

# **Comparison Between Laboratory Leaching Properties and Field Experiments of MSWI Bottom Ash Used as an Unbound Granular Material for Road Pavements**

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## **ABSTRACT**

The potential use of bottom ash from municipal solid waste incineration (MSWI) as an aggregate substitute in unbound granular layers of a road pavement is investigated. The mechanical behavior of bottom ash has already been proven to be acceptable for this application based on the Spanish Road Regulations. Actually, the potential environmental impact is the key point for the use of bottom ash as a road material. The environmental properties of bottom ash in this application were assessed by means of the Dutch availability test NEN 7341 and the single-batch and two-stage batch European CEN 12457 laboratory leaching tests. Furthermore, an experimental unbound pavement transect was constructed to provide information on the leaching behavior under field conditions. In this high infiltration scenario, the results from predicted (based upon laboratory leaching tests) and measured releases (under field conditions) were compared. The depletion period of the extractable fraction of some elements was also quantified.

## **INTRODUCTION**

Bottom ash is the most significant by-product arising from municipal solid waste incineration, since it accounts for about 90% (in weight) of the solid product resulting from this combustion. Incineration reduces the volume of waste by about 90% and its mass by about 70%<sup>1</sup>. Currently, the annual production of bottom ash in Catalonia (NE Spain) reaches 150000 tones, produced in seven municipal solid waste incineration (MSWI) plants. Nowadays, bottom ash in Catalonia is mainly landfilled and only a minor proportion is applied for engineering purposes. The Catalan Administration requires landfilling taxes for this type of material with the aim of promoting the valorization.

Owing to its characteristics, it is thought that bottom ash could be used as a road material replacing natural aggregates<sup>2,3</sup>.

Experimental data showed that MSWI bottom ash meet the physical and mechanical requirements established by Spanish Road Regulations for granular layers, as well as the Catalan Specifications for MSWI bottom ash valorization, environmental requirements referred to leachability of Cu, Pb, Zn, As, Cd and Cr based on DIN 38414 leaching test<sup>4</sup>. Nevertheless, these environmental standards are thought to be permissive because only the above elements are regulated, with relatively tolerant limit values compared with other environmental standards from different EU state members.

Leaching tests provide information on environmental properties of bottom ash to support decisions making on waste management. However, for this specific application the leaching behavior of the environmentally relevant elements under field conditions may differ from the predicted behavior based on results obtained in laboratory leaching tests<sup>3,5</sup>. For this reason, it is of scientific and environmental interest to compare the leaching properties of bottom ash deduced from laboratory leaching tests with the actual leaching occurring in a field scenario of valorization.

The aim of this study is to compare the results from predicted releases, deduced from laboratory tests, with field data when applied as a granular material in a flexible pavement section (made up of unbound layers exclusively). In addition to the comparison of predicted and measured leachable concentrations, the soluble fraction depletion for some elements under field conditions can be evaluated.

## METHODOLOGY

### *Materials*

MSWI bottom ash used in this study was generated in Mataró plant (NE Spain, Figure 1) and the quality was improved by means of a post-incineration treatment performed at Pedreres Rusc facilities (close to the incineration plant), where large particles and a significant proportion of the metallic fraction and unburned material were removed. The treatment included also an ageing during several months. The material was sampled from the stockpiles at treatment plant and characterized prior to the application.

The field test, located in a rural area in Tagamanent (NE Spain, Figure 1), consists of an experimental road transect of 200 m in which MSWI bottom ash was applied as a granular material, as well as in other products such as gravel-ciment or compacted concrete, replacing natural aggregates.

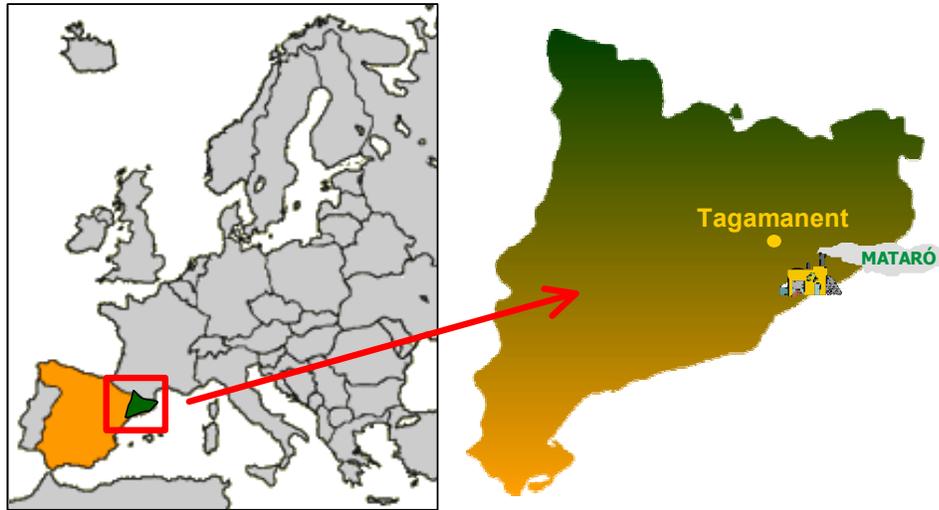


Figure 1. Location of the Mataró MSWI plant and the Tagamanent experimental road.

This study is focused on the use on bottom ash as a granular material in unbound pavement sections. In this case, the pavement section was completed with an unbound granular layer of compacted limestone aggregates on top of the bottom ash layer. In a small transect of this field test, drainage channels of 5m long x 3.7m wide were build (Figure 2), impermeabilized and filled with compacted bottom ash. A tube with drilled holes was placed across the channels to collect the percolation water and drive it towards a sampling tank. Field leachates were sampled with a 4-month periodicity (except for two consecutive extractions), and 7 field extractions were obtained.

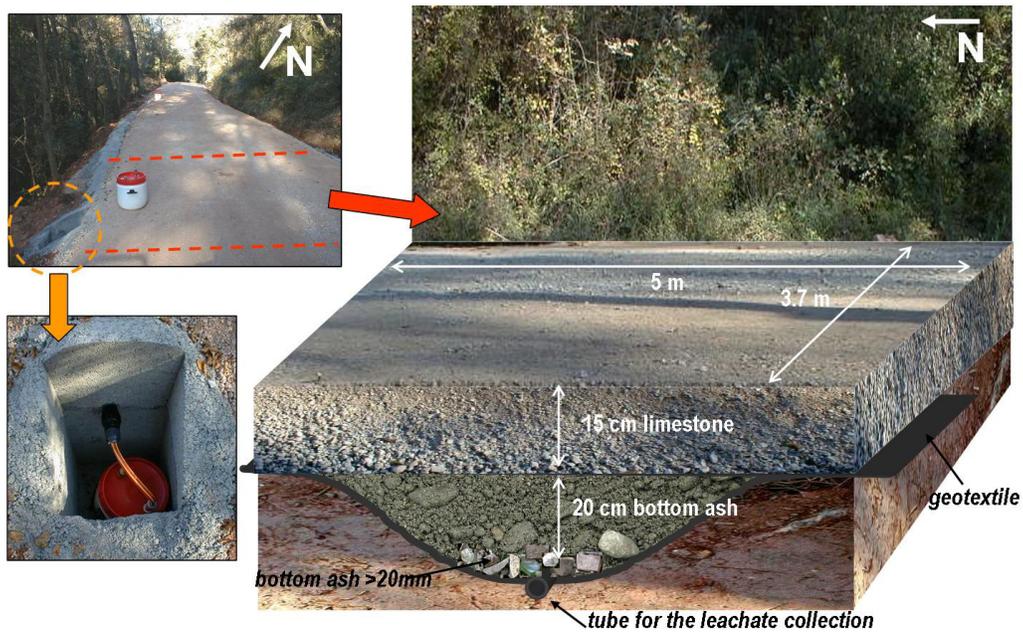


Figure 2. Characteristics of a bottom ash channel, and location of leachate collection and sampling devices.

As a reference material, a limestone channel was also constructed. Results obtained allowed to distinguish the proportion of the field releases corresponding to the leaching of the upper limestone layer, since this proportion should be discounted.

On the other hand, 15 months after the construction core samples from one channel were extracted to determine the total leached amounts of some constituents for this period. Around 15 kg of bottom ash were collected and subsequently mixed and quartered to supply subsamples for the determination of the leaching potential.

### *Characterization methods*

Milling of quartered fractions was carried out for the chemical characterization. Major, minor and trace element concentrations were determined in bottom ash. Samples were acid-digested by using a special two-step digestion method devised for the analysis of trace elements in coal and combustion wastes<sup>6</sup>. Silica contents were determined by X-Ray Fluorescence (XRF) on bulk solid sample, but the other elements were analyzed by means of Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) in the acid digestions. A standard reference material (City Waste Incinerator Fly Ash) was used to test accuracy of the analytical determination. A relative error <3% was obtained for most elements with the exception of K and P (<10%).

### *Laboratory leaching tests*

Since results from laboratory leaching tests were correlated with field data obtained from the experimental road, it has to be taken into account that the road is made up of granular unbound layers, and therefore high L/S ratios are expected. In accordance with this high infiltration scenario, medium-high L/S ratio leaching tests were selected for the environmental characterization of bottom ash. Thus, the environmental performance of bottom ash was assessed by means of NEN 7341<sup>7</sup>, CEN 12457-2<sup>8</sup> and CEN 12457-3<sup>9</sup> leaching tests.

NEN 7341 is a two-step batch and pH-controlled leaching test carried out with deionised water at a cumulative L/S ratio of 100 L/kg. The sample particle size is reduced down to 125 µm and the pH is maintained (using HNO<sub>3</sub>) for three hours at 7.0 and 4.0 in the first and second stages, respectively. CEN tests are applied on granular wastes < 4 mm with deionised water as leachant. CEN 12457-2 is a single batch leaching test performed at L/S=10 L/kg (agitation time of 24 hours). CEN 12457-3 is a two-stage batch leaching test also performed at a cumulative L/S ratio of 10 L/kg (first leaching step, 6 hours at L/S = 2 L/kg; second step, 18 hours at L/S = 8 L/kg).

Leaching tests were carried out by duplicate except for the core samples, performed by triplicate. The contents of major and trace elements in the laboratory and field leachates were determined by means of Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) and High Performance Liquid Chromatography (HPLC) for anionic species.

### *Data treatment*

With the aim of referring the field data obtained in mg/L to cumulative emissions (mg/kg) as a function of the L/S ratio, the percolation water volume and the leached mass for the different leaching episodes are needed.

The mass of bottom ash and limestone for each channel was estimated by means of the geometrical characteristics of the pavement sections. Concerning the percolated water volume, the cumulative rainfalls in the leaching periods were obtained with the data registered in a meteorological observatory (Tagamanent Meteocat station) located close to the field test. Mean annual concentrations of major rainwater components were also obtained from prior studies<sup>10</sup>. In absence of bound layers, the pavement section becomes a high-infiltration scenario, and hence percolation is the main leaching mechanism. However, the experimental road has a slight slope (3-4%) and the surface is a well-graded and highly compacted limestone aggregate layer. Therefore, it is thought that only a certain proportion of the rainfall percolates into the basin and washes the ash.

The infiltration rate was calculated empirically, on the basis of a mass balance between concentrations measured in the field leachates, the volume of precipitation and the decrease of concentrations between the original bottom ash and core-samples. If assumed that the proportion of rainwater that infiltrates into the ash is constant, and taking into account highly soluble species markedly enriched in bottom ash with respect to limestone (since for this calculation the leached amounts must be attributed exclusively to bottom ash) infiltration ratios of 30%, 21%, 34% and 39% were obtained for Na, K, SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>, respectively. This means that around 30% of the total rainfall penetrates into the channel and interacts with the bottom ash.

Once the water volume of each leaching episode was estimated following this procedure, and since the mass of bottom ash is known, the L/S ratio corresponding to the different extractions was estimated.

Leachates from the channel filled with compacted limestone aggregates were also sampled to measure the leached concentrations. With these results, the contribution of the upper limestone layer contained in the bottom ash channels could be subtracted.

## RESULTS AND DISCUSSION

### *pH and conductivity*

The pH measured for field leachates range from 7.3 to 9.2 (Figure 3). These values tend to fluctuate along the study period and to be slightly lower than those determined under CEN tests conditions (8.5-8.9). These differences were also observed by Chandler et al.<sup>2</sup> and could be related to a certain uptake of atmospheric CO<sub>2</sub> during the storage of the leachates before sampling.

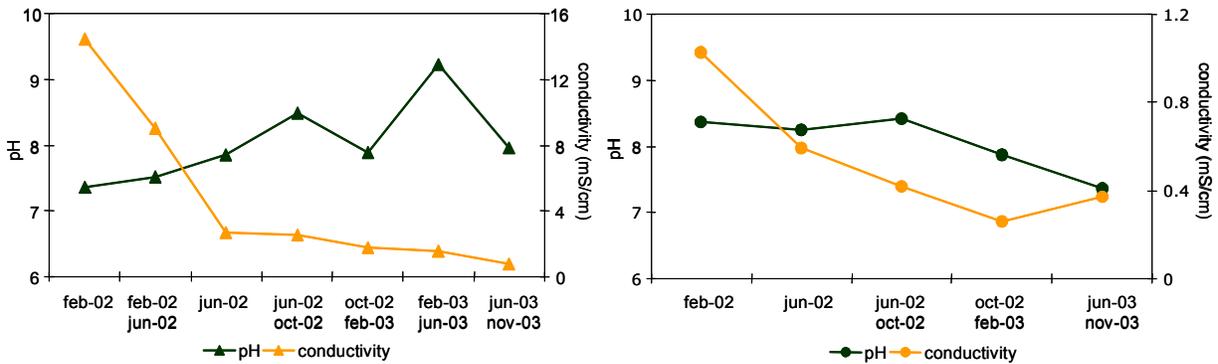


Figure 3. Evolution of pH and conductivity values of bottom ash and limestone channels from experimental road in Tagamanent.

The two initial extractions yielded high conductivity values ( $>8$  mS/cm), while in the subsequent leachates values measured were lower than 2.7 mS/cm and progressively continued decreasing, despite the fact that each extraction took place under different L/S ratio. This trend may be related to the initial quick and high release of constituents occurring as highly soluble salts such as NaCl or KCl. Chlorides are readily depleted but the progressive dissolution of calcium sulfates (less soluble) continued. As shown in Figure 3, after several months, conductivities as well as pH values of bottom ash leachates were similar to those observed in the limestone channel.

#### *Leaching of bottom ash constituents under field conditions*

Figure 4 compares the bulk content of some constituents, the available fraction (NEN 7341), the leachable fraction (according to CEN 12457-2 and CEN 12457-3 tests) and the leached fraction in Tagamanent field test after 2 years of performance. The latter is represented as cumulative emission curves. When the curve of an element acquires an asymptotic morphology, it can be assumed that the soluble fraction is progressively depleted. Since the L/S ratio scale can be converted into a time scale (it could be established that for this field test in the Tagamanent area, 1 year corresponds to an increase of about 1.4 L/kg), the depletion period of the soluble fraction of each constituent can be quantified.

It has to be highlighted that this depletion period is not an intrinsic parameter but a value conditioned by several factors such as the speciation, the magnitude of the leachable fraction or the infiltration rate (which is, in turn, governed by the precipitation regime of the area and the physical characteristics of the structural section of pavement). For these reasons the results cannot be extensive to other situations.

According to the release pattern of the soluble fraction, and if L/S ratio scale is interpreted as time scale, bottom ash constituents can be classified into the following two 2 categories.

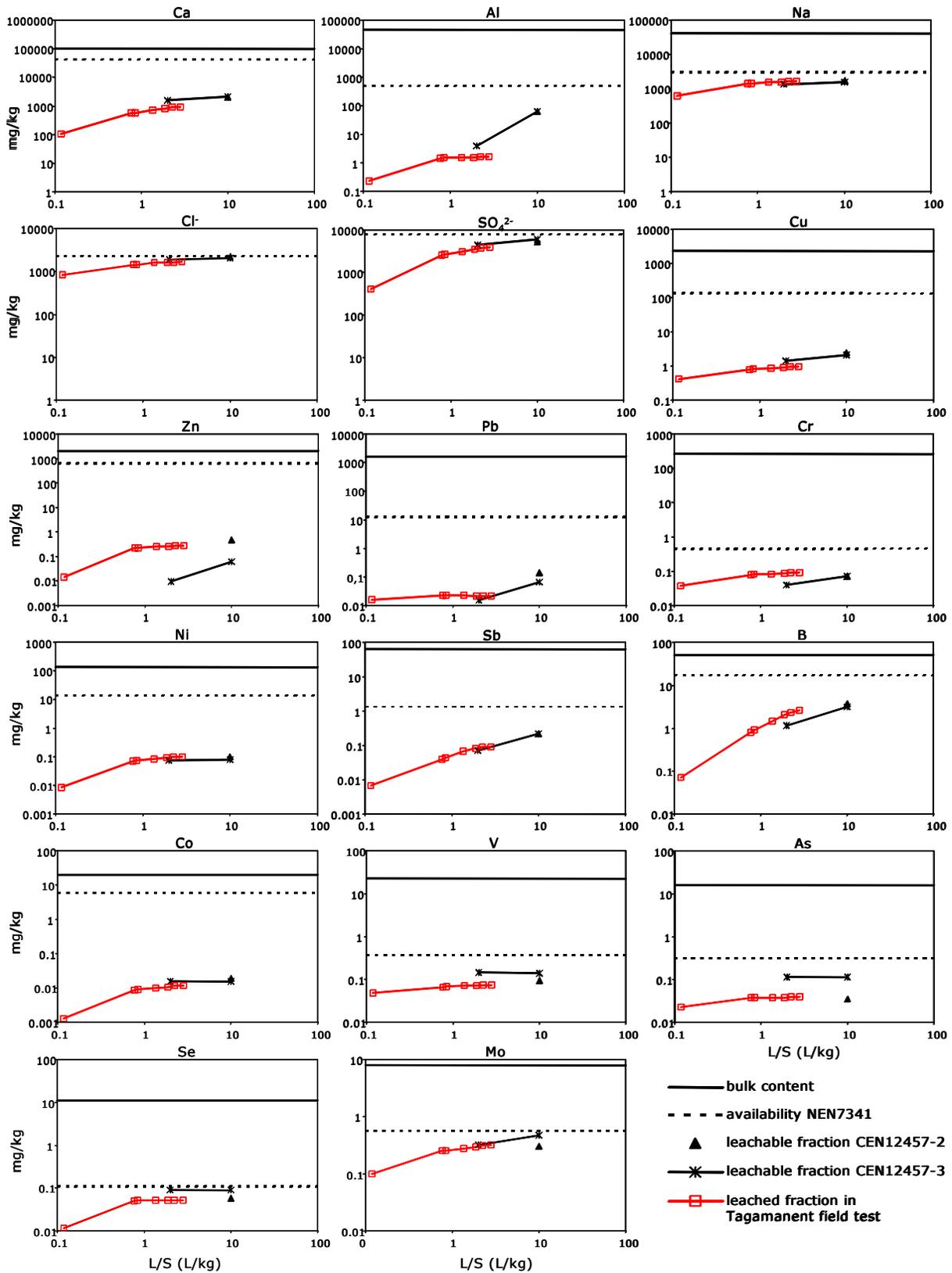


Figure 4. Total content, availability, laboratory leachable fraction and field leached fraction of several elements in bottom ash from Mataró plant.

- Readily leached constituents: The soluble fraction of elements occurring in highly soluble salts (Na, K, Cl<sup>-</sup>, F<sup>-</sup> and NO<sub>3</sub><sup>-</sup>) as well as Cu, Fe, V, Cr, As, Pb, and Cd is nearly depleted in the two initial leaching stages, the first stage accounts for over the 50% of the total cumulative emission. However, the high ionic load in the first leachate may restrain the mobilization of some elements in which the major emission is slightly delayed, taking place in the second extraction. This trend was observed in Al, Ti, Co, Ni, Zn and Se. In both cases, the leached fractions were released in a period that does not exceed 1 year, and subsequent extractions contain very low proportions of these constituents.
- Slowly leached constituents: Some elements are progressively leached with increasing L/S ratio in such a way that each leaching period dissolves similar proportions and thus the emission curves progressively rise up. However, in most cases, the last leachates obtained draw a gentle slope of the curve, suggesting the initial depletion of the extractable fraction. The depletion period can reach 2 years. This trend, characteristic of slightly soluble salts, is the dissolution pattern of Ca, SO<sub>4</sub><sup>2-</sup>, Mg, Rb, Sr and Ba. In addition to these species, elements such as B, Mo, W and Sb exhibit the same behavior. The soluble fraction of these elements is supposed to be in oxianionic form, and therefore subjected to a relatively low and progressive dissolution.

As shown in Figure 4, after 2 years the releases of all these elements (with the exception of B) were clearly low and the depletion period was nearly or completely concluded. In the case of B, no signs of depletion were observed and the progressive dissolution may continue.

Therefore, it can be concluded that in 2 years and for the Tagamanent experimental road, the soluble fraction of most constituents in bottom ash is depleted. If assumed that no drastic changes in mineralogy of bottom ash occur in medium term (which, in turn, can be possible since the used bottom ash was already weathered prior to the application), the cumulative emission of most elements determined after 2 years at L/S ratio of 2.80 L/kg is found to be representative of the cumulative emission that may occur for a period of several years.

#### *Comparison of lab and field tests*

Prior to the comparison of the results obtained from laboratory leaching tests and the field data, it is necessary to emphasize that the pavement test section in Tagamanent is subjected to high infiltration due to the absence of bound layers. In this situation, high L/S ratios are relatively easy to attain and consequently the laboratory leaching tests performed to predict field results would focus on short to medium time frame previsions.

Taking into account a measured increase of 1.4 L/kg in 1 year in Tagamanent road test, the L/S ratio of 10 L/kg (used in CEN tests) is expected in about 7 years. However, cumulative emissions of most elements are stabilized in 2 years. It means that in

Tagamanent test and within 7 years, cumulative emissions may be similar to those currently determined. For this reason, measured field releases can be compared with laboratory predicted releases.

In general, a good agreement between laboratory and field emissions was observed, as seen in Figure 4. For many constituents, such as Al, Fe, Pb, Sn, Sr, W or Sb, field releases are clearly lower than CEN leaching predictions. For some elements, field releases are slightly lower (for example Ca,  $\text{SO}_4^{2-}$ , Ti or Cu) than the predicted values. For other elements occurring in salts, such as Na, K, Mg,  $\text{Cl}^-$ , Cr, V, Se, Mo and B, as well a few metals, such as Ni and Co, field leached concentrations are very close to leachable fractions based on both CEN tests.

Since the soluble fractions of most of those constituents are nearly depleted, it can be concluded that in most cases CEN tests are a valuable tool to make realistic and closely provisions about the leaching potential of bottom ash in a short to medium time frame (several years). However, as previously exposed, B is still progressively leached. Consequently, for this element field releases could exceed predicted releases deduced by laboratory leaching tests.

Only in the case of As and Zn a poor agreement was found between predicted values based on laboratory tests and measured field values, but this is attributed to some discrepancies among the two CEN and field tests. The proportion of Zn leached under field conditions is larger than the leached concentration based on CEN 12457-3 (2-stage batch test), but similar to CEN 12457-2 (single batch). For As, again similar results were obtained for the single extraction test and the field tests, but with leachable concentrations lower than the 2-stage batch test.

It should be noted that for most elements there is also good agreement between emissions from the single and 2-stage batch leaching tests at a cumulative L/S=10 L/kg. However, in a few cases some significant differences were observed. Leachable concentrations of metals such as Zn, Pb, Sn or Fe according to CEN 12457-2 are higher than CEN 12457-3, probably due to solubility restraints. Conversely, V, As, Se and Mo, occurring as oxianionic species, show higher extractable proportions under the 2-stage batch leaching tests conditions. The 2-stage batch leaching test is probably reproducing more closely the field conditions since these include successive leaching stages at low L/S ratios, rather than a single stage at a high L/S ratio. These observations should be taken into account to select the proper leaching test with views to bottom ash management aspects.

Finally, it has to be said that leachable fractions according to CEN tests, as well as leached fractions in Tagamanent road test, are in many cases clearly lower than available fractions determined by availability NEN 7341 test, except for species occurring in soluble salts such as Na, Cl,  $\text{SO}_4^{2-}$ , and to a certain extent other elements in their oxianionic form such as V, Mo, As or Se. This suggests that availability test could be useful to provide information about the leaching potential of those constituents,

but it has a limited application to predict the actual fate of most metals leached from MSWI bottom ash in field applications.

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