

Constituent release predictions for recycled aggregates at field site in Norway

Christian J. Engelsen^{1,4}, Hans A. van der Sloot²,
Gordana Petkovic³, Grethe Wibetoe⁴,
Erik Stoltenberg-Hansson⁵ and Walter Lund⁴

¹ SINTEF Building and Infrastructure. P.O. Box. 123 Blindern. N-0314 Oslo. Norway. Phone: +47 22 96 55 55.
Fax: +47 22 96 57 25. E-mail: christian.engelsen@sintef.no

² Energy Research Centre of the Netherlands. P.O. Box. 1. 1755 ZG Petten. Netherland.
Phone: +31 224 564249. Fax +31 224 568163. E-mail: vandersloot@ecn.nl

³ Norwegian Public Roads Administration. P.O. Box 8142 Dep. N - 0033 Oslo Norway. Phone: +47 22 07 35 00.
Fax + 47 22 07 38 66. E-mail: gordana.petkovic@vegvesen.no.

⁴ University of Oslo. P.O. Box 1033 Blindern. 0315 Oslo.
Phone: +47 22 85 54 46. Fax: +47 22 85 54 41. E-mail: grethe.wibetoe@kjemi.uio.no and
walter.lund@kjemi.uio.no

⁵ Norcem A.S. 3950 Brevik. Norway.
Phone: +47 35 51 11 72. E-mail: estolt-h@online.no

Abstract

The leaching properties of cement and glass based recycled material are being studied in laboratory and at field site. In the laboratory the intrinsic leaching characteristics are determined as a basis for geochemical speciation modelling. At field site the chemical and physical properties of the materials are investigated over time. Two types of recycled aggregate, crushed concrete and foamed glass, in addition to crushed natural stone were applied in the road base in three segments of the road, respectively. An extensive monitoring program was established (October 2004) and is still running on site including on line measurements of field pH, net infiltration rate, temperature profiles, moisture content and the settlement. Water samples are taken automatically for chemical analysis.

Metal concentrations in the leachate are being related to the corresponding infiltration volumes and pH in order to evaluate the impact of different flow regimes. These assessments are being used to measure the deviation from the equilibrium concentrations determined in the laboratory. The field site data will give valuable information to the geochemical speciation model that is based on laboratory data.

This paper presents the field site data from the first 14-month exposure period. During this period the leachate pH decreased from 12.8 to 9.5 in the uncovered crushed concrete field and from 12.8 to 11.5 in the crushed concrete fields in the road base covered with asphalt. In the foam glass aggregate field the pH was fluctuating in the range of 7.6 to 8.6. The full scale demonstration project presented in this paper is a part of The Norwegian Roads Recycling R&D Program (2002-2005) [1].

1. INTRODUCTION

Leaching and emission characterisation are the most relevant ways to determine the environmental properties of construction materials in service life as well as after the demolition stage. Focus has been placed on the latter case as the construction and demolition waste (C&D waste) is the major waste stream in most countries. In Norway and Sweden, approximately 60 % of this waste stream is concrete rubble [2]. Potential release of chemical constituents in an application scenario is therefore of great concern as the objective of most countries is to achieve high level of reuse for this complex material stream. The release scenario currently focused in Norway is the road environment. In the Norwegian Roads Recycling R&D Program different recycled materials were characterised as a part of the chosen risk assessment methodology described elsewhere [3]. This includes basic leaching characterisation, geochemical modelling of the equilibrium concentrations and field site verification. Preliminary results from the field site are presented in this paper.

Generally, the partitioning of chemical species between solid and the aqueous phase determines the solution speciation as well as the ability to bind to reactive solid surfaces. Different surface processes can thus be predominating in different pH domains (1-14) including ion exchange, complexation and precipitation [4]. The overall main controlling factor of those processes is the pH, as recognized in earlier studies [5-7]. In order to identify and describe the surface process at different pH sub domains, a thorough characterization of the leaching behaviour is required as input for geochemical speciation modelling. Characterising the leaching behaviour over a wide range of pH is important due to different acid neutralization capacities (ANC) for different materials as well as the external pH encountered at the field site is different. For example, the pH of recycled concrete aggregates depends on the degree of carbonation, which again depends on the service life exposure of the concrete structures.

In the field experiment the two types of recycled aggregate, crushed concrete and foamed glass, in addition to crushed natural stone were applied in the road base in different segments of the road. The objective of the current field site investigation was to monitor the leachate pH and the constituent release under different infiltration regimes.

2. EXPERIMENTAL

2.1 Materials

The materials studied at field site were recycled concrete aggregate (RCA) and commercial foamed glass aggregate (FGA). The former material originated from a section of the highway E6 (25 km south of Oslo) that was constructed with concrete pavement in the beginning of 1980. Due to reconstruction from two to four highway lanes, the concrete pavement was demolished and used at the place where the field leaching experiments were conducted, see section 2.2. The demolished concrete was subsequently crushed and fractionized into a grain size of 20-120 mm and applied in test segments of the road base in the entrance lane to the north bound lane of E6, see Figure 1. The crushing and fractioning of the old road pavement was conducted only weeks after demolition. Cores of the old road pavement were taken before demolition in order to assess the carbonation level and the concrete quality in terms of compressive strength. The commercial FGA material with the grain size of 10-50 mm, delivered by the HasGroup (Norway), was applied separately in the same way as the RCA. In

addition, a test segment of natural aggregates (NA) with the grain size of 20-120 mm in the road base was also included. In Table 1 the different test fields are summarized.

Table 1 Test fields, materials used and function in the field experiments.

Field ¹	Material	Function	Monitoring
F1	FGA	Leachate collection with membrane	Temp, moist, pH and infiltration volume
F2	FGA	Separation of fields	Temp and moist
F3W	RCA	Leachate collection with membrane	Temp, moist, pH and infiltration volume
F3E	RCA	Leachate collection with membrane	Temp, moist, pH and infiltration volume
F4	RCA	Separation of fields	
F5	NA	Leachate collection with membrane	pH and infiltration volume
F6	NA	Separation of fields	
F7 ²	RCA	Leachate collection with membrane	Temp, moist, pH and infiltration volume
F8 ²	RCA	Separation of fields	
F9	RCA + asphalt	Gain experience with the mix of the two materials	Mechanical properties

¹FGA had a grain size of 10-50 mm, the other test fields were constructed with a grain size 20-120 mm in the subbase.

²Not covered by asphalt and located on the east side of the road i.e. no traffic. Grain size of 20-120 mm.

2.2 Description of the field site

The field site was located at the newly built control station of the road authorities (Taraldrud kontrollstasjon) at the highway E6, 20 km south of Oslo. A section of 50 m of the entrance lane from the control station to the north bound direction of the highway was applied for the field leaching experiments as shown in Figure 1. Different materials were applied as subbase materials in the test section (F1-F9). This also included a test field not covered by asphalt constructed with RCA in order to compare with the covered field.

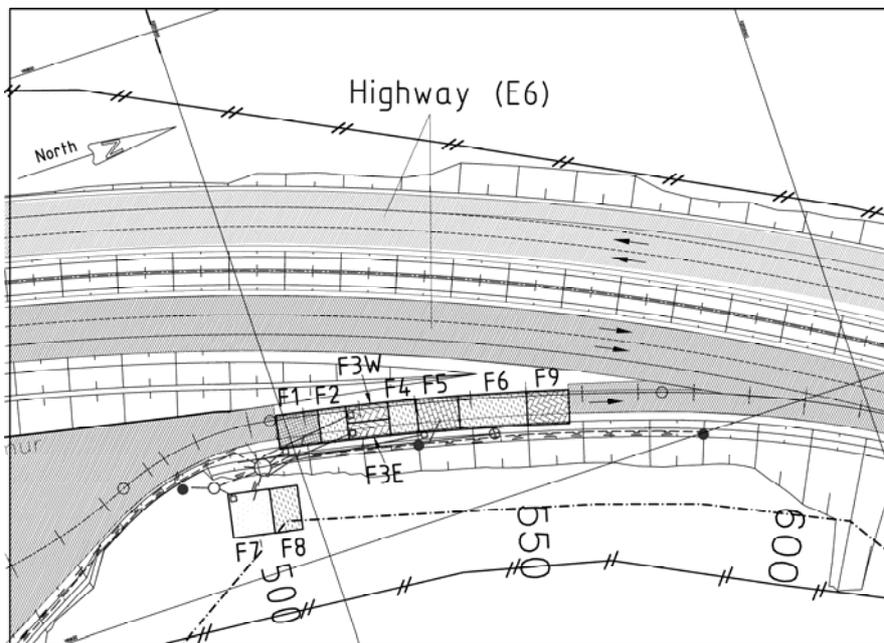


Figure 1 Constructional drawing of the field site. The test fields had a longitudinal fall to the south and a transversal fall to the east.

A cross section of the top structure is shown in Figure 2 and contains the base (top layers of asphalt) and the subbase. A water tight HDPE membrane with drains was placed under the subbase in order to collect the infiltrated water. Each test field was 7.5 m in length (width of 8 m and depth of 0.9 m) and was separated by minimum 5 m to ensure that no cross mixing of drainage was taking place as no membrane was placed under these separation fields. The test road had a longitudinal fall of 4 % and a transversal of 2 %. The drains in the membranes were thus placed in relation to this topography i.e. downstream. In F3 it was chosen to use two drains in the membrane in order to distinguish between two different infiltration rates (due to the transversal fall) as can be seen in Figure 2. This resulted in F3W (west side) and F3E (east side). F1, F5 and F7 were constructed in the same way but with only one drain each. In addition F7 was not covered by asphalt and located to the east side of the road.

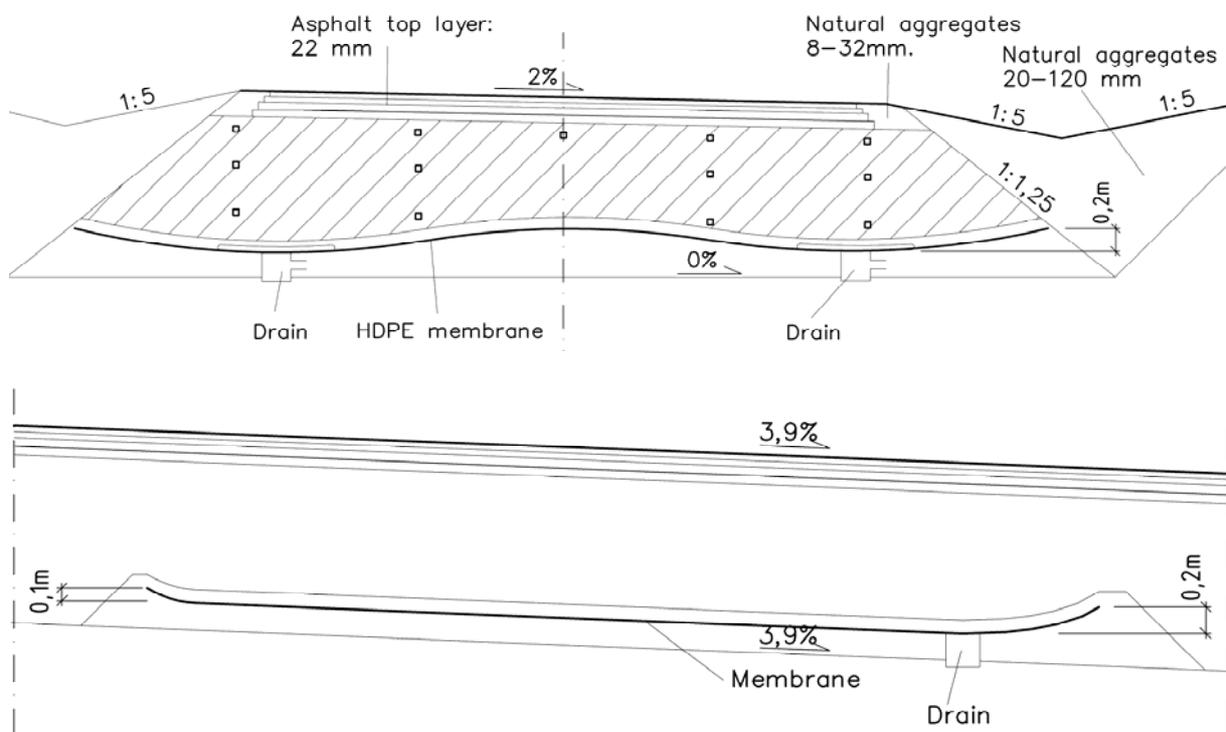


Figure 2 Cross and longitudinal section of the base and subbase. The drawing above: Cross section of the fields F3W (left side) and F3E (right side), the placement of the test material is shown as the shaded area, and open squares indicate the positions of the moist and temperature sensors. The drawing below: Longitudinal section of a test field.

2.3 Monitoring

The drainage water from the different test fields was led in PE pipes to the collection well where it was automatically sampled in separated closed systems. The outlet of each sampling system was connected to Tipping Bucket Flow Gauge (ITAS, Norway) in order to measure the volume collected from each drain. The pH, air temperature, water flow and the precipitation were determined online by using the logging system PC Logger 3100i (Intab Interface-Teknik, Sweden). Temperature and moisture content in the road base were also determined online by connecting the sensors to a Hewlett Packard Logging system.

2.4 Chemical constituents and analysis.

ICP-OES was used to determine major elements (Al, Ca, Si, Fe, S, Ba, Na, K), typical metal cations (Cd, Cu, Ni, Pb, Zn) and elements that under certain conditions form oxyanions (Cr, As, Sb, Se, B, V, Mo). The anions Cl^- , Br^- , SO_4^{2-} and F^- were analyzed by ion chromatography (IC) and the dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) were measured by a Shimadzu carbon analyzer. The ICP-OES, IC and carbon analysis were conducted at Energy Research Centre of the Netherlands (ECN). In eluates not analysed by ion chromatography, Cl^- was determined by titration with silver nitrate using Metrohm 686 Titroprocessor at SINTEF Building and Infrastructure. The concentration of S and P determined by ICP-OES were assumed to be sulphate and phosphate respectively. Sulphate is a major species in geochemical modelling. It is also a major species in the aluminate hydrate phases in the cement paste. Comparison of the sulphur results determined by the two analytical techniques was thus performed, and the results showed in most of the cases less than 15 % deviation. pH measurements were conducted on the computerized titration system Metrohm Basic Titrino 794 and the conductivity was measured with the Hach Sension analyzer carried out at SINTEF Building and Infrastructure. For determination of total element concentrations, FGA and RCA material were pulverised to $< 125 \mu\text{m}$ and decomposed by a mixture of HF and HNO_3 .

3. RESULTS AND DISCUSSION

3.1 Chemical properties of the materials

The element concentrations of the FGA and RCA materials are shown in Table 2. Chemical properties of crushed concrete are to a large extent determined by the properties of the hydrated cement mineral phases. As these hydrate phases are stabilized by the alkali hydroxides at the high $\text{pH} > 13$ in the concrete pore water, the carbonation in service life and leaching of alkalinity in the recycling stage, e.g. road scenario, are largely determining the pH development of the material. The RCA material applied at field site in this study originated from an old concrete pavement covered by an asphalt top layer. Measurements of the carbonation depth prior to demolition and recycling showed insignificant carbonation. This can be seen in Figure 3 as the pH of the material is close to 13. Furthermore, the figure shows the large buffer capacity of the RCA material and the drop of material pH is thus dependent on the leaching rate of alkalinity (also carbonation) which in turn is dependent on the scenario conditions. The ANC curve will thus give a first approximation of the expected resistance to pH drop at field site and is therefore important in the release prediction, as the leaching of elements forming both cations (e.g. Zn, Cu, Pb, Ni) and oxyanions (e.g. As, Mo, Sb, Cr and V) are to be largely dependent on pH in terms of equilibrium concentrations [7, 8].

The ANC for FGA material is different from the RCA as the buffer capacity is small, which is also shown in Figure 3. The external pH encountered at field site will be more controlling for this material than in the case of the uncarbonated RCA.

Table 2 Total element concentrations of the RCA and FGA materials

Element	Total content (weight %)		Element	Total content (mg/kg)	
	RCA	FGA		RCA	FGA
Al	6.25	0.86	Cu	12	163
Ca	6.26	6.35	Ni	14	19
Si	28.32	32.69	Pb	31	841
S	0.24	0.07	Cd	< 0.72	1.29
Fe	2.37	0.28	Cr	69	234
Na	1.95	9.71	As	< 10	32
K	2.58	0.52	Sb	< 3.3	51
Al	6.25	0.86	V	53	16

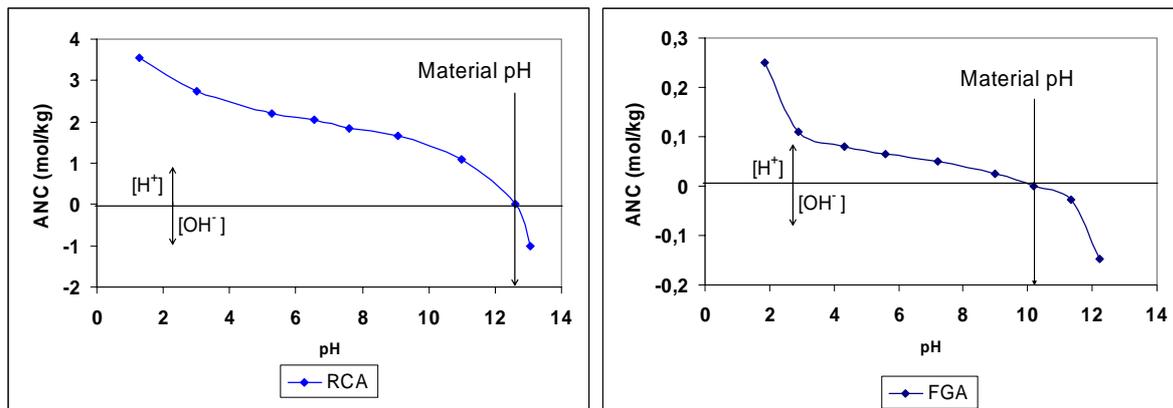


Figure 3 Acid Neutralization Capacity (ANC) determined in laboratory for crushed concrete (RCA) and foam glass (FGA) applied at field site.

3.2 Field pH and infiltration

The pH of the collected infiltrates differed as expected. In the RCA test fields (F3W, F3E and F7) the start pH corresponded to the ANC results from the laboratory tests as can be seen in Figure 4. The pH development, however, differs between F3 and F7 as the latter is uncovered and more sensitive to carbonation. After the first year of exposure the pH is less than 9.5 in F7. Physical examination of the aggregates in this field is currently ongoing and preliminary cross section results showed a significant carbonated layer around the aggregates of approximately 3-10 mm in the upper layer of the field. The examinations will continue for the other fields as well including vertical carbonation profiles.

The pH in F1 (FGA) shown in Figure 4 varied between 7.6 and 8.6 with respect to the monthly average and even more for single measurements. The FGA consist of a glass based structure with low buffering capacity and the pH is thus more dependent of the external pH. Compared to natural aggregates (F5) the pH is higher. However, the FGA materials own pH (material pH) is above 10 which was never observed under real conditions and this shows that the leachate pH is imposed by the external field pH.

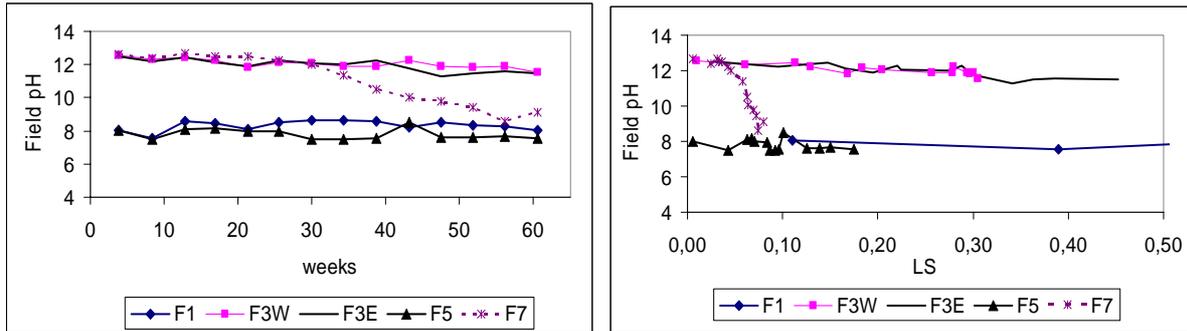


Figure 4 Left: Online pH measurements as function of time in the infiltrates from the different test fields. The curves are based on monthly averages. Right: pH as function of accumulated liquid/solid ratio (L/S). For F1 the LS continued to 4.7.

In Figure 5 the cumulative infiltration volumes are shown. The FGA material showed the highest drainage capacity. It can also be seen that the infiltrated volume increased in the sequence F3W, F3E and F7, as expected from the design of the construction in Figure 2.

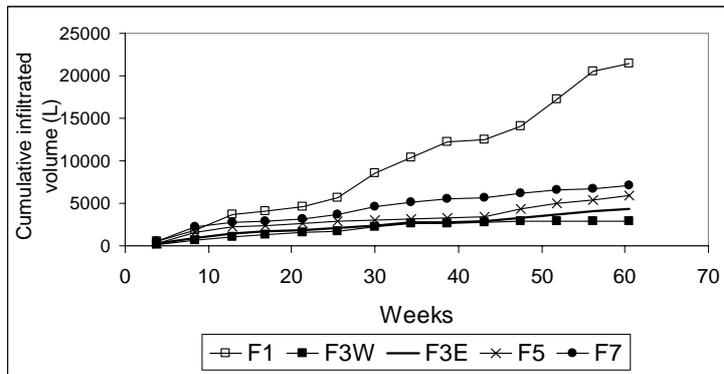


Figure 5 Cumulative infiltration volumes determined in the different fields as function of time

3.3 Release

Water samples were taken periodically taking into consideration the accumulated infiltrated volume and the change in pH. A second selection of these samples was needed due to large number of samples. In these final selected samples chemical analysis were carried out for the constituents specified in paragraph 2.4. For practical reasons the results from F3E will be left out in the following.

3.3.1 Major elements

Major constituents of the paste in RCA are important parameters to measure as the hydrate phases such as calcium silicate hydrates, calcium hydroxide, and calcium sulfoaluminates start to dissolve in a certain order when the pH decreases [9]. The pH static leaching characterization for this RCA material indicated the presence of the sulfoaluminate phases, when compared with the leaching patterns of other RCA materials [8]. At field site the processes are more complex as the decrease in the pH is a combination of carbonation and leaching of the alkaline species and these processes are dependent on the field condition e.g. precipitation (rainfall). In addition, the dry-wet cycles interfere with the physical nature of the material which can change the chemistry of the infiltration water. In order to give a qualitative overview of the field site, the release of Al, Ca, Si and S in F3W and F7 is compared in Figure 6 over the 14-month period. The data were plotted as function of the cumulative liquid/solid

ratio (L/S) defined as the infiltrated volume (L) divided by the mass in contact with infiltrate (kg). The concentrations are related to the total mass in order to give a comparable basis, as the precipitation catchment area and total mass exposed are different for the two fields. It is shown in Figure 6 that the concentrations of Al and S decreased faster in F7 than in F3W. This shows the effect of carbonation as this field was not covered by asphalt and natural aggregates.

It must be emphasized that carbonation and leaching of alkalinity are expected to occur at low L/S (compared to a column experiment) as only the outer layer of the coarse particles plays a role at this relatively early stage. In later stages of the field experiment, the precipitated or leached layer will be changed.

The results from the F1 samples (FGA material) showed significant concentrations (10-100 mg/L) for Ca, Si, and S. In laboratory the acid solubility of this material was determined to 0.3 weight % compared to 15 weight % in the RCA material. Due to less experience with FGA material in regard to modelling of chemical leaching processes, it is at the present stage difficult to explain the leaching behaviour. However, modelling research is under way. From the full pH dependent leaching characterisation in laboratory the pH was found to have small effects on the S and Si equilibrium concentrations.

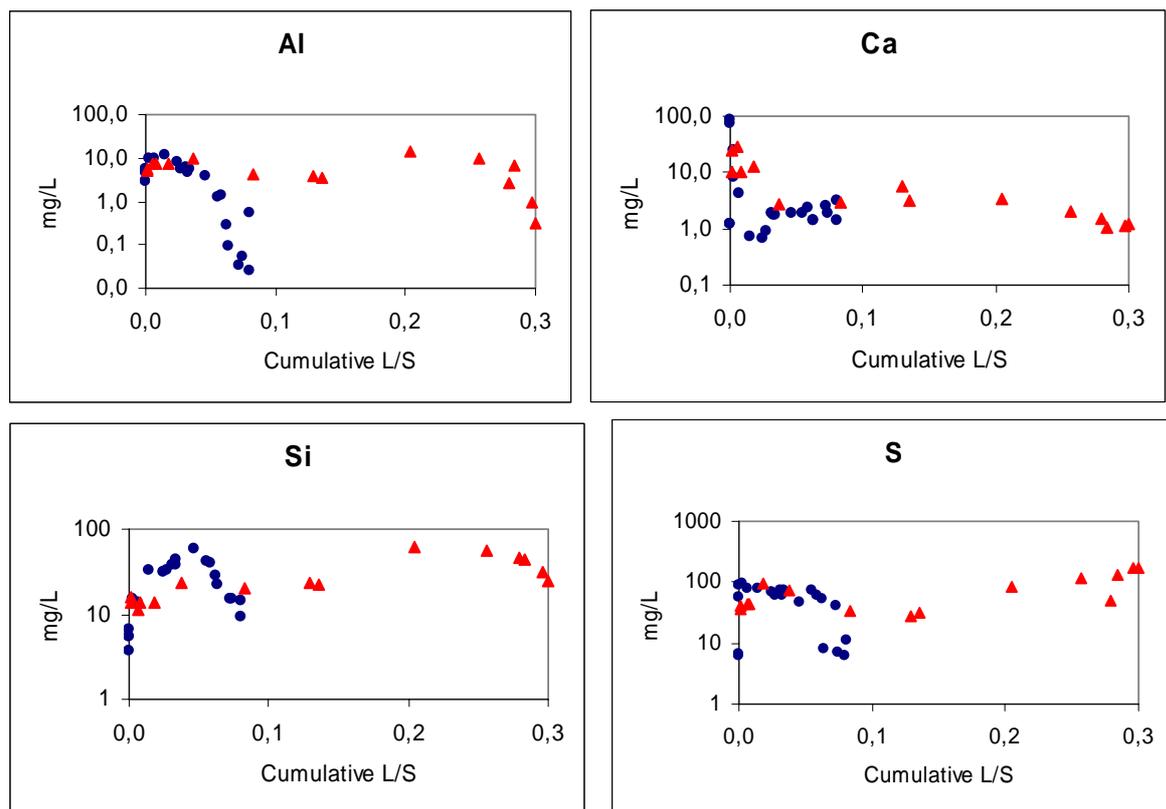


Figure 6 Release of Al, Ca, Si and S as function of cumulative L/S determined in water samples from F3W (triangles) and F7 (bullets) during the first 14-month.

3.3.2 Minor and trace elements

Of the elements that typically form metal cations (see paragraph 2.4) Cu and Ni leaching results were chosen to be presented due to detectable concentrations and leaching trends,

shown in Figure 7. The results are determined from the fields with FGA (F1) and the RCA (F3W and F7). In the fields with RCA a combined effect of carbonation and formation of a leached layer can be seen in F7 (open to air) as the concentration of both elements decreases during the period of 14-month. In Figure 4 the pH of this field was found to decrease significantly after approximately 30 weeks, i.e. after $L/S = 0.05$. This seems to influence the Ni and Cu concentrations after the first “wash out” period and this effect can be seen in Figure 7.

In F3W (in road base), however, the metal concentrations tend to increase after the first exposure period. In the reference field, F5, the Cu and Ni concentrations were lower in the same period; $1-10 \mu\text{g/L}$ and $< 5 \mu\text{g/L}$ respectively. As the field pH of F3W had much smaller decrease compared to F7 (see Figure 4) the fluctuation can be due to the fact that the sampling has been conducted at different infiltration rates in F3W, i.e. at high and low rain fall. This remains to be verified by studying the monitoring data further.

The data from F1 varied in the range $5-100 \mu\text{g/L}$ for both Ni and Cu. The former element seems to fluctuate in a systematic way and further studies of the monitoring data will be carried out. The dashed line in Figure 7 shows the Norwegian limits for pore water concentrations in sensitive land use calculated by K_d values given by Norwegian pollution authorities [10]. None of the concentrations were found to exceed the limits. It is also worth mentioning that the total content of Pb was 840 mg/kg in the FGA material used at field site. Most concentrations measured in F1 were found to be $< 10 \mu\text{g/L}$.

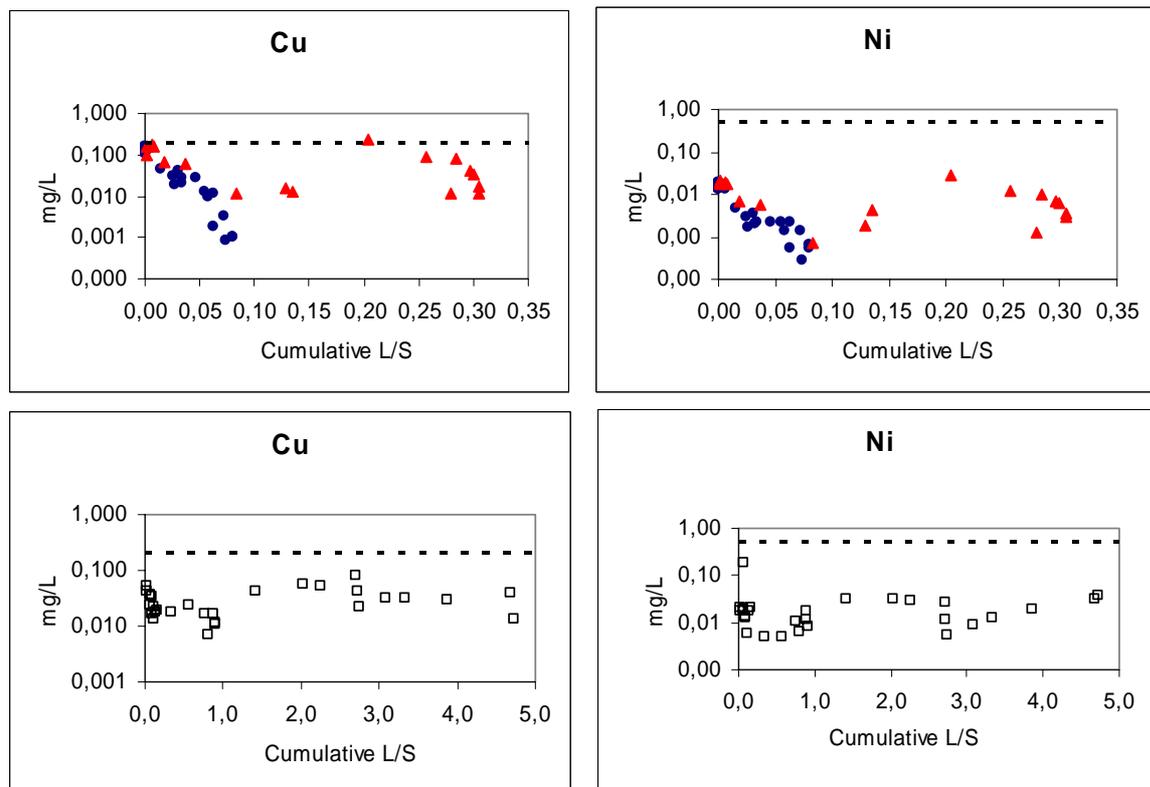


Figure 7 Release of Cu and Ni as function of cumulative LS determined in water samples from F3W (triangles), F7 (bullets) and F1 (open squares) during the first 14-month. Dashed lines indicate the criteria for Norwegian limits for pore water concentrations, based on K_d values.

In oxyanion species the chemical bonding between the element and the oxygen is covalent. Formation of oxyanions is favoured for elements that are close to oxygen in electronegativity, for example the transition metals Mo, Cr and W form MoO_4^{2-} , CrO_4^{2-} and WO_4^{2-} , respectively. Although transition metals exist in different oxidation states depending on the pH and redox conditions, their oxyanions are most stable when the elements are in their highest oxidation states [11].

For cement based products as well as aggregates in concrete, oxyanions are more important than their cationic form because the latter has low solubility in the relevant pH domain. [6]. Moreover, oxyanions are Lewis bases and tend to react with Lewis acids to form oxyanion salts by ionic bonding. The most stable oxyanionic salts are thus formed with alkaline and earth-alkaline metals. In cement systems (alkaline conditions) Ca is one of the major elements in the solid and aqueous phase. Thus, the oxyanions tend to form equilibrium with Ca metallates, i.e. $\text{CaMoO}_4(\text{s})$ and $\text{CaCrO}_4(\text{s})$ in such systems. Moreover, more complex calcium minerals also tend to precipitate as oxyanion complexes/minerals, forming equilibrium with the corresponding anion [12].

Laboratory leaching characterisation and subsequent geochemical modelling of different oxyanion species carried out for different RCA material, including the RCA material used in this field study is presented elsewhere [8]. Of the elements that have the tendency to form oxyanions (see paragraph 2.4), field data for Cr, As, V and Sb are shown in Figure 8. The results are from the fields with FGA (F1) and the RCA (F3W and F7). In the fields with RCA the effect of carbonation and possible leached layer formation can be seen as pointed out above. Significant decrease in concentrations of Cr, As and V was found to start after 30 weeks of exposure ($L/S = 0.05$). For V and As, however, the concentrations increased for the first weeks. From the raw data it was found that the increase took place up to 5-10 weeks. A similar effect was not observed for the metals that typically do not form oxyanions.

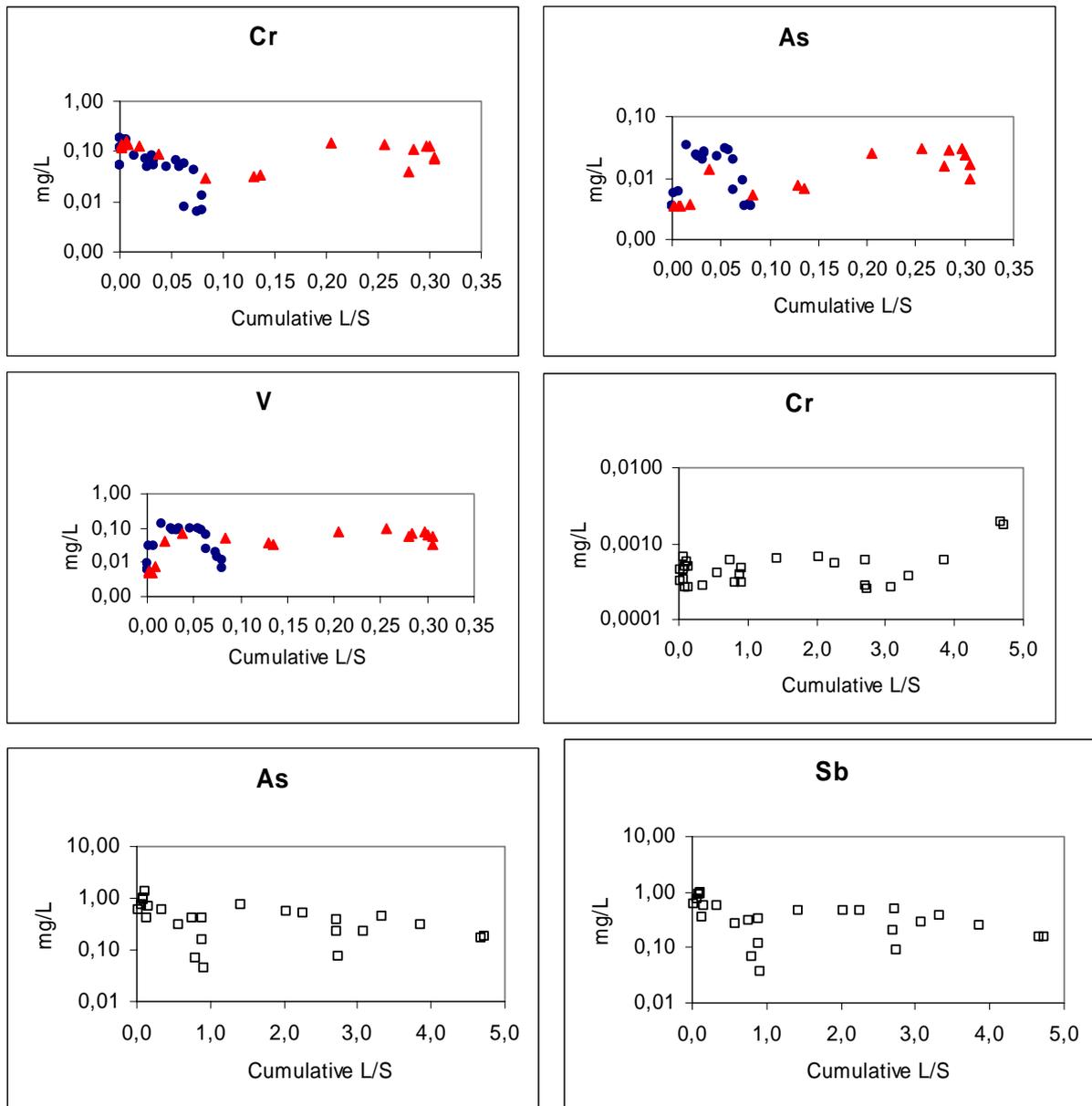


Figure 8 Release of Cr, As, V and Sb as function of cumulative L/S determined in water samples from F3W (triangles), F7 (bullets) and F1 (open squares) during the first 14-month.

The release of Cr, As and Sb from F1 in Figure 8 was relatively consistent as the concentrations seem to reach a constant level. For As and Sb the behaviour is almost identical in regard to the released quantity and the time where the concentration drops occur. To decide if the latter is due to a chemical effect (e.g. solubility control) modelling of the pore water chemistry will be applied.

The Norwegian limits for pore water concentrations based on K_d values in sensitive land use for As and Cr are $69 \mu\text{g/L}$ and $833 \mu\text{g/L}$ respectively. Most of the As concentrations determined in F1 exceeded this limit. However, applying an extended risk evaluation taking into account the actual chosen field scenario resulted in an acceptable level risk.

4. CONCLUSIONS

The FGA and RCA materials showed different behaviour in terms of leaching patterns and quantity of elements released. As the acid neutralization capacity was less for the FGA material, the field pH of the leachates was more influenced by the external field conditions. The test fields were constructed in a way that reflected different infiltration volumes and the results agreed with this variation of infiltration volumes. Significant carbonation and leaching of alkalinity was observed. The pH of the concrete fields covered with asphalt (F3W and F3E) decreased from 12.7 to 11 compared to a decrease in pH from 12.8 to 9.5 for the concrete field without asphalt cover (F7). This effect could also be seen in the constituent leaching from these fields.

The leaching patterns for some of the elements that have the tendency to form oxyanions (V and As) was different compared to the leaching of Cu and Ni. This indicates solubility controlled leaching. However, this still needs to be verified by coupling the monitoring data i.e. the infiltration volumes, rain fall, temperatures, and concentrations further with the laboratory data and the geochemical modelling approach.

Relatively high total contents of Cu, Cr, Pb and As in the FGA material did not exceed the criteria for pore water concentrations given by Norwegian pollution authorities.

The monitoring of the field experiment will continue and more generic data sets will be generated and characterisation of physical and chemical properties of the characterisation exposed materials will also be carried out.

5. REFERENCES

- [1] Norwegian Public Roads Administration: "The Norwegian Roads Recycled Materials R&D Program", www.gjenbruksprosjektet.net, Project web site
- [2] C.J. Engelsen, J. Mehus, C. Pade, D.H. Sæther, Carbon dioxide uptake in demolished and crushed concrete, NBI project report 395 (2005).
- [3] G. Petkovic, C.J. Engelsen, A.O. Haoya, G. Breedveld, Environmental impact from the use of recycled materials in road construction: method for decision-making in Norway, *Resources Conservation and Recycling* 42 (3) (2004) 249-264.
- [4] J.J. Dijkstra, J.C.L. Meeussen, R.N.J. Comans, Leaching of heavy metals from contaminated soils: An experimental and modeling study, *Environmental Science & Technology* 38 (16) (2004) 4390-4395.
- [5] B. Ludwig, P. Khanna, J. Prenzel, F. Beese, Heavy metal release from different ashes during serial batch tests using water and acid, *Waste Management* 25 (10) (2005) 1055-1066.
- [6] H.A. van der Sloot, Characterization of the leaching behaviour of concrete mortars and of cement-stabilized wastes with different waste loading for long term environmental assessment, *Waste Management* 22 (2) (2002) 181-186.

- [7] H.A. van der Sloot, Comparison of the characteristic leaching behavior of cements using standard (EN 196-1) cement mortar and an assessment of their long-term environmental behavior in construction products during service life and recycling, *Cement and Concrete Research* 30 (7) (2000) 1079-1096.
- [8] C.J. Engelsen, H.A. van der Sloot, G. Wibetoe, E. Stoltenberg-Hansson, G. Petkovic, W. Lund, Constituent Release Characterization of different recycled concrete aggregates, *Cement and Concrete Research*, in preparation (2006).
- [9] B. Lagerblad "Carbon Dioxide Uptake during Concrete life cycle," CBI report 2:2005.
- [10] Norwegian State pollution Authorities (SFT: 99:01), Risk assessments of contaminated ground grunn (1999), in Norwegian.
- [11] D. Shriver, P. Atkins *Inorganic Chemistry* 3 ed.; Oxford University Press.
- [12] C.A. Johnson, Metal binding in the cement matrix: an overview, Internal report (2002).