


Figure 1 Cross section of the utility trench. All sampling points are located in the unsaturated zone. The fibre membrane

### 2.3 Chemical constituents and analysis.

The following inorganic and organic constituents were determined in the samples: As (arsenic), Pb (lead), Cd (cadmium), Cr (chromium), Cu (copper), Hg (mercury), Ni (nickel), Zn (zinc), Ca (calcium), Mg (Magnesium), PCB (polychlorinated biphenyls) and PAH (polycyclic aromatic hydrocarbons). In addition, the pH in the eluates and the collected soil samples were determined.

The chemical analyses were performed with standard techniques as reported in the literature [8, 9]. Atomic absorption spectrometry (GFAAS and FAAS) and inductively coupled plasma atomic emission spectrometry (ICP-AES) were used to determine the content of As, Pb, Cd, Cr, Cu, Hg, Ni, Zn and Ca. For the determination of PCB and PAH, gas chromatography (GC) was used.

## 3. RESULTS AND DISCUSSION

### 3.1 Short term leaching of Ca and Mg

Figure 2 shows the leached amounts of Ca, Mg and the leachate pH after 24 h. According to the standard (EN 1744-3), the leachant was demineralised water. Due to the alkaline nature of the test material (size reduced concrete and masonry) and the absence of buffering capacity in demineralised water, a large increase of the pH in the leachate was expected. This obvious fact was also confirmed by the results. Consequently, the extent of increasing pH can be related to the leached amount of Ca. A significant increase in the release of Ca with increasing pH confirms this relation, as shown in Figure 2.

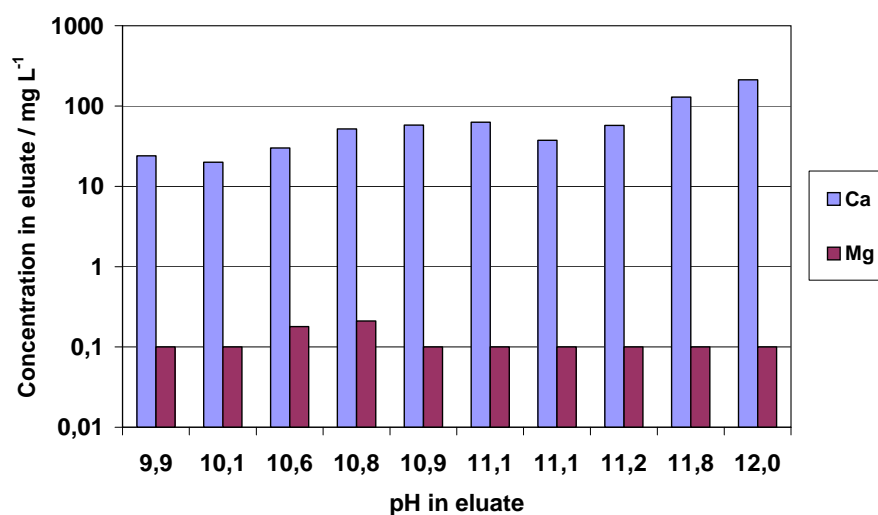


Figure 2 Eluate concentrations of Ca and Mg versus pH after 24 h leaching in 10 different experiments.

The released amount of Mg can further be seen in Figure 2. In most of the experiments the concentrations were below the analytical limit of detection (0,1 mg/L). Mg as well as Ca are natural constituents in soil and soil/ground water, and consequently, the establishment of leaching behaviour in laboratory can give valuable information on expected behaviour under field conditions.

### 3.2 Short-term leaching of metals

In the eluate samples only significant amounts of Cu, Cr and Hg were detected, Table 2. Due to the absence of Norwegian leaching criteria, these amounts have been compared to criteria in Germany, Austria and Switzerland [10, 11]. The achieved results in RESIBA did not exceed any of these criteria with respect to the metals of concern.

The authors realize the differences in the test methods in these countries. In Germany and Austria the German test, DEV-S4, is used [12]. EN 1744-3 is a modified version of DEV-S4, differing in the grain size using < 10 mm and < 32 mm respectively [13]. The German test also requires continues motion of the material contrary to the stirring of leachant in EN 1744-3 during the exposure. Still, both methods use 24 h exposure, L/S = 10 and leaching at own pH. The Swiss test is different from the other two methods by performing the leaching at fixed pH (4-4,5) adjusted with CO<sub>2</sub> [14]. Even though, the comparison is thought to be relevant in the preliminary study. The differences pointed out in the previous paragraph will be investigated in the ongoing national R&D program.

Table 2 Leaching of metals, results attained in laboratory.

Sample designation	Eluate concentration / $\mu\text{gL}^{-1}$							
	As	Pb	Cd	Cu	Cr	Hg	Ni	Zn
CM1	< 0,3	< 1	< 0,2	29	32	< 0,1	< 10	< 50
CM2	< 0,3	< 1	< 0,2	31	33	< 0,1	< 10	< 50
CM3	< 0,3	< 1	< 0,2	4,0	7	< 0,1	< 10	< 50
CM4	< 0,3	< 1	0,4	< 2,0	7	< 0,1	< 10	< 50
CM5	< 1,0	< 10	< 0,5	11	35	0,03	< 5	8
CM6	< 1,0	< 10	< 0,5	13	15	0,02	< 5	5
CM7	< 1,0	< 10	< 0,5	< 5,0	< 5	< 0,05	< 5	< 5
CM8	< 1,0	< 10	< 0,5	< 5,0	< 5	< 0,05	< 5	< 5
C1	< 1,0	< 10	< 0,5	< 5,0	30	< 0,05	< 5	< 5
C2	< 1,0	< 10	< 0,5	< 5,0	< 5	< 0,05	< 5	< 5
LECA CM	< 1,0	< 10	< 0,5	46	39	0,16	6	< 5
LECA Ref	< 1,0	< 10	< 0,5	23	43	< 0,05	< 5	< 5

Due to the risk of contamination when performing analysis in low concentration ranges (ppb), caution has to be taken during each step in the method, including sampling, leachate preparation, exposure and analysis. This subject was carefully considered, and the procedure was carried out without test material (blank sample) in order to check possible contamination from the equipment used. No contamination was found with respect to the metals of concern. More elaborated methods are studied in the ongoing R&D program, including detailed study of the leaching effect from different grain sizes using different grinding equipment. Accordingly, the contamination risk will furthermore be studied.

### 3.3 Short-term leaching of PAH and PCB

EN 1744-3 was originally developed for leaching of inorganic constituents. At the time RESIBA was initiated (1998), suitable test methods concerning leaching of organic compounds from demolition waste did not exist. As a consequence, PAH and PCB were determined in the same eluate samples as for the inorganic constituents.

The analytical limit of detection for the determination of PCB in eluate was 0,005 µg/L. PCB was not found in concentrations above this limit. Contrary PAH was detected in most of the eluate samples. These concentrations are given as the total amount of the 16 most common PAH compounds according to the USEPA (United States Environmental Protection Agency). The leached amounts of PAH were in the range 0,20 µg/L - 1,60 µg/L.

The drinking water quality guidelines in Norway regulates the total concentration of the following 4 PAH compounds [15]:

- benzo(b)fluoranten
- benzo(k)fluoranten
- benzo(ghi)perylene
- indeno(1,2,3-cd)pyren.

The maximum concentration according to Norwegian regulation is 0,1 µg/L. The achieved results regarding only these 4 PAH compounds were below 0,02 µg/L. Further investigation on this subject are already in progress.

### 3.4 Ca and Mg in soil water

The intention with the field site experiment was to prove any influence on the soil system caused by RCA exposed for 3 years in the trench. This was carried out by investigating the soil system adjacent to the RCA (S1-S4) and the reference sampling points (R1 and R2) located some distance away (1 meter), in order to demonstrate possible accumulation of the constituents of concern, see Figure 1.

In Figure 3 the concentrations of Ca and Mg in the soil water samples are shown. Significant differences between S1-S3 and R1 were only found with respect to the concentration of Ca. Mg appeared approximately in the same concentration in all samples. Similar results were achieved in both sampling periods (spring 2000 and autumn 2001).

A thorough investigation of the soil-pH was carried out to complete the measurements of Ca and Mg. The results are shown in Figure 4, and indicated significant increase in soil-pH for S1-S4 compared to the references R1 and R2. This clearly proves the influence of RCA with respect to leaching of alkaline substances such as Ca species [Ca(OH)<sub>2</sub> and CaCO<sub>3</sub>]. Considering the facts that Ca and Mg are natural constituents in soil and their concentrations are in the same order of magnitude, the achieved results demonstrate the influence from RCA on the soil system in terms of the Ca leaching and increased soil pH.

The measurements of Ca and Mg at field site are in accordance with the short term leaching in laboratory. In future field site experiments effective diffusion coefficients can easily be assessed by more detailed study of the ratio between these constituents. Ca can be considered as a natural tracer, which is easily determined regarding release from cementitious waste materials.

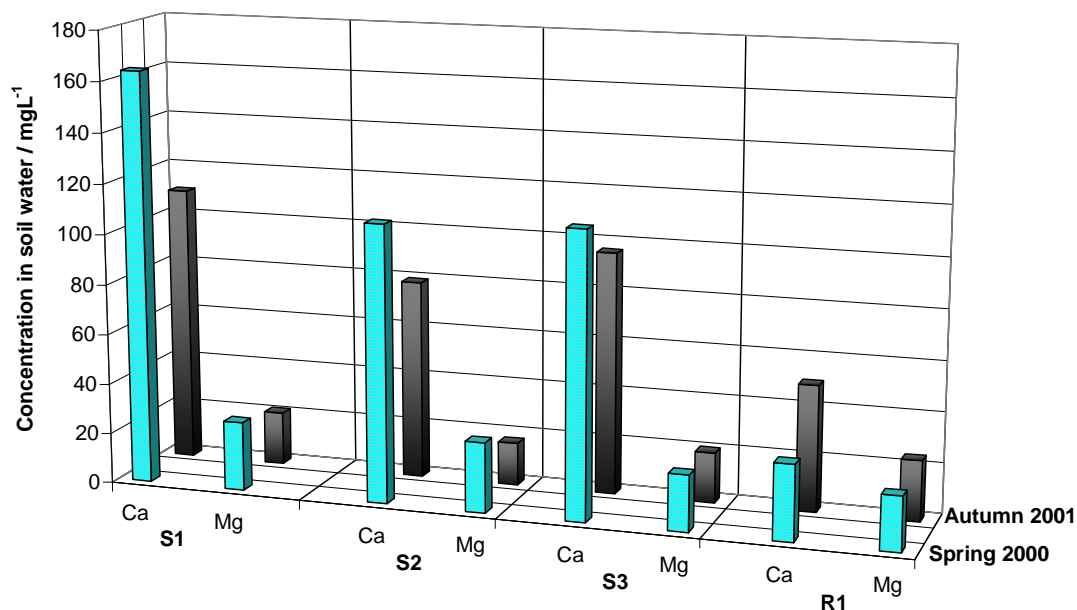


Figure 3 Concentration of Ca and Mg in sample S1-S3 and in reference sample R1 in soil water. Samples were taken in the spring of 2000 and the autumn of 2001.

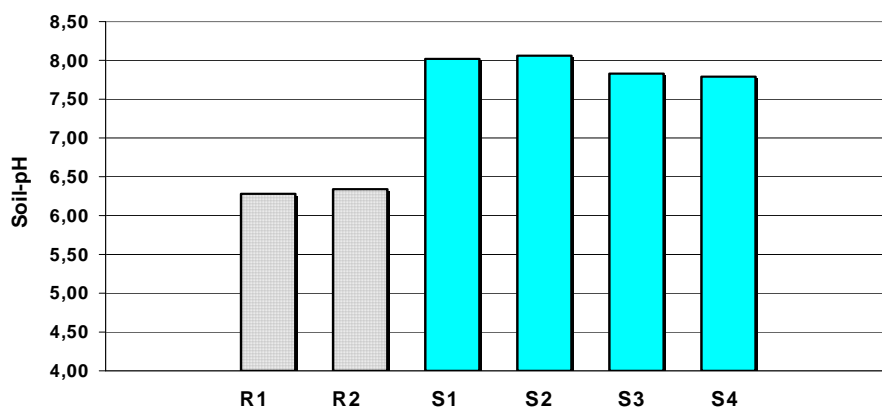


Figure 4 Soil pH in S1-S4 and in the references R1-R2.

### 3.5 Inorganic and organic constituents in soil water

Concentrations of the constituents in S1-S3 are compared to R1 and typical concentrations in Norwegian surface water, shown in Table 3 [16]. The results achieved spring 2000 for S1-S3 indicated increased values only for PAH and Zn compared to R1. Considering typical values in surface water only the results for Zn were above these values.

Results from the autumn of 2001 were almost similar to the results achieved the year before except for Zn, Hg and PAH, (Table 3). In this sampling period lower Zn values were found in S1-S3 compared to the reference, indicating a large spread in concentration of this element. In the case of Hg, increased values for both S1-S3 and R1 were found. The concentrations of PAH were above the values for both R1 and typical concentrations in surface water.

Due to the large spread in Zn values and the increase of Hg in both sampling areas (S1-S3 and R1), the difficulties in finding reasonable conclusions regarding Zn and Hg are obvious. In the case of PAH, laboratory experiments showed significant release of this compound. Even so, no conclusion can be drawn at the present stage considering the fact that soil water samples are related to uncertainty, specially regarding organic compounds such as PAH. In addition, the total content of PAH in the soil samples were found at normal level as will be shown later.

Table 3 Inorganic and organic constituents in soil water

Parameter	Concentration / $\mu\text{gL}^{-1}$				
	Spring 2000		Autumn 2001		Surface water <sup>1</sup>
	S1-S3	R1	S1-S3	R1	
As	< 1	< 1	< 1	< 1	NM
Pb	< 10	< 10	< 10	< 10	< 1 - 8
Cd	< 1	1	< 0,5	< 0,5	< 0,1 - 0,4
Cu	< 5 - 26	62	12 - 41	56	3 - 15
Cr	< 5	< 5	< 5	< 5	< 1 -12
Hg	< 0,01	0,01	0,01 - 0,20	0,89	< 0,20
Ni	< 5 - 7	23	< 5 - 24	42	< 1 - 10
Zn	< 5- 237	25	7 - 26	151	< 5 - 140
PCB	< 0,05	< 0,1	< 0,05	NM	NM
$\Sigma$ PAH	< 0,1 - 0,19	< 0,1	< 0,30 - 1,6	< 0,30	0,1 - 0,8

<sup>1</sup> Chemical constituents in surface water in Norway [16]

<sup>2</sup> Not Measured

### 3.6 Inorganic and organic constituents in soil

The ability of soil to adsorb metals and organic components such as PCB and PAH is well documented [17]. In polluted soils these substances can often be detected. In view of this fact, soil samples were examined in order to prove any possible accumulation. The results are given in Table 4, and were compared to the soil requirements in sensitive areas with respect to human exposure [18]. With few exceptions no significant difference was found between S1-S4 and R1-R2. Compared to the soil requirements, only As was found in significantly higher amount. Since no significant difference between S1-S4 and R1-R2 was found for As, these concentrations could be natural background level or originating from other sources than RCA.

Table 4 Total content of different substances in the soil samples.

Parameter	Total content / $\text{mg kg}^{-1}$						Regulated value <sup>1</sup>
	S1	S2	S3	S4	R1	R2	
As	3,6	3,2	2,7	2,8	3,1	2,5	2
Pb	15	20	12	13	21	14	60
Cd	0,1	0,1	0,1	0,05	0,1	< 0,02	3
Cu	13	15	10	13	22	15	100
Cr	23	27	23	24	28	28	25
Hg	0,019	0,020	0,014	0,017	0,039	0,015	1
Ni	21	21	18	20	22	26	50
Zn	72	68	55	20	85	66	100
PCB	< 0,01	0,01	< 0,01	< 0,01	< 0,01	< 0,01	0,01
PAH	2,5	0,14	0,04	0,2	0,24	< 0,01	2

<sup>1</sup> Soil criteria in most sensitive areas i.e. areas where people are exposed by defined routes. Norwegian Pollution Control Authority issues these values [18].



### **3.7 Ongoing research activities**

One of the objectives in the ongoing national R&D program carried out by NRPA is to develop a fast and convenient risk evaluation method regarding different waste management scenarios in road construction. The work is proceeded within the frames of Norwegian and European regulations [18, 19]. Waste management today requires case-to-case evaluation and this seems to be an obstacle for applying waste materials. The working group 2 in the project decided to use the European methodology guideline ENV 12920 combined with national risk evaluation guideline SFT 99:01 issued by Norwegian Pollution Control authority [18, 20]. ENV 12920 focuses on the management scenario in the testing and evaluation of waste material. By applying this standard systematically for different scenarios in road construction, relevant evaluations of the waste materials are achieved. Results from these approaches will be input in the risk assessment using SFT 99:01-guideline in order to make the relevant decisions. Possibilities of using generalized default assumptions contrary to site specific data will be the key issue. Cementitious and glass based materials, shredded tires and recycled asphalt will be tested in the project.

### **4. CONCLUSION**

Leaching results achieved in RESIBA comply with the criteria in Germany, Switzerland and Austria using pr-EN 1744-3 as test method with respect to leaching of metals. Owing to the fact that different methods are used in these countries, further work has to be done in order to make a valid comparison. The eluate concentration of PAH did not exceed the Norwegian drinking water criteria, but detection of other PAH compounds not regulated in Norway results in further investigation.

No influence from RCA with respect to metals, PCB and PAH on soil was found at field site. The only influence on soil from RCA was the release of Ca and consequently an increased soil pH. This was not considered to be a negative environmental impact.

The RESIBA project verified some essential facts regarding recycled aggregates in Norway, but it also gave rise to some unanswered questions discussed in this context. The work needed for achieving the relevant answers, are already in progress in the ongoing national R&D program.

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