IRON AND MANGANESE SURVEY IN AQUIFERS SURROUNDING LANDFILLS IN WALLONIA (BELGIUM)

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SUMMARY: Fe(II) and Mn(II) are first importance markers for characterizing and surveying aquifers contaminated with landfill leachates. However, they are not included in lists of required parameters imposed by Belgian or international environmental policiesn probably because they are not hazardous. In this study, aquifers under 4 municipal solid waste (MSW) landfills have been sampled using existing monitoring wells. It covers four different types of aquifers. For each of them, sharp increases of Fe(II) and Mn(II) concentrations are observed in some of the wells downgradient from the sites. Regional mean and maximal values for corresponding aquifers can be strongly exceeded without detecting significant anomalies of other markers.

1. CONTEXT

The groundwater biogeochemistry around waste disposal facilities has been widely studied since twenty years (Christensen & al, 2001). It is well known that leachates produced by water percolation through waste solid often contain high concentrations of dissolved Fe and Mn (Christensen & al, 1997).

Fe(III) and Mn(IV) also enter in the composition of aquifers matrix (silts, sand, rocks). It constitutes a second reserve of electrons acceptors for further anaerobic biodegradation in aquifer environment.

Previous works reviewed by Chrinstensen & al (2001) demonstrate that many parameters influence the Fe(II) and Mn(II) concentrations measured in the groundwater and their spatio-temporal variations downstream landfills. Among others the following are often cited:

- The chemical composition of source leachates (Andreotolla & al, 1990) ;
- The redox potential (Eh) of the aquifer, which is linked to dissolved O_2 concentration in the water, thus to sampling depth (Appelo and Postma, 1994 - chapt. 7; Cozzarelli et al, 2001) ;
- The natural background concentrations directly depending on aquifer matrix (sand, silt, rocks) composition (Polo-C. et al, 2003) and its heterogeneity (Scholl & al, 1999);
- The groundwater level (McGuire & al., 2000), varying with climatic conditions, weighted by the aquifer response time;
- The hydrogeological parameters such as dilution rate, transmissivity, transport and dispersion parameters (Boulding, 1995 - chapt. 4 ; Appelo and Postma, 1994 - chapt. 9) ;
- The sampling materials and methods (Boulding, 1995 – chapt. 5).

Space-time distribution and evolution of Fe(II) and Mn(II) in aquifer is thus, up to now, not yet fully understood. However, one observation is common to every fully investigated site downstream of waste disposals: high rated Fe-Mn zones are formed downstream landfills.

Therefore, iron and manganese should be considered by environmental authorities and landfill owners as important markers when achieving aquifer quality survey around waste disposals. On the other hand, Fe and Mn are not included in the lists of "hazardous materials" published by environmental authorities or health organisations. They are neither toxic nor carcinogen. Consequently, little environmental policies make them required for aquifer periodic control around landfills. In particular Fe(II) and Mn(II) are not listed as "to be controlled groundwater parameters" in waste management legislations in Belgium (Wallonia). Neither "sectoral conditions" nor landfill exploitation licences include these two parameters. In practice, most of aquifer surveying programs achieved semi-annually for many years by landfill owners do not include any iron or manganese analyses.

In addition to these imposed periodic auto-controls, the environmental authorities of Wallonia (DGRNE) realize independent environmental surveys of every municipal solid waste (MSW) landfills. ISSeP has been fulfilling this surveying mission for 10 years with freedom to select sampling points and control frequencies, investigation methods and chemical analysis protocols. Within this framework, Fe and Mn concentrations have been, at least once, measured in aquifers around each landfill. For four particular landfills, those analyses revealed significant iron and manganese enrichments in groundwater downstream actual or old filling zones. This constatation led to a specific investigation, focused on Fe-Mn problem, which is presented in this paper.

2. SITES SELECTION AND DESCRIPTION

DGRNE's landfills surveying network comprises 12 MSW landfills located all around the Wallonia. It allows investigating various exploitation methods (filling of abandoned quarries, tumulus building and topographic depression levelling) and varied panel of aquifer types (primary fractured rocks, secondary chalk, tertiary sand, quaternary alluvial deposit). On each location, sampling wells have been installed by owners following their environmental licenses. The four investigated sites have been selected with the aim of conserving this variety with the additional condition to have high iron and manganese in at least one observation well.

The selected sites for Fe-Mn specific investigations are Tenneville (TEN), Cronfestu (CRO), Mont-Saint-Guibert (MSG) and Happe-Chapois (CHA).

2.1 TEN landfill

TEN is a "tumulus building" landfill exploited since 1979. The first filling cell (phase 1 on fig. 1.a) was exploited during the eighties. It had not been equipped with adequate composite basis liner under the waste mass. This part of the landfill has been rehabilitated with conform top-liner and biogaz recuperation system. The second tumulus (phase 2) has been finished in 2006. The exploitation of the third sector just began in 2007.

The first aquifer beneath the landfill is located in the fractured upper part of primary bedrock, constituted of Praguian sedimentary rocks (shale, siltstone and sandstones). The local weathered fringe is thin (no more than 1 m). The high proportion of shale gives to the aquifer low transmissivity and limited capacity for exploitation. The groundwaterflow is strongly influenced by superficial relief and drained by a brook. Flow direction is first northwards, before deviating to the west.

Six piezometers are available on the site (figure 1a), which were all sampled for this work.

2.2 CRO landfill

CRO landfill (Figure 1b) is an old chalk quarry that was filled with domestic waste from the sixties until the year 1994. No basis liner has been installed. The upper part of the landfill has already been rehabilitated with upper composite liner. The site is in post-management phase. In the North of the landfill, a second quarry was filled with non-hazardous industrial waste (NHIW).

The chalk aquifer was higher than the bottom level of the quarry in its early stage of filling. Due to large scale exploitation it has been remaining a few meters under this level for more than 10 years, even during high-water periods. Under the landfill, the chalk watertable shows a 1,6% gradient to the West. Local measured transmissivity varies between 0,9 and $13,0.10^{-3}$ m^2/s for a thirty meters thickness saturated layer. Chalk presents double porosity (pores and fractures) giving very good exploitability.

Height monitoring wells are available around the site (see Figure 1b). They were all selected for sampling.

2.3 MSG landfill

MSG is one of the biggest landfill in Wallonia. Its exploitation begins in 1958 at a small scale with the filling of a first old sand quarry. The filling of the main second quarry followed the first project in two phases. The first phase (northeast) has been exploited at low rhythm during the eighties. These two first waste masses ("old disposal" and "phase 1" on figure 1c) have been stored without basis liners. The second phase is exploited since 1994 with best available techniques. A small zone of actual exploited area is reserved for NHIW disposal.

The groundwater level in tertiary sand aquifer under MSG is among one meter lower than the bottom of the original sand quarry. The groundwater flow is globally oriented westwards. Surface-water drainage creates local curvature in water table contour with flow direction deviating to the North-West and to the South-West, especially in the West part of landfill. Directly downstream from the landfill, the aquifer thickness reduces fast, giving to the water reserve lower potentiality for exploitation. Local transmissivities were measured from 0,12 and $1,8.10^{-3}$ m²/s.

A wide network of 30 sampling wells has been installed in two concentric rings. Risers of some wells cross sand aquifer. Others have been bored deeper and equipped in the underlying bedrock aquifer. For this study, 11 sand wells have been selected for sampling, among which one (P20) is upstream and 3 belong to the second ring.

2.4 CHA landfill

CHA landfill is as smaller landfill (Figure 1d). The exploitation method consists in levelling a natural small valley gradually (from its head). In the past, a small intermittent superficial flow was present in the bottom of this depression. Before starting the work, this flow has been collected in a pipe. Nevertheless, some saturated alluvial sediment remains present under the waste disposal. Phase I (northwest) has been achieved without bottom liner. Phase II has been designed with needed protection avoiding leachates leakage.

Under the landfill, alluvial water is in contact with the groundwater located in the weathered and fractured primary bedrock (upper Devonian). This bedrock is constituted of two distinct geological formations. The first one ("Famenne" layer) is mainly composed of shale's and has aquiclude hydrogeological properties.
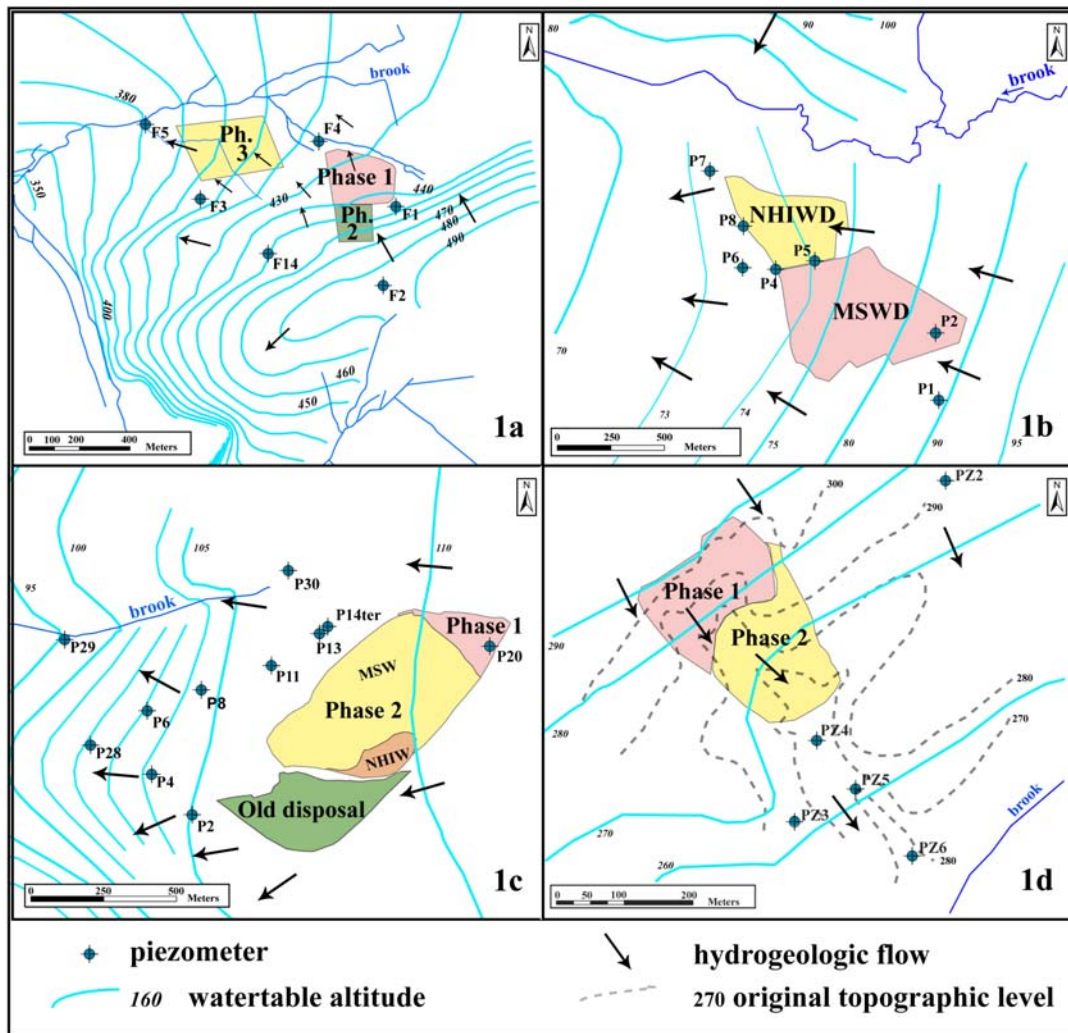
Figure 1: schematic representation of TEN (1a), CRO (1b), MSG (1c) and CHA (1d) landfills – position of selected monitoring piezometers and simplified groundwater table contours

In the southeast part of the site, "Famenne" shales are covered by "Esneux" formation. It is constituted of siltsones and sandstones. The weahthered fringe of this geological layer is more or less sandy and contains groundwater, which is also stored in deeper fractures of rock. Hydrogeological characteristics of Esneux aquifer are rarely good enough for large scale exploitation. On CHA location the thickness of permeable layer is low, giving a watertable strongly influenced by topographic relief. It is drained in the direction of a brook (southeast), then following brook's flow (south-west).

Six wells are available for groundwater monitoring. Each of them has been sampled in this study. Upstream well (Pz2) 1 is located in the "Famenne" shale formation when the others are in the "Esneux" siltstones.

Table 1: Regional background concentrations (RBG) computed by Polo-C. & al (2003) compared with concentrations measured upstream studied landfills

		"Ardennes" bedrock		*TEN*	"Mons bassin " chalk		*CRO*	"Bruxelles" sand		*MSG*	"Esneux" bedrock		*CHA*
		RBG			RBG			RBG			RBG		
		mean	max	*F2*	mean	max	*P1*	mean	max	*P20*	mean	max	*Pz2*
Fe $_{tot}$	µg / l	240	4540	*90*	140	4000	*16*	32	220	*24*	31	138	7460
Mn $_{tot}$	µg / l	47,7	878	*30*	5	76	*<6,3*	3,1	30	*<6,3*	6	43	404
Cl $^-$	mg / l	16,2	103	*27*	44,8	93,2	*47*	47,2	68,8	*62*	23,1	73	24
NH$_4^+$	mg / l	0,001	0,06	*<0,05*	0,08	0,76	*<0,05*	0,01	0,09	*<0,04*	0	0,03	*<0,04*
Ni	µg / l	5,63	27,8	*26*	4,33	17,7	*7,4*	2,89	14,5	*<3,1*	2,69	21,5	70

Table 2: Fe-Mn grades measured during former ISSeP study in TEN, MSG and CHA leachates.

		TEN	MSG	CHA
Fe $_{tot}$	µg / l	13431	5203	1490
Mn $_{tot}$	µg / l	4344	373	2054

2.5 Background concentration in aquifers

Table 1 presents mean and maximum regional background concentrations (RBG) of common "waste leachates tracers" according to Polo-C. & al (2003). These background values were computed from DGRNE's groundwater database and water supply company's data for each regional aquifer. It shows that upstream piezometers at TEN, CRO and MSG present normal or weak iron and manganese total contents compared with regional data. On CHA, iron and manganese upstream values are higher than regional maxima, but Pz2 is implanted in "Famenne" formation, not included in RBG computation as it is not exploited for water supply.

2.6 Leachates composition

Chemical composition of CRO remains unknown because it is totally "encapsulated" without sampling possibility. The other leachates are controlled, for a few tracers, by owners every year and, for a largest set of parameters, during each ISSeP campaign. Table 2 present total iron and manganese values measured by ISSeP in previous campaigns. TEN's leachate shows the absolute highest Fe-Mn concentrations. MSG's leachate presents relative lower Mn content than the others. CHA's leachate is the only one where Mn is higher than Fe.

3. SAMPLING AND CHEMICAL ANALYSES METHODS

DGRNE's landfills surveying network do not dispose of proper budget for placing monitoring wells. ISSeP's sampling campaigns have been achieved using existing sampling wells. The position of the monitoring wells was thus imposed before the work, and not always optimised as suggested in scientific literature (Christensen & al, 1992). Moreover, these wells were designed with two requirements: providing sampling points for survey and leaving the possibility of pumping to stop or recover pollution if it happens. They were thus designed as water collecting wells with large diameters, long strainers and without multilevel sampling possibilities. In particular, MSG and CRO samples are representative of the total aquifer's thickness whereas TEN and CHA samples characterize 15 to 20 meters of fractured rocks. With such sampling

devices, the vertical redox zonation observed on "reference landfills" such as Grindsted, Denmark (Bjerg & al, 1995), Norman, USA (Cozzarelly & all, 2000) or Borden, Canada (Nicholson & al, 1983) is unobservable.

On site filtration (0,45 µm) was achieved for determining Fe(II) and Mn(II) content. Samples have been analysed by ISSeP laboratories (reference labs in Wallonia) for a wide set of inorganic and organic parameters using normalized analytical methods.

4. RESULTS

Tables 3 to 6 present most significant results for each investigated landfill. In those tables, "chlorinated HC" values are the sums of PCE, TCE and 1,2cis-DCE measured concentrations.

On TEN landfill (Table 3), results shows strong Fe(II) and Mn(II) enrichment in three downstream observation wells. Measured concentrations are slightly higher maximal regional concentrations (Polo-C. & al, 2003). Observed downgradient/upgradient factors are very high. Only for the closest piezometer (F4), other anomalies are detected (higher conductivity, Cl^-, NH_4^+, TOC, SO_4^{--} and Ni) but in much weaker proportions. F5 and F14 are only high in Fe(II) and Mn(II) (and Ni in F14): even Cl^-, often considered as conservative ion, is not detected. There are two possible explanations to these observations:

- First explanation: higher Fe-Mn concentrations in F5 and F14 are due to natural geochemical variations. The structure and composition of Ardennes bedrock allows such interpretation: folded geology can create brutal change in lithology with higher or lower Fe(III)-Mn(IV) grade in rocks.
- Second explanation: Fe(II) and Mn(II) high concentrations are due to the dispersion and degradation of a leachate contamination but concentrations of other contaminant are too weak to be detected.

Fe(II) and Mn(II) results in Table 4 show a strong impact of leachate contamination on two CRO downstream observation wells (P5 and P4). From up to downgradient, values are multiplied by more than 100 (P5) and 10 (P4) reaching two times the maximal values measured regionally in the chalk aquifer (Polo-C. & al, 2003).

Table 3: Groundwater geochemistry around TEN landfill

		upstream		downstream			
		F2	F1	F3	F4	F5	F14
conductivity	µS / cm	154	101	92	269	167	119
Fe^{++}	µg/l	70	87	39	4243	5613	4428
Mn^{++}	µg/l	30	64	620	1141	636	877
Cl^-	mg / l	27	10	4,2	50	3,5	3,5
NH_4^+	mg / l	<0,05	<0,05	<0,05	0,40	<0,05	<0,05
SO_4^{--}	mg / l	11,1	2	2,1	8,4	2,6	1,52
TOC	mg C / l	0,50	<0,3	<0,3	1,8	<0,3	0,7
Ni	µg/l	26	12,9	3,7	63	12,8	57
chlorinated HC*	µg / l	<0,3	<0,3	<0,3	<0,3	<0,3	<0,3
BTEXN	µg / l	<0,7	<0,7	<0,7	<0,7	<0,7	<0,7

Table 4: Groundwater geochemistry around CRO landfill

		upstream		downstream				
		P1	P2	P4	P5	P6	P7	P8
Conductivity	µS / cm	941	777	909	1620	917	877	874
Fe^{++}	µg / l	4,4	50	491	7792	2	2,6	2,86
Mn^{++}	µg / l	<5	<5	49,6	220	11,5	<5	< 5
Cl^-	mg / l	47	39	49	188	42	47	35
NH_4^+	mg / l	<0,05	<0,05	4,3	45	0,69	<0,05	<0,05
SO_4^{--}	mg / l	234	156,6	214	83	187,8	154	179
TOC	mg C / l	0,9	0,9	3,4	21,9	1,4	1,7	0,9
Ni	µg / l	7,4	4	10,4	31,6	8,8	8,1	7,5
chlorinated HC*	µg / l	<0,3	<0,3	<0,3	<0,5	12,9	8,1	8
BTEXN	µg / l	<0,7	<0,7	<0,7	<0,8	<0,7	<0,7	<0,7

The plume shows limited axial and lateral extensions. Fe-Mn concentrations evolve in the same way as other leachates markers (high Cl^-, TOC, COD, Ni and trace of chlorinated solvents) Other redox attenuation phenomenon's are observed (SO_4^{--} consumption and NH_4^+ oxidation). Fe and Mn concentration are probably mainly governed by aerobic oxidation above and just below water table level. Dispersion-dilution phenomenon dominates deeper evolution in the chalk. Secondary anaerobic reduction is not possible due to the lack of Fe(III)-Mn(IV) in aquifer's matrix (pure $CaCO_3$).

Geochemistry around MSG (see Table 5) is better known thanks to more available wells. Fe(II) and Mn(II) increase strongly from upgradient to downgradient, reaching respectively 5 and 50 times maximal regional values. The conservative behaviour of chloride ion is better observed than in CRO and TEN monitoring wells. The plume is also larger, probably due to multiple sources (old disposal and phase 1) and to the wider range of flow directions. The measured Fe/Mn proportion shows variable values in "first ring" wells: Mn predominates in the South part whereas Fe is in majority in the North part.

Table 5: Groundwater geochemistry around MSG landfill

		upstr.	downstream									
		P20	P2	P4	P6	P8	P11	P13	P14ter	P28	P29	P30
conductivity	µS / cm	480	2010	1571	1335	761	3060	521	238	1201	1331	1300
Fe^{++}	µg / l	5	6272	327	< 2	506	1103	17,9	892	2,2	2,1	495
Mn^{++}	µg / l	< 5	1875	1466	64	<5	794	<5	667	201	91	224
Cl^-	mg / l	62	238	170	173	65	519	41	49	139	158	n.m.
NH_4^+	mg / l	<0,04	96	32	< 0,04	<0,	0,05	<0,04	1,66	<0,04	<0,04	n.m.
NO_3^-	mg / l	19,3	12	15,4	1,27	37	<0,1	11,9	0,23	4,7	3,9	n.m.
SO_4^{--}	mg / l	81	70	89	81,8	<0,04	42,2	69	52	77	110	n.m.
TOC	mg C / l	1	28,7	18	11,7	1	54	0,5	2,4	10,9	13,8	n.m.
Ni	µg / l	<3,1	38	52	31	3,5	133	<3,1	11,4	29	42	33
chlor. HC*	µg / l	<0,3	<0,8	<0,3	<0,3	<0,3	<0,4	<0,5	<0,3	<0,3	<0,4	n.m.
BTEXN	µg / l	<0,7	<0,8	<0,7	<0,7	<0,7	<1,4	<0,7	<0,7	<0,7	<0,7	n.m.

Table 6: Groundwater geochemistry around CHA landfill

		upstr.	downstream			
		Pz2	Pz3	Pz4bis	Pz5	Pz6
conductivity	µS / cm	280	370	322	338	393
Fe_{tot}	µg / l	7460	1857	972	16	9914
Fe^{++}	µg / l	19	5,1	11,5	<2	11,4
Mn_{tot}	µg / l	404	125	173	234	513
Mn^{++}	µg / l	83	<5	158	144	29
Cl^-	mg / l	24	26	17,9	11	43
NH_4^+	mg / l	<0,04	<0,04	0,067	<0,04	<0,04
NO_3^-	mg / l	89	38	32	0,81	3,7
SO_4^{--}	mg / l	14,5	30	28	47	46
TOC	mg C / l	1,1	1,5	1	0,5	0,8
BTEXN	µg / l	<0,7	<0,9	<0,7	<0,7	<0,7

There's not enough data to explain such variation. It can be due to double sources with different original proportion, to aquifer's heterogeneity or to REDOX zonation. At the level of the second ring (P28, P29), normal Fe(II) concentration are recovered whereas Mn(II) remains higher than upstream value. It could be an illustration of the further axial position of manganese peak in leachates contaminated aquifers (Christensen & al, 2001 – fig. 3). At P30 (third « second ring well ») iron remains higher than upgradient. Composition of P13 water is strange as it seems not polluted whereas it is between P14ter and P11, both clearly influenced by leachate.

Geochemical results at CHA location are quite different (see Table 6). One had selected the landfill on basis of older Fe_{tot} and Mn_{tot} measurements. On Fe** and Mn** analyses, one can see that iron and manganese detected are almost total in solid oxidised phase. There is no clear evidence of an impact of landfill leachate on groundwater quality. Moreover, high Fe-Mn total concentrations are measured in upstream Pz 2, which contribute to complicate the interpretation.

Chlorinated and aromatic hydrocarbons are quite never detected in any monitoring wells of any location. It could be explained by their complete elimination by natural attenuation. It's also possible that dilution induced by monitoring wells design reduces their concentrations enough to make them undetectable by analytical devices.

5. DISCUSSION

Aquifers around four MSW landfills have been investigated using sampling wells that were dimensioned as water exploitation pumping wells. Moreover there are often too little sampling points positioned in a not optimal way. The available sampling facilities are far from full scale 3D sampling networks required for complete understanding of leachate contaminated plumes in groundwater. It gives a partial and "smoothed" image of the contaminated zones. However it allows obtaining some interesting results that can be resumed as follow:
- Whatever the type of aquifer, leachates contamination plumes are limited in space, which confirms, but on varied aquifer types, the observations made on reference sites in sandy aquifers.
- The only phenomenon observable by mean of any sampling network on any site seems to be substantial Fe(II) and Mn(II) concentration increases in downstream sampling wells. Iron and

manganese down/up factors are higher than those of any other leachate marker. This could be explained by the vertical distribution of redox zones as described on reference locations (Christensen & al, 2001): high concentrations of Fe(II) and Mn(II) are often detected on wider range of depth than any other redox markers or leachate conservative contaminant.

- Associated nickel and chlorides anomalies are often observable but not systematically and in weaker proportions. In particular, the apparent behaviour of chloride seems less conservative in fractured aquifers conditions.

Without controlling Fe-Mn concentrations, conventional monitoring wells design are thus not sufficient for evaluating impact of landfills on underlying aquifers, at the scale of their total thickness. On the other hand, monitoring wells designed with full aquifer thickness risers has a big advantage. It gives the concentrations that would be measured at a domestic water tap if one pumping well was installed at the same place. It gives thus the best evaluation of sanitary risks linked to the groundwater contamination. At this rough measuring scale, the effect of a landfill is to create a limited area of ferro-manganeous water. In case of exploitation, such water would not be toxic, but iron and manganese oxides would precipitate. It would create aesthetic (brown water) and water pipeline stopping problems.

5. CONCLUSIONS

By undertaking a sampling campaign focused on iron and manganese detection in groundwater around 4 MSW landfills in Wallonia, ISSeP hoped to open a discussion with scientific and politic authorities. The obtained results presented above are consistent enough to achieve this goal. They clearly demonstrate to administration that iron and manganese are required parameters for efficient detection and characterisation of leachates contaminated plumes. The study also worked on a more varied panel of aquifer types than previously. For each situation, it contributes to demonstrate that natural auto-remediation potentials of aquifers are high enough to avoid large-scale dispersion of contaminants.

ISSeP aims at looking further into this discussion by launching future dialog with local and international environmental authorities in order to make include Fe and Mn in future policies. At the scale of DGRNE's control network of MSW landfill, the use of multilevel sampling techniques and the optimization of number and position of monitoring wells will be considered case-by-case with owners. From scientific point of view, this paper could help to initiate future collaborations with research teams specialized in the thematic of leachates contaminated aquifers. These teams hold essential knowledge's and know-how whereas DGRNE and ISSeP dispose of a wide variety of potentially interesting case studies, in an open network of sites.

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