

# Monitoring of leachate at a test road using treated fly ash from municipal solid waste incinerator

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## Abstract

A treatment process for municipal solid waste incinerator fly ash has been jointly developed by the Université Libre in Brussels and the Solvay company. This experimental process has led to an experimental project with a multi-disciplinary team from Solvay along with several companies and organisations, plus support from the ADEME agency. After a laboratory assessment of the treated fly ash (TFA), its environmental properties and the optimal fields of application in road construction, as part of the *Revasol* project, a full-scale analysis was organised for both environmental and mechanical purposes. A test section using TFA was designed in order to collect percolation water; TFA was used as a road base component (a cement-bound graded aggregate containing 12% TFA). A reference section with the same structure was also built using natural materials.

After a description of the treatment process and the test road design, the main TFA characteristics are presented. Then, the environmental behaviour of both sections over a 1-year period following construction is detailed: infiltration, leachate quality, cumulative release. This monitoring effort highlights not only the evolution in TFA behaviour, but also the general permeability of the road structure to various external influences.

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## 1. Introduction

The most dramatic change in municipal solid waste (MSW) incineration technology over the last decades pertains to improvements in air pollution control (APC) systems. This trend has resulted in increasing both the variety and quantity of APC residues, among which fly ash constitutes the particulate matter conveyed from the combustion chamber of the MSW incinerator and removed from the flue gas stream prior to the addition of any type of sorbent material [1].

Solvay developed a dry process for treating incinerator fumes (Neutrec<sup>®</sup>-Gaz); the first step in this process consists of filtering the fly ash (whose production amounts to around 25 kg/t of incinerated MSW). Another process developed by Solvay (the *Novosol* process) has been designed to treat this fly ash [2]. The *Revasol* project was aimed at validating the *Novosol* process at a pilot scale and creating reuse opportunities for the treated fly ash (TFA), in particular as an environmentally-friendly road construction material. The project was financially supported by the French environment and energy management agency (ADEME).

Today, it remains difficult to forecast material behaviour under actual road use conditions on the sole basis of laboratory tests. Current knowledge on both the evolution in surface course permeability under the joint effect of traffic and climatic agents and water circulation patterns within the road structure is still quite poor [3,4]. In the field of road engineering, building a test section represents an essential step in the normal validation procedure for a new technique. Within the framework of the *Revasol* project, the construction of a temporary test road was thus decided in order to assess one of the possible technical solutions involving use of TFA. A reference section, made of classical materials, was also constructed as a means of placing the TFA effects into perspective. For the environmental purpose, the full-scale analysis consisted in a 1-year monitoring of the hydrology of the test road and of the quality of leachates, in the evaluation of release for the obvious most significant chemical parameters and in the appraisal of the physicochemical evolution of leachates.

## 2. Materials

The amount of information available on APC residue properties in general, and on fly ash properties in particular, is not as extensive as that found on MSW incinerator bottom ash, due

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Table 1  
Summary analysis table of the 10 MSW incinerator fly ash samples

Analysis	Grading ( $\mu\text{m}$ )		Density ( $\text{kg/m}^3$ ) <sup>a</sup>	Main constituents <sup>b</sup>					Weight loss <sup>c</sup> (g/kg)
	$D_{50}^c$	$D_{50}^d$		$\text{Al}_2\text{O}_3$ (g/kg)	CaO (g/kg)	$\text{Fe}_2\text{O}_3$ (g/kg)	MgO (g/kg)	$\text{SiO}_2$ (g/kg)	
Mean value	174	45	544	98	235	27	27	230	65
Standard deviation	51	15	154	29	42	11	4	5	23

Analysis	Soluble salts <sup>f</sup>				Trace elements <sup>g</sup>				
	NaCl (g/kg)	KCl (g/kg)	$\text{CaSO}_4$ (g/kg)	CaO (g/kg)	Zn (g/kg)	Pb (g/kg)	Cr (g/kg)	Cd (g/kg)	Hg (g/kg)
Mean value	58	79	135	23	16	6.1	0.57	0.28	0.002
Standard deviation	32	38	46	25	7.8	4.4	0.22	0.16	0.002

<sup>a</sup> Free flowing density.

<sup>b</sup> X-ray fluorescence.

<sup>c</sup> Sieve size distribution.

<sup>d</sup> Laser size distribution dispersed in saturated alcohol.

<sup>e</sup> At 1000 °C.

<sup>f</sup> Chemical analysis.

<sup>g</sup> Inductively-coupled plasma.

in part to the “newer” status of the former and in part to the apparently more limited use potential [1].

As for the *Revasol* project, during an initial stage, random grab samples of 10 European MSW incinerator fly ash (from Belgium, France, Germany, Italy, Switzerland and the Netherlands) were collected and analysed regarding their physical characteristics, major chemicals and trace elements (see Table 1). The 10 fly ash samples proved to be in conformance with former analyses carried out at larger scale [5]. The main mineral phases detected and assessed were amorphous silicates and alumino silicates, calcium sulphate anhydride, halite and sylvite, calcite, microcline and gehlinite, quartz and rutile. During a second stage, five of the initial 10 samples, which exhibited either typical or extreme compositions, were selected and submitted to an in-depth analysis according to the protocol described by Piantone et al. [6].

### 3. Methods

#### 3.1. Treatment process

The main steps within the *Novosol* process applied to fly ash (see Fig. 1) are, in succession: a washing intended to remove

soluble salts, a phosphation intended to trap heavy metals in stable crystalline phases as apatite (calcium phosphate), and lastly a calcination intended to oxidise organic compounds, in particular dioxins and furans [2].

This process was applied at the laboratory scale on the five selected fly ash samples. During a third stage, two of these samples were further selected to be produced at the pilot scale (1 t/day capacity) in order to compare results between laboratory batches and continuous pilot runs. Then, in a fourth stage, in order to represent the worst scenario on pollutant leaching considering the treatment process, the fly ash sample most heavily polluted in chromium and rich in calcium sulphate, was selected. From this fly ash, 20 t of TFA were produced for building the test road.

#### 3.2. Design and construction of the test road

Within the project working group, a wide array of scenarios for TFA use were studied by the Eurovia company and the Laboratoire Central des Ponts et Chaussées (LCPC) [7]. Since TFA displays cementitious properties that enable substitution for a portion of the cement amount, a cement-bound graded aggregate

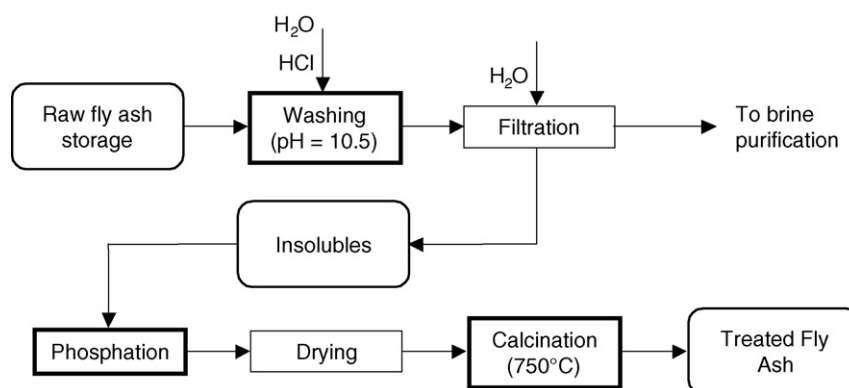


Fig. 1. Principle of the *Novosol* process applied to MSW incinerator fly ash.

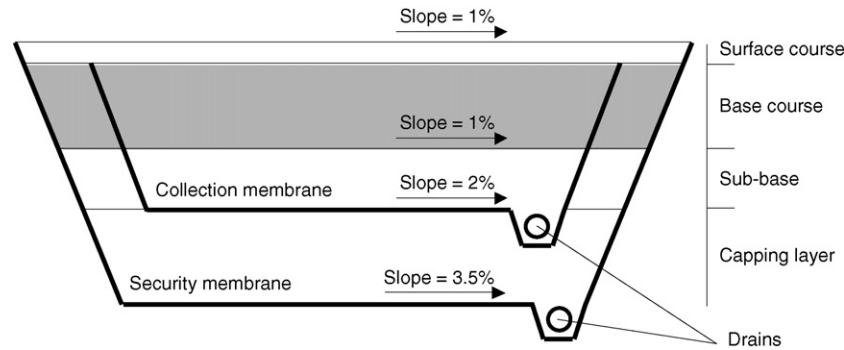


Fig. 2. Structure of the test section water collection system.

gate with 12% TFA as a road base was selected for the full-scale testing.

Two road sections (25 m long and 5.5 m wide) were thus built: the test section including TFA, and the reference section containing natural materials. The geo-membrane laid underneath each section was limited to a length of 21.5 m so as to avoid the cross-contamination of leachates; it was designated as the “collection membrane”. The test section was also equipped with a second, more deeply-positioned geo-membrane (called the “security membrane”) intended to collect all percolating waters and control their quality before discharge. The double membrane collection system was devised not only in order to collect all the leachate but also in order to avoid the drawback of a possible preferential flow from the road surface. Indeed, for such a test road, from a new wearing course (i.e. without any surface crack), the risk of preferential flow from the surface lies mainly in a preferential infiltration where the vertical wall of the membrane is in contact with the surface. The structure of the test section is presented in Fig. 2. It prevents preferential flow along the wall of the security membrane to interfere with the hydrology of the test section. Once infiltrated into the surface course the water is then expected to flow evenly down to the studied base course and the collection membrane.

In order to facilitate the infiltration of rainfall into the road structure, the transverse slope of the surface course was reduced (1%) in comparison with typical practice (which calls for 2.5%) in road construction. The transverse slopes of both the sub-base and capping layer were more in conformance with standard practices (2% and 3.5%, respectively). The structure of these two sections was designed according to the details provided in Table 2.

For both sections, the surface area covered by the collection membrane amounted to 118 m<sup>2</sup>. The volume of material involved in the monitoring campaign was 28.3 m<sup>3</sup> for the test section and 23.4 m<sup>3</sup> for the reference section, with this variation due to a slight difference in base course thickness. The base course mass for this experiment was 57.9 t (dry density = 2.05 t/m<sup>3</sup>) and 50.3 t (dry density = 2.15 t/m<sup>3</sup>) in the test and reference sections, respectively. The test road was built by Eurovia on the site of the Solvay Company, with the assistance of the Nancy (eastern France) Regional Public Works Laboratory (Laboratoire Régional des Ponts et Chaussées—LRPC). It was opened to traffic (60–70 trucks/day) on 27th June 2002.

### 3.3. Monitoring activities

The monitoring of the test road started from its opening to traffic and lasted until 9th September 2003. Volumes of collected water were measured by the LRPC-Nancy Laboratory, which was also responsible for sampling and some of on-site measurements on leachate (temperature, electric conductivity, pH, redox potential). Leachates were collected in a 1.2 m<sup>3</sup> high-density polyethylene-tank. On the whole monitoring period, 14 samplings were carried out for each section. From sampling no. 1 (2nd July 2002) to sampling no. 8 (10th October 2002), samples were collected when the tank was full. Then, in a second phase, from sampling no. 9 (4th December 2002) to the end of the monitoring, leachates were collected regularly in order to constitute mean samples on a period of approximately 2 months. Rainfall was measured on-site by Solvay. The complete analysis of leachates was commissioned to a laboratory independent of the project. For both sections leachate analysis, an initial list of 34 parameters was defined, based on the TFA chemical and

Table 2  
Structure of both the test and reference sections

Structural component	Test section	Reference section
Surface course	5 cm <sup>a</sup> bituminous concrete	5 cm <sup>a</sup> bituminous concrete
Base course	24 cm <sup>a</sup> cement-bound graded aggregate (0/20 mm) with 12% TFA and 3.5% cement	20 cm <sup>a</sup> cement-bound graded aggregate (0/20 cm) with 4.5% cement
Sub-base (draining layer)	15 cm unbound graded aggregate (0/14 mm)—silty material	15 cm unbound graded aggregate (0/14 mm)—silty material
“Collection membrane”	Geo-membrane	Geo-membrane
Capping layer (draining layer)	20 cm unbound graded aggregate (0/14 mm)—silty material	—
“Security membrane”	Geo-membrane	—

<sup>a</sup> Actual mean thickness measured on core samples.

Table 3  
Parameters of the leachate monitoring of both sections

Parameters	Test section	Reference section
pH	Full	Full
Electric conductivity	Full	Full
Redox potential	Full	Full
Chemical oxygen demand	Stopped (not valid)	Stopped (not valid)
Suspended solids	Full	Full
Kjeldahl nitrogen	Full	Full
Ammonia nitrogen	Full	Full
Nitrites	Full	Full
Nitrates	Full	Full
Adsorbable organic bound halogens (AOX)	Stopped (not valid)	Stopped (not valid)
Total organic carbon (TOC)	Full	Full
Total CN	Stopped (<DL1)	Stopped (<DL1)
Phenol index	Full	Full
Total hydrocarbon	Full	Full
Fluorides	Full	Full
Sulfates	Full	Full
Chlorides	Full	Full
Aluminium	Full	Full
Arsenic	Full	Full
Cadmium	Stopped (<DL2)	Stopped (<DL2)
Chromium	Full	Stopped (<DL2)
Chromium VI	Full	Stopped (<DL2)
Copper	Stopped (<DL2)	Stopped (<DL2)
Iron	Full	Full
Mercury	Full	Full
Nickel	Stopped (<DL2)	Stopped (<DL2)
Manganese	Full	Full
Molybdenum	Full	Full
Lead	Stopped (<DL2)	Stopped (<DL2)
Selenium	Full	Full
Tin	Full	Full
Zinc	Stopped (<DL2)	Stopped (<DL2)
Antimony	Full	Full
Barium	Full	Full

DL1: detection limit = 0.01 mg/l; DL2: detection limit = 0.05 mg/l.

leaching characteristics observed thanks to preliminary laboratory results [8], but also based on wishes of the administration in charge of environment protection. The analytical programme of monitoring is described in Table 3. Not all the parameters were monitored through the end of the study. Indeed, depending on the section, the monitoring of some parameters was stopped at sampling no. 9, either because it was not valid (COD, AOX) or due to lingering concentrations below detection limits (see Table 3). Such a decision was taken after checking its consistency with the preliminary laboratory leaching results [8].

## 4. Results

The main characteristics of the TFA observed in the laboratory (chemical composition and leaching), together with the leaching observed as part of test road monitoring, are presented herein. Not only are concentrations in the leachate of interest to interpret, but the release (mass of element leached per dry weight of material [1], here the studied base course) also lend considerable insight. Collected water volumes serve to calculate the actual liquid-to-solid ratios ( $L/S$ , expressed in l/kg).

### 4.1. Treated fly ash characteristics

The characteristics and composition of TFA are given in Table 4. Those figures are related to mean values on production samples, which present rather small dispersion. Grading and composition are similar to the raw material, except for the content of  $P$  which had risen due to the phosphation process. Major mineral phases analysed by X-ray diffraction are gehlinite ( $\text{Ca}_2\text{Al}_2\text{SiO}_7$ ), microcline ( $\text{KAlSi}_3\text{O}_8$ ), anhydrite ( $\text{CaSO}_4$ ), calcite ( $\text{CaCO}_3$ ), apatite ( $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH},\text{F},\text{Cl})_2$ ), and vitreous silicates. Phosphorous is present in the form of apatite and traps most of the metal ions in cationic form [6].

### 4.2. Hydrology of the test road

The total volume of water collected from the test section equals 25,616 l, while this figure climbs to 29,851 l for the reference section. Relative to the drained surface area of the two sections, these volumes lead to an infiltrated water depth of 216.6 and 252.4 mm, respectively. In comparison with rainfall measurements recorded during the monitoring period (701.6 mm), this means that over the entire period, the infiltration rate was

Table 4  
Primary characteristics of treated fly ash (TFA)

Analysis	Grading <sup>a</sup>			Main constituents <sup>b</sup>						Weight loss <sup>c</sup> (g/kg)
	$D_{90}$ ( $\mu\text{m}$ )	$D_{50}$ ( $\mu\text{m}$ )	$D_{10}$ ( $\mu\text{m}$ )	$\text{Al}_2\text{O}_3$ (g/kg)	$\text{CaO}$ (g/kg)	$\text{Fe}_2\text{O}_3$ (g/kg)	$\text{MgO}$ (g/kg)	$\text{SiO}_2$ (g/kg)	$\text{P}_2\text{O}_5$ (g/kg)	
Value	200	31	2	110	288	28	33	222	106	19
Analysis	True density ( $\text{kg}/\text{m}^3$ )	Specif. Surf. ( $\text{cm}^2/\text{g}$ ) <sup>d</sup>	Leaching <sup>e</sup>			Trace elements <sup>f</sup>				
			$\text{Cl}^-$ (g/kg)	$\text{SO}_4^{2-}$ (g/kg)	pH	Zn (g/kg)	Pb (g/kg)	Cr (g/kg)	Cd (g/kg)	Hg (g/kg)
Value	2777	8150	$1.6 \pm 0.5$	$22 \pm 4$	$7.7 \pm 1.8$	12.7	4.7	1.02	0.23	n/a

<sup>a</sup> Laser size distribution.

<sup>b</sup> X-ray fluorescence.

<sup>c</sup> At 1000 °C.

<sup>d</sup> Blaine specific surface (EN 196-6 Standard).

<sup>e</sup> According to EN 12457-2 standard.

<sup>f</sup> Inductively-coupled plasma.

31% for the test section and 36% for the reference section, which matches existing figures [9].

Measurement of leached volumes over time provides a description of the  $L/S$  ratio evolution for each section. As compared to the previous and subsequent periods, the winter period (from October 2002 to February 2003) is characterised by increased percolation. The trend for both sections is similar. In agreement with its lower infiltration rate, the test section displays a final  $L/S$  ratio of 0.44 l/kg, whereas the reference section stands at 0.59 l/kg.

#### 4.3. Leachate quality monitoring

The set of parameters analysed through the end of the study for both sections consisted of: pH, electrical conductivity, redox potential, suspended matter, Kjeldhal nitrogen, ammonia nitrogen, nitrates, nitrites, total organic carbon (TOC), phenol index, total hydrocarbons, fluorides, sulphates, chlorides, aluminium, arsenic, iron, mercury, manganese, molybdenum, selenium, tin, antimony and barium. Among these parameters however, the comparison between leachates from both sections shows that some of them are not differentiated: concentrations are similar, or even lower in the test section leachate (notably Al, As and Hg). Chromium and chromate were detected only in the test section. The significant parameters of TFA leachate quality ultimately appear to be chlorides, sulphates and chromium.

Chlorides show high concentrations (up to 4000 mg/l), with the same trends recorded on both sections. An initial difference of +1000 mg/l is observed in the test section leachate, which disappears within 4 months. This difference has been explained by the Meurthe River origin of the aggregate used in the draining sub-base (salty watershed). Because of these high chloride concentrations, the measurements of COD and AOX were systematically wrong for both sections. The winter period is marked by an increase in chloride concentration, in relation to de-icing operations conducted on the road. Sulphates also show higher concentrations in the test section (up to 2500 mg/l) than in the reference section leachate (15–2300 mg/l and typically 100 mg/l). Concentrations in both chlorides and sulphates in the

test section leachate are always above 300 mg/l. The chromium concentration in the test section leachate moves between 0.10 and 0.20 mg/l, with a peak of 0.80 mg/l on one occasion (March 2003). As regards chromate, with the exception of one sample during the initial period, concentrations always remain strong (between 0.06 and 0.29 mg/l). The evolution of chromium and chromate concentration in the leachates of the test section is presented in Fig. 3 (the detection limit for both is 0.05 mg/l). In the reference section leachate, a significantly higher level of aluminium is observed (0.08–0.75 mg/l), as compared to the test section (0–0.18 mg/l).

#### 4.4. Calculation of releases

At the end of the monitoring campaign, cumulative chloride releases from both sections are identical (500 mg/kg). A difference between the two sections only appears during the first half of the study. Releases increase during the winter period and then subside as of March 2003. On the other hand, the final sulphate release from the test section (250 mg/kg) is higher than that from the reference section (150 mg/kg). The latter does not change from October 2002 onwards, while the former keeps on increasing, although more slowly during the spring–summer period than during the previous winter rainy season. Cumulative

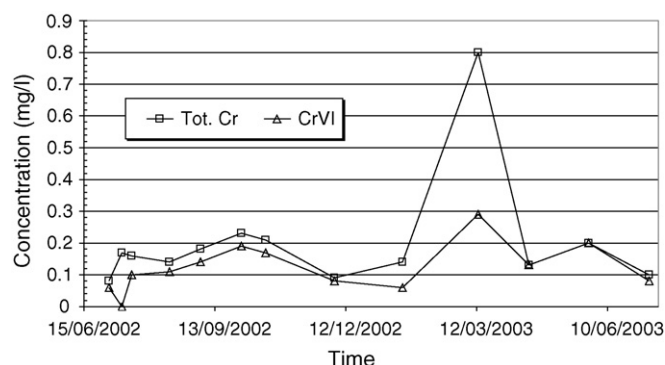


Fig. 3. Leachate concentration in chromium and chromate from the test section.

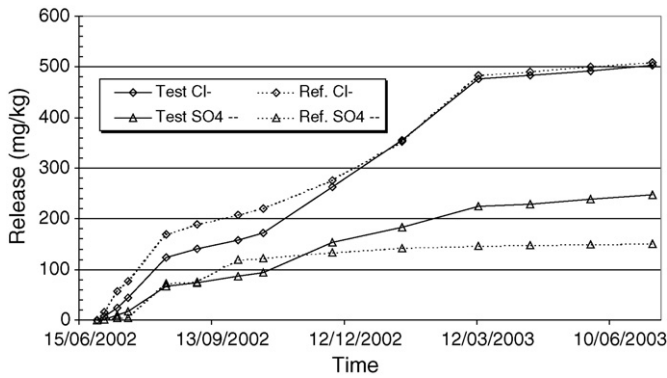


Fig. 4. Cumulative release of chlorides and sulphates from both sections.

releases are presented in Fig. 4 for both the test and reference (denoted “Ref.”) sections.

The chromium release from the test section increases exponentially from the opening of the road until March 2003, from which time it almost stops moving (see Fig. 5). The final release is 0.12 mg/kg. The chromate release evolves similarly since it always represents at least half the total chromium release; the final chromate release equals 0.06 mg/kg.

#### 4.5. Physicochemical parameters

Upon opening the road to traffic, the electrical conductivity of the test section leachate is perceptibly higher ( $>15$  mS/cm) than that of the reference section ( $<9$  mS/cm). This finding agrees with the initial release of chloride and sulphate. It rapidly decreases to 2.5 mS/cm during autumn 2002, before increasing again during the winter de-icing period and regaining a new equilibrium level in spring 2003, at below 4 mS/cm.

Due to implementation of the cement-bound graded aggregate, the initial pH of the reference section leachate is alkaline (pH 11), while that of the test section is neutral to slightly acidic (Fig. 6), with respect to TFA characteristics. The pH of both sections quickly converges towards an equilibrium value of 8 (reached in September 2002). In October 2002, a spillage of quicklime on the test section resulted in a sudden pH increase (pH $\approx$ 12), the effect of which lasted until March 2003. The phenomenon reached the reference section a few months lat-

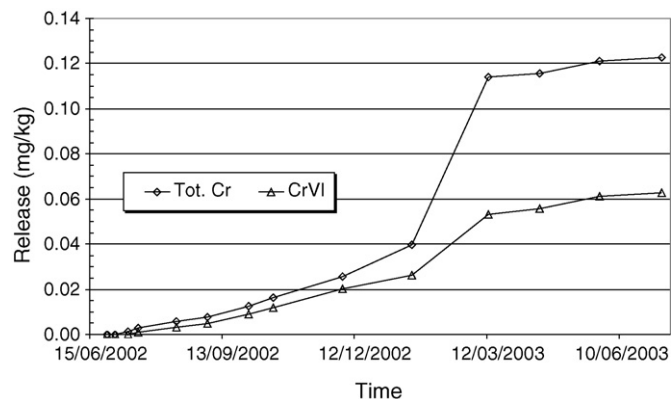


Fig. 5. Cumulative release of chromium and chromate from the test section.

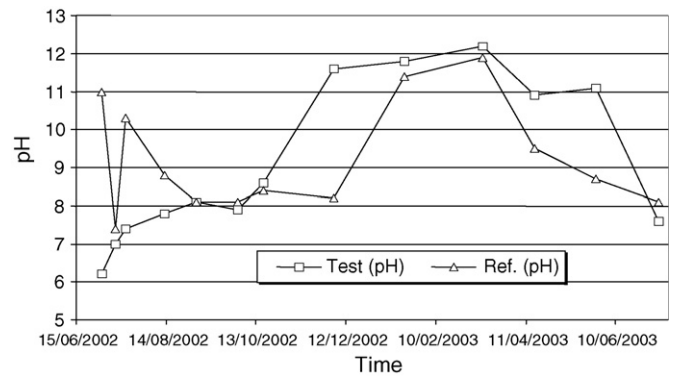


Fig. 6. Evolution in pH over time for both sections.

ter (January 2003) with the same intensity. Both sections only really recovered their equilibrium pH upon completion of the study, i.e. after 9 and 7 months, respectively.

The redox potential measured on-site is quite stable over the whole monitoring period; for both sections, Eh evolves between 230 and 370 mV, i.e. around the redox neutrality of water for the observed range of pH [10].

## 5. Discussion

From the initial list of 34 parameters defined for leachate analysis (based on known TFA characteristics and water regulation constraints), some of them rapidly and permanently display values below analytical detection limits. For both sections, such was the case for total cyanides, cadmium, copper, nickel, lead and zinc. For these latter metals, the results indicate the efficiency of phosphation stabilisation.

The comparative analysis of release between the test and reference sections reveals no effect of TFA regarding chlorides (a high release of 500 mg/kg after 1 year), thus demonstrating high efficiency in chloride removal from the process. In contrast, sulphate release from the test section exceeds that from the reference section (250 mg/kg compared to 150 mg/kg after 1 year, respectively). The concentration however tends to become similar between the two sections and then decreases slowly towards the World Health Organisation’s drinkable limit value (250 mg/l).

While no chromium release is detected from the reference section (concentration always below 0.5 mg/l), in the test section a high value appears: 0.12 mg Cr/kg of road base (half being constituted by chromate). The studied fly ash sample was intentionally chosen however for its particularly high chromium content ( $1.02 \pm 0.34$  g/kg of the insoluble fraction), although after 9 months this release almost ceased.

For all other elements, releases from the test section are similar to those of the reference section (fluorides, nitrates, nitrites, ammonia nitrogen, molybdenum, selenium, antimony, barium, iron, phenols, total hydrocarbons and TOC) and, in some instances, even lower (aluminium, arsenic, mercury).

The high and persistent accidental pH increase from October 2002, as well as the outlet of chlorides, or the high rate of infiltration ( $>30\%$ ) after a complete annual cycle demonstrates a

relatively high permeability of the road structure to various external influences. This can be partially explained by the thickness and transverse slope of the surface course, which were intentionally reduced in comparison with standard road practices [11], in order to enhanced rainfall infiltration into the road structure. Under more typical conditions, percolation would probably be lower during the 1st year, but would then progressively rise with increased surface course wear (cracking, stripping). However, after 1 year, no crack was observed on the road surface.

The sudden accidental pH gain observed as of autumn 2002, while both study sections had reached physicochemical equilibrium, provided an opportunity to observe the effect of a possible hazard during the life cycle of an actual road structure [12]. This event has proved to be responsible for an extended disruption of the system equilibrium. However, despite a high variation of pH in the test section during the 1-year monitoring (from 6.5 to 12), no significant variation of leachate concentrations was observed. This shows the efficiency of the treatment process. In addition to this, after the disruption, the pH of both systems (test and reference sections) converges again toward the value of 8 (its initial stabilized value). Such a stability of the system is important to consider regarding its longer-term behaviour.

## 6. Conclusion

The 1-year monitoring campaign conducted on a large number of water quality parameters ultimately highlights just three chemical parameters for which instant concentrations in the test section leachate and releases are significantly higher than in the reference section, i.e. sulphates, total chromium, and chromate. It must be kept in mind however that the TFA used in this experiment had been chosen for its high chromium content and potential release, out of a group of 10 samples collected from across Europe.

Percolation through the road structure and the subsequent release were enhanced by the test road design, in comparison with a classical road structure. On the other hand, with the exception of chromium (whose solubility may have been temporarily modified), the sizeable accidental pH increase did not result in any major impact on the release of the chemical species studied herein. The lesson provided by such an experiment is of interest to the entire scope of using alternative materials in road construction.

The monitoring campaign conducted in the framework of the *Revasol* project was rather long and detailed compared to similar field studies, however, despite positive results (obvious efficiency of the treatment process and stability of the system resulting in low release), its duration is short compared to the life of an actual road. The monitoring of leachate is continuing but at

the end of the experiment, due to the regulatory classification of MSWI fly ash (and as a consequence of TFA) as hazardous waste, the test section of the experimental road will be dismantled.

## Acknowledgments

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