

# Biological removal of pharmaceuticals physicochemical characterisation of influen...

[Design](#)



The influx of the effluent was monitored during the period between September 2010 and August 2011. These informations showed that the influx of the effluent ranged between 271000 and 298400 m<sup>3</sup>/day. The physico-chemical parametric quantities of the influent during the sampling period exhibit big fluctuations.

The effluent temperature values varied from 23 & A ; deg ; C at winter and 35 & A ; deg ; C at summer. The pH values varied from 7. 0 to 7. 2 and the values of N, phosphoric, chemical O demand ( COD ) , EC and suspended solids ( SS ) fluctuate during the sample twelvemonth ( see appendix ) .

## **Performance of effluent intervention procedures of Sulaibiya**

The physico-chemical belongingss of the wastewater are presented in appendix.

The entire P content and the entire Kjeldahl N content decreased from 6. 3 mg/L to 1. 6 mg/L and 43. 5 to 2. 5 mg/L, severally.

Phosphorus and N are usually known as modification foods for eutrophication in natural balance of aquatic ecosystems. Therefore, careful manage of their discharge is of import to forestall inordinate algal growing ( Andersen et al. , 2006 ) . The primary and secondary interventions of the effluent efficaciously reduced the P and N by 75 and 94 % , severally. Significantly the wastewater had a better quality in respects to the N and organic contents due to the efficiency of the activated sludge procedure in the WWTP where mean Collect remotions were 93 % . The suspended solids in the secondary wastewater are much lower than the influent by 95 % . During the primary

and secondary interventions, the cations concentrations did not alter significantly which can be seen by the lone little alteration in the electrical conduction between the influent and wastewater with mean decrease of 17 % where other research workers' consequences ( 7 % ) as reported by Tchobanoglous et al.

( 2007 ) .

### **Happening of pharmaceuticals in effluent influents**

The concentrations of the mark pharmaceuticals in the influent over the twelvemonth long sampling period at Sulaibiya are summarized in ( Figures ) . Trimethoprim, sulphamethoxazole, paracetamol and Zantac were found in all influent samples whereas, Flagyls were not detected in October and November. Metronidazole sensing was ranging between 4ng/l at December and 58ng/l at April lower than other reported ( Rosal et al. , 2010 ) .

Trimethoprim and sulphamethoxazole were found in the influent in the scope of 61-1814 and 11-1669, severally. The highest concentration of trimethoprim was found at August and the lowest was found at April whereas the highest concentration of sulphamethoxazole was found at October and the lowest was at February. Trimethoprim was reported at 290 ng/L in natural influent H<sub>2</sub>O in Switzerland ( Goebel et al.

, 2005 ) and at comparatively high concentrations from 2100-7900 ng/L in the US ( Batt et al. , 2007 ) . On the other hand, sulphamethoxazole was reported at high concentration was 6000ng/l ( Batt et al. , 2005 ) and at lower concentration was 70ng/l which was higher than our consequences.

Paracetamol and Zantac was found in the influent at concentrations

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significantly higher than other mark drugs which were the top 10 pharmaceuticals dispensed in Kuwait.

Paracetamol was detected in all of the effluent samples at concentrations running from 101-2086 ng/L with the highest concentrations in November 2010 and lowest concentrations in February 2011. These concentrations were to some extent lower than those reported antecedently ( Pham and Proulx, 1997 ; Ternes 1998 ; Blanchard et al. , 2004 ) . On the other manus, Zantac was running from 365 to 2009 ng/L. These concentrations are consistent with other surveies with concentrations of lower sensing up to 580 ng/L ( Kolpin et al. , 2002 ) and higher sensing at 1700 ng/l ( Gomez et al. , 2006 ) . Harmonizing to the fluctuated temperature during the sampling between summer and winter. This fact might bespeak that the concentrations of pharmaceuticals in the influent may be related to higher ingestion during the cold periods of the twelvemonth when more illness occurs.

## **Figure 5-9: Variation of concentration of assorted mark compounds ( ng/L ) in the**

**influent- each circle in line represents one sample**

**Figure: Concentrations of pharmaceuticals ( ng/L ) in the influent and the remotion per centum by the secondary intervention procedures of Sulaibiya.**

### **Removal of pharmaceuticals during the primary and secondary intervention in Sulaibiya**

The remotion rates of pharmaceuticals during the sampling period were shown in Figure. Paracetamol was removed expeditiously by the secondary intervention, at an mean 97.5 % with highest remotion reached 99.9 % and lowest remotion was 86.1 % . Trimethoprim was removed lower than paracetamol with mean removal 86.

1 % where the highest remotion was 96.1 % and the lowest remotion was 63 % . Removal efficiency of Flagyl in secondary intervention was at mean 83.4 % with highest remotion was 93.9 % and lowest remotion was 59.4 % .

Sulphamethoxazole and Zantac were the lowest remotion efficiency with mean 77.5 % where the highest remotion of sulphamethoxazole was 98.7 % and lowest remotion was 31.3 % while the highest remotion of Zantac was 99.2 % and lowest remotion was 47.4 % . In general, the remotion efficiencies found in this survey were consistent with other WWTP utilizing primary and secondary intervention with activated sludge. For illustration, 75 % remotion rate in German ( Ternes, 1998 ; Stumpf et al.

, 1999 ) , up to 90 % in Spain ( Santos et al. , 2007 ) and up to 99 % in Japan ( Nakada et al. , 2006 ) were reported.

These removal rates for a individual compound can change greatly from one WWTP to another depending on the type of intervention ( e. g. biological and physico-chemical ) and the abode clip of H<sub>2</sub>O in the primary deposit armored combat vehicle ( Santos et al.

, 2007 ) . Removal efficiency of Flagyl has been reported with a big variableness scope from 65-80 % in Spain ( Gros et al. , 2010 ) . On the other manus trimethoprim was reported uncomplete remotion during conventional intervention by several surveies ( Gobel et al.

, 2007 ; Jelic et al. , 2011 ) while Gros et al., 2010 study 65 to 80 % removal efficiency in the workss with higher hydraulic keeping times. Similar observation for the removal efficiency of sulphamethoxazole and Zantac were found by other research worker where they report removal efficiency 30-92 % and 50-98 % , severally ( Gros et al. , 2010 ) . In Germany paracetamol was found to be removed expeditiously at 95 % due to its biodegradability and was detected in less than 10 % of all sewerage wastewaters ( Ternes, 1998 ; Kolpin et al. , 2004 ; Roberts and Thomas, 2006 ) .

Concentrations of the mark pharmaceuticals detected in the WWTP wastewater in a scope of 1-1000ng/L are presented in Figure. This is in understanding with Ternes et Al ( 1998 ) , who reported that many pharmaceuticals were detected in the wastewaters and measured at high

concentrations due to incomplete removal in German sewerage intervention works. The efficiency of modern effluent interventions has increased the removal of pharmaceuticals from influent with the debut of the activated sludge procedure.

Elimination of pharmaceuticals in the activated sludge procedure occurs due to several grounds surface assimilation, biological or chemical debasement and biotransformation. Ternes ( 1998 ) suggested that activated sludge removes high sums of pharmaceuticals than other intervention, most likely to the bacterial activity in the activated sludge. The consequences of this survey showed there was non complete riddance of hint pharmaceuticals in the wastewater. Therefore implementing other engineerings such as membrane systems would be necessary for complete removal of these hints.

### **Consequence of temperature on the removal efficiency**

Although the entire concentrations of mark compounds in the influent samples through-out the annual sampling fluctuated, the removal procedure in the effluent intervention works worked as expeditiously during the summer months as during the winter months. Therefore, consequence of temperature was statically analysed utilizing ANOVA. The correlativity between the temperature and the removal of COD, BOD, organic N, TKN, MLVSS, and mark pharmaceuticals was extremely important ( table ) .

This decision contradicts other research workers who found that the removal processes in effluent intervention works was higher in summer

than in winter ( Vieno et al. , 2005 ) . They suggested that the ground was the lower biodegradation in the works because of low temperature in winter.

## **Variables**

### **Factor**

### **p-value**

### **Confidence degree**

### **Degree of Significance**

COD remotionTemp0100Highly SignificantBOD remotionTemp0100Highly SignificantOrganic N remotionTemp0100Highly SignificantTKN remotionTemp0100Highly SignificantMLVSSTemp0100Highly SignificantMetronidazole remotionTemp0100Highly SignificantTrimethoprim remotionTemp0100Highly SignificantSulphamethoxazole remotionTemp0100Highly SignificantParacetamol remotionTemp0100Highly SignificantRanitidine remotionTemp0100Highly Significant

## **Consequence of pharmaceuticals concentration on the remotion efficiency**

The consequence of pharmaceuticals concentration was extremely important on most responses except sulphamethoxazole which was important consequence on COD, BOD, organic N, and TKN remotion, while was extremely important on MLVSS and sulphamethoxazole remotion efficiency. This extremely important consequence of most mark pharmaceuticals on the remotion COD, BOD, organic N, TKN, drug, and MLVSS agrees with the old qualitative analysis discussed earlier ( Sections ) , since the remotion efficiency and biomass concentrations and features were chiefly affected by



the dominant factor which was the pharmaceuticals concentration. This extremely important consequence on removal efficiencies need to be carefully addressed.

## **Variables**

### **Factor Concentration**

### **p-value**

### **Confidence degree**

### **Degree of Significance**

COD removal Metronidazole 0.100 Highly Significant Trimethoprim 0.

0.0299. 8 Highly Significant Sulphamethoxazole 0.

0.0694 Significant Paracetamol 0. 0.00199. 9 Highly

Significant Ranitidine 0.100 Highly Significant BOD

removal Metronidazole 0.100 Highly Significant Trimethoprim 0.

0.00399. 7 Highly Significant Sulphamethoxazole 0. 0.06493.

0.06 Significant Paracetamol 0. 0.00199. 9 Highly Significant Ranitidine 0.100 Highly

Significant Organic N removal Metronidazole 0.100 Highly

Significant Trimethoprim 0. 0.00299. 8 Highly Significant Sulphamethoxazole 0.

0.05394. 7 Significant Paracetamol 0.

0.00199. 9 Highly Significant Ranitidine 0.100 Highly Significant TKN

removal Metronidazole 0.100 Highly Significant Trimethoprim 0. 0.00399. 7 Highly

Significant Sulphamethoxazole 0.

06393. 7SignificantParacetamol0. 00199. 9Highly

SignificantRanitidine0100Highly SignificantMLVSSMetronidazole0100Highly

SignificantTrimethoprim0100Highly

SignificantSulphamethoxazole0100Highly SignificantParacetamol0100Highly

SignificantRanitidine0100Highly SignificantDrug

remotionMetronidazole0100Highly SignificantTrimethoprim0.

00299. 8Highly SignificantSulphamethoxazole0. 0595Highly

SignificantParacetamol0. 00199. 9Highly SignificantRanitidine0100Highly

SignificantThe primary and secondary effluent intervention gave moderate to high remotion efficiencies of pharmaceuticals.

However, the wastewater still had considerable concentrations of some of these drugs. These concentrations were in the scope of 1-1000 ng/L, as most of the pharmaceuticals present in the influent were found in the wastewater, which indicate the demand for farther interventions to take these pollutants compounds. In this subdivision, an probe was made to compare the degree of pharmaceuticals concentration between the influent and wastewater of a effluent intervention works every bit good as the finding of removal efficiency by the effluent intervention processes. In the following subdivision, the mass balance of rearward osmosis and the remotion of micropollutants by progress engineerings of H<sub>2</sub>O recycling procedures of LPWRP will be described.

## Physical remotion of pharmaceuticals during the WWTP

### Physicochemical Features

The physicochemical features of the provender and permeate for the extremist filtration procedure during the sampling twelvemonth are presented in Appendix. The mean value of the physicochemical features of the recess of extremist filtration was pH ( 7.

04 ) , TSS ( 8. 68mg/l ) , TDS ( 437. 1mg/l ) , COD ( 24mg/l ) , BOD ( 4.

07mg/l ) , entire Fe ( 1. 38mg/l ) , and entire coliforms ( 426261. 2

colonies/100ml ) . The mean removal efficiencies of the extremist filtration procedure were 98 % , 95 % and 99 % for the TSS, entire Fe and entire coliforms measurings, while there were no important alterations in the TDS measurings. The concentrations of COD were measured for extremist filtration provender and permeate. Consequences did non demo high remotion of hint organic contaminations through the filtration processes where the mean remotion of COD was 42 % while the BOD was 72 % .

Therefore, the extremist filtration procedure provides an indispensable pre-treatment for the RO by taking particulate and colloidal stuff from the provender but the remotion is limited to atoms larger than the membrane pore size ( Van der Bruggen et al. , 2003a ) . The mean removal efficiencies of the RO procedure for the TSS and entire coliforms measurings were 68 % and 99 % , while there were extremely important remotion in the TDS measurings with mean removal 96 % . Furthermore, the concentrations measurings of BOD for RO provender and pervade show high remotion of hint organic contaminations through the filtration processes with mean

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removal 90 % . Trace Organic Compounds may be wholly or partly degraded in effluent intervention a works that takes topographic point largely in the activated biological sludge procedure. Pharmaceuticals fluctuate in their debasement in assorted effluent intervention processes. The staying pharmaceuticals are passed into the extremist filtration so to RO systems. Pharmaceutical compounds were detected in the RO provender that was derived from the WWTP.

These fluctuations were once a consequence of one-year fluctuations of compounds in natural effluent in add-on to other procedures involved in effluent intervention. Most of pharmaceuticals, viz. metronidazole, Trimethoprim, sulphamethoxazole, paracetamol, and Zantac were found in all sample from RO recesss during the sampling twelvemonth. The mean concentrations of these compounds found in the RO recesss were 4ng/l, 61ng/l, 47ng/l, 8ng/l, and 210ng/l for Flagyl, trimethoprim, sulphamethoxazole, paracetamol, ranitidine severally ( Figures ) . The highest removal efficiency of these compounds was 97 % for Zantac so 92 % for sulphamethoxazole and paracetamol as for Trimethoprim was 86 % . Last the lowest remotion efficiency between these compounds was 56 % for Flagyl due to low concentration found in RO recesss. The solubility of these pharmaceuticals varies ; some are reasonably soluble such as sulphamethoxazole and Trimethoprim where the solubility was 281mg/l, 400 mg/L, severally ; some are extremely soluble like Zantac, paracetamol and Flagyl where the solubility were 24. 7g/l, 14g/l and 10g/l, severally.

Log Kow values of these pharmaceuticals ranged between -0.02 and 0.92. The scatter plot of log Kow versus removal efficiency showed a weak positive (Figure).

On the other hand, the solubility and log Kow did not correlate with the behavior of these pharmaceuticals in the RO. Harmonizing to Tolls (2001) found that log Kow may not be a good index of the behavior of pharmaceuticals in the environment. It has been reported that the removal efficiency of solute by membrane filtration and RO is affected by a different parametric quantities such as pH, solute charge, molecular weight and geometry, membrane charge and hydrophobicity, every bit good as the membrane surface charge (Van der Bruggen et al., 1998; Van der Bruggen et al.

, 1999; Kiso et al., 2000; Kiso et al., 2001; Ozaki and Li, 2002; Kimura et al., 2003b; Kimura et al., 2004).

Probes have been done by old surveys on the removal efficiency of RO compared to other types of membranes and low force per unit area contrary osmosis where they found a great advantage of utilizing RO in bringing forth a high quality of recycled H<sub>2</sub>O. Harmonizing to Lopez-Ramirez et al., (2006) who found that the recycled effluent for were widely exceed the RO membrane the drinking H<sub>2</sub>O criteria by. However, the removal consequences of the RO membrane represent extremely reduced pollutants in permeate. Furthermore, microorganisms were removed from the RO permeate, which would let safe reuse of H<sub>2</sub>O.

## **Consequence of temperature and pH on the remotion efficiency**

Similarly to biological intervention the temperature was impacting the remotion processes in the RO system. The concentrations of mark pharmaceuticals in the RO recesss samples during the trying twelvemonth was fluctuated during the summer and winter months. Therefore, consequence of temperature and pH was besides statically analysed utilizing ANOVA.

The correlativity between the temperature and pH with the remotion of BOD, TSS, TDS, entire coliforms and mark pharmaceuticals was extremely important ( table ) . In this survey, there was no relationship between the remotion of pharmaceuticals and the other remotion parametric quantity tested such TSS, TDS, BOD, and entire coliforms distributed in the RO watercourse. The arrested development analysis shows no correlativity between the removal efficiency of pharmaceuticals and TSS, TDS, BOD, and entire coliforms.

This might be due to the complexness of RO provender in intervention workss and to a wide scope of rejection. Therefore, it ' s really hard to tie in these operating parametric quantities with these remotion rates. Although to the broad scope of variableness and restriction of informations, there was no possible to find the relationship between the remotion and molecular weight or molecular size.

Harmonizing to Kimura et al. , ( 2003b ) there was a additive relationship between molecular weight of the non-charged compounds and remotion.

However, in this survey, the relationship between the molecular weight and the removal of Flagyl, trimethoprim and Zantac was observed a additive arrested development while sulphamethoxazole and was non on the arrested development line. The physico-chemical features of tried drugs in this survey differ from each other. Therefore, a relationship between any of these removals could be described by different physico-chemical features such the charge, form of compounds. Positive correlativity between hydrophobicity of non-phenolic compounds (  $\log K_{ow}$  ) and their rejection by nanofiltration was reported by Kiso et al. , 2000.

The rejection of nonylphenol and bisphenol A varied greatly particularly for NP. Rejection varied between 0 to 100 % for nonylphenol and 53 % to 100 % for bisphenol A ( see Figure 6-20 ) . Rejection mechanisms by RO were investigated by many research workers ( Ozaki and Li, 2002 ; Van der Bruggen et al. , 2003b ) . Rejections may be influenced by dipole moment of compounds, hydrophobicity of compounds represented by  $K_{ow}$  and molecular size. As reported by Ozaki and Li ( 2002 ) , it is hard and complicated to clarify the nucleus mechanisms for the rejection of hint organic compounds by RO under existent conditions. However, most surveies on rejection mechanisms by RO were conducted on pilot graduated tables utilizing virgin membranes, high concentrations and utilizing either base or man-made H<sub>2</sub>O. These pilot graduated tables were run under ideal conditions.

Snyder and colleagues ( 2007 ) suggested that the compounds which breached RO under the full graduated table were non consistent, and no

clear relationship between molecular construction and membrane could be established. Breaching of the RO could be the consequence of diffusion into and through the membrane, short-circuiting of the membrane or back upping media failure.

## 6. 8 Product H2O

The concentrations of PhACs and EDCs were reduced as these compounds passed through the double membrane systems in the H2O recycling works. Clofibric acid, diclofenac and diphenylhydantoin were non detected in any merchandise H2O samples. Other compounds ( nonylphenol, Lopid, isobutylphenyl propionic acid, Orudis, naproxen, Datriil, Mysoline, salicylic acid and carbamazepine ) were detected in most merchandise H2O samples with a maximal concentration of 120 ng/L ( Table 6-6 ) . Bisphenol A ( BPA ) was detected in all merchandise H2O samples with concentrations ranging from 20-464 ng/L.

These concentrations are non surprising since BPA had the 2nd highest concentration among the compounds found in the influent. The presence of BPA in the H2O merchandise would be considered to hold a possible impact on the environment and the concluding user tract. Recent surveies ( Nakada et al. , 2006 ; Roberts and Thomas, 2006 ; Gomez et al. , 2007 ; Santos et al. , 2007 ) have shown that degrees of compounds found in the wastewater of WWTP are frequently higher by more than one order of magnitude than the consequences obtained in this survey.

Therefore, it is most likely that the remotion of PhACs and other compounds is more effectual in the advanced intervention works utilizing RO systems



than other conventional intervention works ( Snyder et al. , 2007 ) .

Recently, Snyder et Al. ( 2007 ) have investigated the removal of a wide scope of EDCs during imbibing and effluent intervention processes at bench, pilot and full graduated table. They found that RO membranes removed about wholly investigated compounds to degrees less than method describing bounds ( MDL ) ( 1-10 ng/L ) . However, hint degrees of some contaminations were still noticeable ( & A ; It ; MDL ) in RO permeate ( e. g. , gemfibrozil and naproxen ) .

It should be noted that the latter survey was a snapshot and it does non give such a comprehensive image as the work presented here.

## **Chlorination consequence on the pharmaceuticals during the intervention**

The wastewaters of the RO were treated farther by CI before discharge. Most of pharmaceuticals escape from the RO system and the hint were detected in the RO wastewater.

The maximal concentration detected was 19ng/l and 15ng/l for Trimethoprim and ranitidine where the other pharmaceuticals were detected at 7ng/l, 5ng/l, and 4ng/l severally. On the other manus, the lowest concentration detected in the RO wastewaters for Flagyl, trimethoprim, sulphamethoxazole, paracetamol, and Zantac were 0. 2ng/l, 1ng/l, 0. 2ng/l, 1ng/l, and 1ng/l severally. The happening of pharmaceuticals in the environment, chiefly in H<sub>2</sub>O, is a subject that has attracted a strong attending since the last 10 old ages. In this clip, several curative categories of drugs and their homo and animate being metabolites have been found in

the aqueous environment at degrees making several mg/L in effluent ( Petrovic and Barcelo, 2007 ; Reemtsma and Jekel, 2006 ) . The uncomplete remotion of these pollutants at effluent intervention workss ( WWTPs ) , non designed for this undertaking, has permitted their spread through surface Watersss ( Boyd et al. , 2003 ; Carballa et al.

, 2004 ; Kim et al. , 2007 ; Metcalfe et al. , 2003 ; Okuda et al. , 2008 ; Pa? xeus, 2004 ; Reemtsma et al.

, 2006 ; Tauxe-Wuersch et al. , 2005 ; Ternes, 1998 ) , which is really a primary beginning of imbibing H<sub>2</sub>O. Subsequently, some pharmaceuticals are once more non wholly removed during imbibing H<sub>2</sub>O production and therefore, they have been identified in potable H<sub>2</sub>O at the ng/L degree ( Drewes et al. , 2002 ; Kim et al. , 2007 ; Mompelat et al. , 2009 ; Stackelberg et al.

, 2004, 2007 ; Ternes et al. , 2002 ) . Paracetamol is easy degraded by deacetylation to para-aminophenolP-aminophenol is an parallel and metabolite of common family anodynes, such as Datril.

It is well-known that Datril in overdose can do terrible hepatic centrilobular mortification in worlds and experimental animate beings ( Thomas, 1993 and Boyd and Berezky, 1966 ) . Like Datril, p-aminophenol-induced hepatotoxicity may besides affect a chemically reactive intermediate and GSH may play an of import function in its toxicity. This possibility has non yet been explored ( Klos et al. , 1992 ) .

P-aminophenol is a nephrotoxic metabolite of Datriil. It is 5 times more powerful than acetaminophen as nephrotoxicant in F344 rats. Inhibition of acetaminophen deacetylation to p-aminophenol lessened nephritic toxicity, proposing that acetaminophen nephritic toxicity is partially mediated by formation of p-aminophenol.

P-aminophenol nephrotoxicity is site-specific for the S3 section of the proximal tubule ( Harmon, 2006 ) . Molecular weight 171. 2aLog Kow-0. 02 at 25 & A ; deg ; CaWater solubility 10 g/L at 25 & A ; deg ; Cb

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log Kow = 0. 91290. 32Solubility in H2O: 400 mg/l @ 25 deg.

C. Log Kow sulphamethoxazoleMolecular Weight ( g/mol ) 253. 28Water Solubility ( mg/L ) 281Log Kow 0. 92Molecular Weight151.

16CorrosivityDisassociationpKa = 9. 38log P ( octanol-water )log Kow = 0. 46SolubilitiesVery somewhat sol in cold H2O, well more sol in hot H2O ; colloidal suspension in methyl alcohol, ethyl alcohol, dimethylformamide, ethylene bichloride, propanone, ethyl ethanoate ; somewhat sol in quintessence ; practically insol in crude oil quintessence, pentane, benzine. In H2O, 14 g/L at 25 deg CKow 0. 27Molecular weight: 350. 9Water Solubility24. 7 g/L