Treated wastewater reuse for a seawater intrusion hydraulic barrier implementation in the Llobregat delta aquifer (Barcelona, Spain). First phase.

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Abstract. Sampling results of a wells network to monitor a hydraulic barrier for seawater intrusion control at the Llobregat delta deep aquifer (Barcelona, Spain) are presented here for the period March 2007-2008. The monitoring programme consisted in periodical water sampling and piezometric level control in wells, local network and wastewater treatment plant. The first-phase results of the sampling campaign show a decrease in electric conductivity in the wells closer to the injection points. Also a cation exchange index decrease is observed, indicating a continuous increase in the water mixing process in the wells. Regarding emerging contaminants, enhanced wastewater treatment removed the majority of contaminants before injection. The preliminary field results of hydrogeological control of injected water indicate that improved pre-treatment of wastewater has been appropriate for contaminants removal.

Keywords. Hydraulic barrier – Seawater intrusion – Treated wastewater – Llobregat aquifer – Microcontaminants.

Recyclage des eaux usées traitées pour la mise en place d'une barrière hydraulique à l'intrusion de l'eau de mer dans l'aquifère du delta du Llobregat (Barcelone, Espagne). Première phase

Résumé. *Dans le présent travail, on passe en revue les résultats de l'échantillonnage d'un réseau de puits mis en place afin de suivre une barrière hydraulique pour le contrôle de l'intrusion marine dans l'aquifère du delta du Llobregat (Barcelone, Espagne), dans la période mars 2007-2008. Le programme de suivi est basé sur le prélèvement périodique d'échantillons d'eau et le contrôle du niveau piézométrique dans les puits, le réseau local et la station de traitement des eaux usées. Les résultats de la première phase d'échantillonnage montrent une réduction de la conductivité électrique dans les puits les plus proches des points d'injection. Parallèlement, on observe une diminution de l'indice d'échange cationique, qui indique un accroissement continu du processus de mélange des eaux dans les puits. Quant aux contaminants émergents, l'amélioration du traitement des eaux usées permet d'éliminer la plupart des contaminants avant l'injection. Les résultats préliminaires du contrôle hydrogéologique des eaux injectées confirment l'importance d'améliorer le pré-traitement des eaux usées pour éliminer les contaminants.*

Mots-clés. *Barrière hydraulique – Intrusion de l'eau de mer – Eaux usées traitées – Aquifère de Llobregat – Micro-contaminants.*

I – Introduction

The reuse of water originating from sewage treatment plants for environmental applications (generally focussing on clogging processes) assumes that the possible polluting agents are eliminated through conventional treatment. However, not all polluting agents are eliminated through standard treatments (Ternes *et al.*, 2002). Between these "persistent" compounds is the emergent polluting agent group, constituted by chemical compounds of a highly diverse origin, characterised by its high production and consumption entailing its continuous presence in the environment. Currently, a great interest has been taken in studying the presence of these compounds (Sedlak *et al.*, 2000), whose effects on the environment and human health still remain unknown. Among them drugs (antibiotics, painkillers, etc.) are included, diagnosis products, steroids and hormones, antiseptics, personal care products (sun creams, perfumes, etc.), petrol additives, etc. Although in the literature a great number of studies dealing with the application of recycled waters exist, the quality criteria used (Levine and Asano, 2004) do not contemplate the presence of these polluting agents.

Spain is one of the countries that employs a great amount of recycled water although in insignificant total amounts; no more than 5% of collected wastewaters is recycled nowadays. For 2015, and following the new instruments for the integrated water management implementation, the reuse up to 1200 hm^3 is foreseen. In such respect, the new Royal Decree (1620/2007, BOE 294, 8 December 2007) establishes the legal regulations for water reuse plans, reclaimed water definition and treatments to achieve the sanitary and environmental quality for its further use.

In Barcelona metropolitan area, water is a limited resource and of great importance due to the presence of an extended industrial activity and water supply demand. In order to reduce this deficit, wastewater reuse from the Depurbaix sewage treatment plant (Cazurra, 2006), located in the Baix Llobregat, is intended, thus contributing to the sustainable management of water. The volume of water to be reused is destined to satisfy different demands such as the contribution to ecological flow, agricultural irrigation, maintenance of wetlands and the construction of a hydraulic barrier against the existing sea water intrusion in the Llobregat river delta deep aquifer.

The objective of this paper is to present the preliminary results from the first phase implementation of the hydraulic barrier against seawater intrusion in the Llobregat delta. Besides, the quality assessment of the applied wastewater treatment for emerging contaminants is a secondary objective of this research.

II – Geology of the Llobregat delta

The Llobregat river delta (Barcelona, Spain) (Figure 1) is a delta formation covering 97 km^2 formed by Quaternary material deposits of a detritic nature over materials of a Pliocene age, with the exception of the margins, which overly materials of an older age.

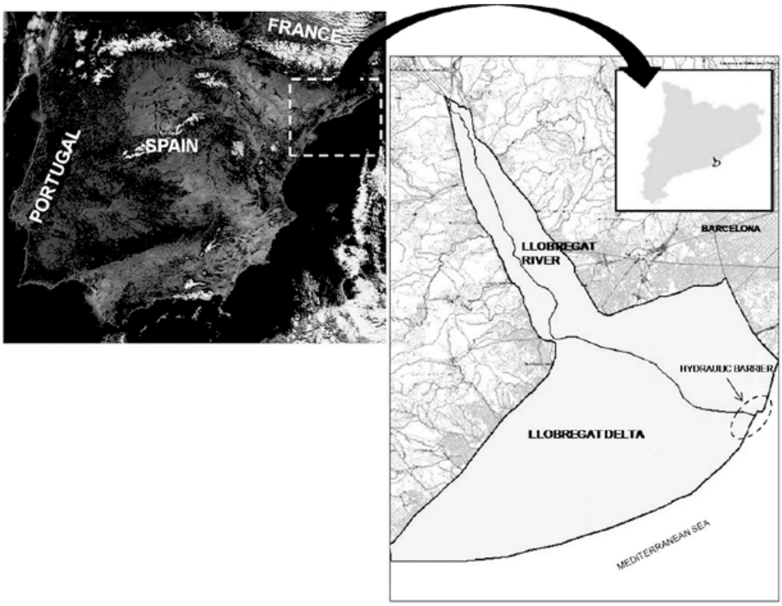

Figure 1. Situation of the study area.

This delta is characterised by two aquifers, the upper and lower aquifer which are separated by a silt wedge confining the lower aquifer, except at the borders of the delta, where this silt package disappears and the upper and lower aquifers are connected by fine sands. The lower aquifer, with an average thickness of 6 m constituted by fine sands and gravels, occupies almost the entire delta surface, extending under the sea below the silt wedge. This aquifer formation has been highly exploited and has produced a general lowering of the water table since the 1970's. This has led to the presence of sea water intrusion in different parts of the aquifer, which has been enhanced by excavations of the impermeable layer, the contact between aquifer and the sea. As a result, a progressive deterioration in the groundwater quality has been observed, which reduces the possibilities of exploiting the aquifer.

With the objective of controlling the sea water intrusion wedge produced in the lower aquifer, studies of different nature have been carried out. Among the numerous articles published, it is possible to mention, for example, Iríbar *et al.* (1997) and Abarca *et al.* (2006) focussing on the hydrogeological aspects, or those describing the construction of a hydraulic barrier by means of the injection of recycled water originating from the Depurbaix sewage treatment plant (Cazurra, 2006; Ortuño *et al.*, 2008).

III – Methodology

The hydraulic barrier consists of injecting treated recycled water to conform a pressure ridge along the coast, therefore stopping the sea water from entering the aquifer. Sampling is currently carried out by the Comunitat d'Usuaris d'Aigües del Delta del Llobregat (CUADLL), Agéncia Catalana del Aigua (ACA) and UPC. The analysis of the components is accomplished by different organisations: detection of major elements (ACA), polluting agents, priority substances and heavy metals (University of Almería and University of Jaén), microbiological analyses (Autonomous University of Barcelona) and analysis of toxicity (Department of the Environment, INIA).

IV– Injection wells

For the first stage, four injection wells were constructed (P1 to P4) at a depth of 70 m, that totally penetrate the lower aquifer (59-65 m). These wells are located 1500 m inland, aligned parallel to the coastline, separated by a distance of 300 m between them (Figure 2). The wells have a diameter of 610 mm, lined with stainless steel to 350 mm. After drilling, pump tests were carried out to define the hydraulic characteristics of the lower aquifer. Results obtained demonstrate that wells P1 and P3 show the highest transmissivity values (700 to 2000 m^2/day), whereas P2 and P4 present smaller transmissivity values (100 to 200 m^2/day).

The injection in wells P3 and P4 commenced in March, 2007. Later, injection began in P2, and in July, 2007, the injection in P4 ceased, leaving only P2 and P3 operative which is the current situation. The initial injected volume was 620 m^3/day during 8 hours per day, until November, 2007, when the volume was increased to 2500 m^3/day during 24 hours.

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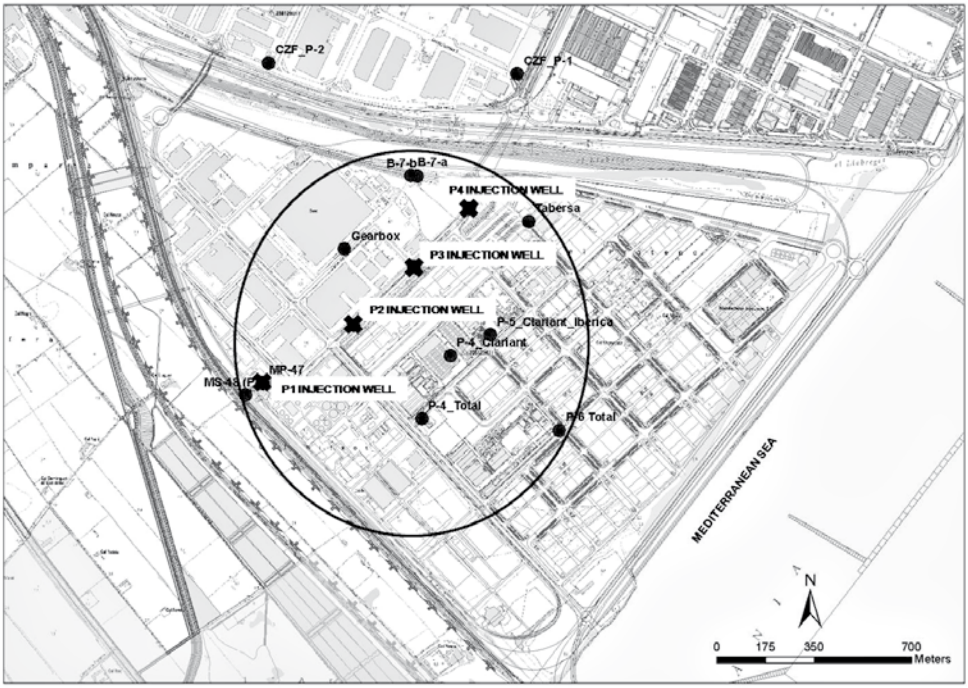

Figure 2. Situation of the injection wells and the local wells network.

V – Injected water

The injected water for the hydraulic barrier initially consisted of an equal mixture between treated wastewater and tap water, following a similar model of the Orange County Water District barrier (California). The water mixture was replaced by recycled water in March, 2008.

The injected treated wastewater consists of outflow water from the Depurbaix tertiary sewage treatment plant that receives an additional treatment. After the conventional tertiary treatment (coagulation-flocculation processes, lamellar clarification, filtration and UV disinfection), water is additionally treated by ultra-filtration, reverse osmosis and UV disinfection before being stored in a deposit for further injection in wells.

VI – Monitoring

In order to control and monitor the hydraulic barrier performance, samples of the influent and effluent of the tertiary treatment were taken, along with the injection water storage deposit and groundwater from the local network (located within a 1 km radius around injection wells). Monitoring started in March 2007.

On a fortnight basis, *in situ* measurements of temperature, dissolved oxygen, Eh, pH, conductivity and piezometric level are taken from wells in the local network and in the water tank for injection. Water samples of the local network and water to be injected are carried out every second month, while influent and effluent samples of the tertiary treatment water were carried out on a monthly basis since April 2007 until May 2008.

For the emerging contaminants and priority substances, 105 substances between which drugs, personal hygiene products, volatile metals, priority polluting agents, polycyclic aromatic hydrocarbons and other priority polluting agents have been selected (Table 1).

Before the injection period commenced on the 26th of March, 2007, samples were taken from injection and local network wells with the objective to determine the background level of the aquifer in respect to its contents of micropollutants. Until the present day 10 sampling events have taken place; however, not all the analytical data is currently available yet.

Table 1. Emerging contaminants and priority substances selected.

Pharmaceuticals	N-acetyl-4-amino-antipiryne (4-AAA) *	Hexachlorobenzene
4-amino-antipiryne (4-AA) *	Naproxen *	Pentachlorobenzene
4-dimethylaminoantipiryne (4-DAA) *	N-formyl-4-amino-antipiryne (4-FAA) *	Alfa-hexachlorocyclohexane
4MAA	Nicotine *	Beta-hexachlorocyclohexane
Acetaminophen	Ofloxacin *	Gamma-hexachlorocyclohexane (lindane)
Antipyrine	Omeprazole	Delta-hexachlorohexane
Atenolol *	Paraxanthine *	Alachlor
Benzafibrate *	Paroxethine	Tetra-brominated diphenyl ether
Biphenylol *	Propanolol hydrochloride	Penta-brominated diphenyl ether 2
Caffeine *	Ranitidine *	Penta-brominated diphenyl ether 3
Carbamazepine *	Salbutamol	**PAHs**
Carb,Epoxide	Sotalol	Acenaphthene
Cefotaxime	Sulfamethoxazole *	Acenaphthylene
Chlorophene	Terbutaline	Anthracene
Ciprofloxacin *	Triclosan *	Benzo (b) fluoranthene
Clofibric acid	Trimethoprim *	Benzo (a) anthracene
Codeine *	**Personal care products**	Benzo (a) fluoranthene
Diatrizoate	3-(4-methylbenzylidene) camphor	Benzo (a) pyrene
Diazepan *	Benzophenone-3	Benzo (k) fluoranthene
Diclofenac *	Celestolide	Chrysene
Erythromycin *	Ethylhexyl methoxycinnamate	Fluoranthene
Fenofibrate	Galaxolide *	Fluorene
Fenofibric Acid	Octocrylene	Naphthalene
Fenoprofen	Octyl-triazone	Phenanthrene
Fluoxethine	Phantolide	Pyrene
Furosemide *	Tonalide *	**Other priority pollutants**
Gemfibrozil *	Traseolide	2,3,7,8-tetrachloro-dibenzo-p-dioxin
Hydrochlorothiazide *	**Metals**	2,7/2,8-dichloro-dibenzo-p-dioxin
Ibuprofen	Ni *	Alpha-Endosulfan
Indomethacine *	Cd	Atrazine
Ketoprofen	Hg *	Beta-endosulfan
Ketorolac	Pb *	Chlorfenvinphos
Mefenamic Acid	**Volatile prioritary pollutants**	Chlorpyriphos-Methyl
Mepivacaine	1,2,3-trichlorobenzene	Diuron *
Methylprednisolone 6-alpha sodium succinate (Urbason)	1,2,4-trichlorobenzene	Endosulfan sulphate
Metoprolol	1,3,5-trichlorobenzene	Isoproturon
Metronidazole *	Hexachloro 1,3-butadiene	Simazine

*: Compounds with concentration higher than 0.1μg/l present in the tertiary treatment effluent.

VII – Results and discussion

The distribution of water volumes injected in the different wells was based on transmissivity data collected from pump tests once drilling had ceased, where more volume is injected in the higher transmissivity wells and the more saline ones. In July, 2007, P4 was abandoned due to malfunctioning, and since then P2 and P3 remain operative, through which an approximated proportion of 15% to 85% of the total volume is injected.

According to the records from September 2007, a volume of 286837 m^3 has been injected up until January 2008.

1. Piezometric evolution at the injection points

In the wells under operation, the dynamic water level is currently located at around -4.7 masl (P2) and -3.5 masl (P3), which represents an increase in the absolute level of 0.58 m and 1.68 m respectively. In agreement with the observed values, and considering the volume of water injected in each well, the flow rate/piezometric uplift relation seems to stay more or less constant, which indicates no evidence of possible clogging until now. Due to problems related to the data acquisition system and to the calibration of the wells pressure sensors, data were not available before September 2007, making piezometric level monitoring more difficult.

2. Evolution of the salinity

In spite of the reduced volume of water injected in the aquifer, a slight reduction of the electric conductivity has been detected in some points of the local network such as at Gearbox Prat (where the conductivity has decreased from 15,723 to 9,770 µS/cm), and in MP-47 and Clariant P4, where the reduction is smaller (around 200 µS/cm in both points). Both wells are located at 640 m and 327 m of distance from the injection wells (Figure 2).

3. Presence of emerging contaminants

From the analytical data available from samples taken from the tertiary treatment plant corresponding to the months of April, May and July (2007), it is observed that some compounds are not totally eliminated by the treatment process. A total of 33 out of the 105 micropollutants analysed were found in concentrations above 0.1 µg/l (accepted maximum value for micropollutants) (Table 1).

In regards to the samples taken from the injecting water deposit, it has been observed that micropollutants that were present in the tertiary treatment effluent tend to disappear after undergoing the further treatment of ultrafiltration, reverse osmosis and UV disinfection. Only one compound, Galaxolide, was found in a concentration greater than 0.1 µg/l.

Finally, for the local network points and injection wells, data is available for the months of March, June and July, 2007. The March sampling was carried out previous to the commencement of the injection with the objective to define the emerging compounds background state of the aquifer. In this initial sampling event, caffeine was detected at concentrations superior to 0.1 µg/l. In the following sampling events at the beginning of the injection period in June and July, concentrations superior to 0.1 µg/l were found for caffeine along with three other compounds: Paraxantine, Nicotine and Galaxolide. Caffeine, which has been detected in concentrations superior to 0.5 µg/l, is an alkaloid compound present in many common drinks; Paraxantine is a metabolite or degradation product of Caffeine. Nicotine, also a composed alkaloid, originates from the tobacco plant and is used in insecticides commonly employed in greenhouses. Galaxolide, found in concentrations that reach 0.35 µg/l, is a fragrance used to manufacture detergents, deodorants, cosmetics, etc.

VIII – Conclusions

The analysis of monitoring records from the first phase of the injection hydraulic barrier along a year has shown a decrease in electric conductivity in wells in the mixing front of groundwater and injected water.

The treated water quality has been one of the most important issues in order to avoid further aquifer impacts. In the case of the treated wastewater destined to the delta deep aquifer injection, the water employed is of the highest quality. The results obtained show that having undergone additional water treatment, the quality of the recycled water is improved to the point where it complies with the criteria required for optimum water reuse in aquifer recharge regimes. Moreover, the additional treatment impairs possible clogging effects of wells due to physico-chemical and biochemical reactions.

Regarding the presence of caffeine, Paraxantine, Galaxolide and Nicotine, given that these compounds were not found in samples taken from the injection water deposit, it seems probable that their presence is related to natural aquifer recharge.

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References

Abarca E., Vázquez-Suñé E., Carrera J., Capino B., Gámez D. and Battle F., 2006. Optimal design of measures to correct seawater intrusion. *Water Resources Research*, W09415, doi: 10.1029/2005 WR004524.

Cazurra T., 2006. Water Reuse of South Barcelona´s wastewater reclamation plant. *Desalination,* 218 (1-3):43-51.

Iríbar V., Carrera J., Custodio E. and Medina A., 1997. Inverse modeling of seawater intrusion in the Llobregat delta deep aquifer. *Journal of Hydrology*, 198: 226-244.

Levine A. and Asano T., 2004. Recovering sustainable water from wastewater. *Environmental Science Technology*, 201A.

Ortuño F., Niñerola J.M., Teijón G. and Candela L., 2008. Desarrollo de la primera fase de la barrera hidráulica contra la intrusión marina en el acuífero principal del Delta del Llobregat. In IX Simposium of Hydrogeology, Elche, Spain, 28-30 Junary, 2008.

Sedlak D.L., Gray J.L. and Pinkston K.E., 2000. Understanding microcontaminants in recycled water. *Environmental Science Technology*, 34(23): 509-515.

Ternes T., Meisenheimer M., Mcdowell D., Sacher F., Brauch H. and Haist-Gulde B., 2002. Removal of pharmaceuticals during drinking water treatment. *Environmental Science Technology*, 36:3855-63.