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Treatability studies of hospital wastewaters with AOPs by Taguchi's experimental design

Yonar T.[1] and Kurt A.[2,*]

[1]Uludag University, Faculty of Engineering, Environmental Engineering, Gorukle Campus, Bursa 16059, Turkey

[2]Central Research Laboratory for the Scientific and Technological Supports, Duzce Universitesi, Konuralp Campus, Duzce 81620, Duzce, Turkey

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*to whom all correspondence should be addressed: e-mail: kurtayse1987@gmail.com

Abstract

In this study, it was studied various advanced oxidation treatment processes; Fenton, UV/H_2O_2, $UV/O_3/H_2O_2$ for treatability of hospital wastewaters containing antibiotics (cephalosporines). Taguchi's L25 orthogonal array design was applied to design of advanced oxidation processes, for simplification of the analysis and calculations. 95,7%, 90,65%, 91,8% COD and 55,86%, 60,83%, 70,8% TOC removal efficiencies were obtained under the best operation conditions for UV/H_2O_2, $UV/O_3/H_2O_2$ and Fenton processes, respectively. According to the ANOVA results, pH was of great importance in COD removal for Fenton. For the UV/H_2O_2 processes, H_2O_2 has significance in COD and TOC removals. As for the $O_3/UV/H_2O_2$ processes, O_3/UV reaction time was found as an important parameter effecting the removal rates. Also, cephalosporine antibiotic active compounds (cefradine and cefaclor) were degraded completely within minutes for all of the processes. Taguchi's Method was found useful for the environmental applications and simplifications of advanced oxidation processes for treatment of hospital wastewaters and calculations.

Keywords: advanced oxidation, ozone, hydrogen peroxide, Fenton, cephalosporine, orthogonal array

1. Introduction

Pollutants, effecting natural water sources are changing and increasing rapidly day by day, in terms of quantity and composition. Wastewaters are accepted as the main source of these pollutants which affect environment adversely. They may contain heavy metals, radioactive and toxic substances and pharmaceuticals at high level (Gupta *et al.*, 2009; Kajitvichyanukul and Suntronvipart, 2006). However, analysis and treatment technologies of wastewaters are inadequate.

It is well known that, pharmaceutical pollutants have a major impact on the aquatic environment and human health which are found in wastewaters mostly (Daughton and Ruhoy, 2009). They affect human health and aquatic environment adversely and have toxic and endocrine disrupting properties (Walsh, 2003; Sanderson *et al.*, 2004; Kim and Aga, 2007; de Souza *et al.*, 2009; Yu *et al.*, 2009). Among these endocrine disruptor pharmaceuticals antibiotics are accepted as the most dangerous types, because of the microorganism resistance and bioaccumulation in the environment (Petrovic *et al.*, 2005; Blackwell *et al.*, 2004). They are used highly for protection and treatment of human and animal health and improving agricultural processes (Oberlé *et al.*, 2012).

Synthetic antibiotic compounds attract attention with their widespread use in Turkey and worldwide (Balcıoğlu and Arslan, 1997). In Turkey, human and animal applications of antibiotics are much more higher than in northern European countries. According to the IMS (2013-2014) datas, consumptions of cephalosporins, penicillins and quinolones were found very high in Turkey (IMS, 2013 and IMS, 2014). These antibiotic compounds are excreted into the environment through the metabolism of living organisms and agricultural activities. They contaminate aquatic environment by water and wastewater discharges, mostly.

However, hospital wastewaters contain antibiotics at high rate, particularly. They contain pharmaceuticals, heavy metals, radioactive and toxic substances etc. highly (Gupta *et al.*, 2009; Kajitvichyanukul and Suntronvipart, 2006). They are generally treated with centralized domestic wastewater treatment systems without pretreatment processes (Santos *et al.*, 2013; Verlicchi *et al.*, 2010; Thomas *et al.*, 2007). Unfortunately, conventional treatment plants for treatment of these compounds are inadequate in terms of efficiency (Kim and Aga, 2007; Sukul and Spiteller, 2007). Advanced treatment processes such as carbon adsorption, advanced oxidation processes (AOPs), modern pressure-driven membrane processes with reverse osmosis or nanofiltration are recommended for the removal of antibiotics and other pharmaceuticals from wastewaters, instead of conventional treatment methods (Balcioğlu and Arslan, 1997).

Some countries such as China and Japan treat hospital wastewaters on-site by conventional wastewater treatment plants or membrane bioreactors against the pathogen risks (Liu *et al.*, 2010; Pauwels and Verstraete, 2006). In Europe: Marienhospital Gelsenkirchen and Waldbrol (Germany), Isala clinics in Zwolle (The Netherlands), Cantonal Hospital of Baden (Switzerland) (Pills Project, 2012), a hospital in Ioannina (Greece) (Kosma *et al.*, 2010) and Herlev hospital (Denmark) (Nielsen *et al.*, 2013) treat wastewaters seperately for the removal of pharmaceuticals. Advanced oxidation processes (AOPs) are promising alternative methods for treatment of hospital wastewaters because of their efficiency and cheapness (Wols and Hofman-Caris, 2012; Sirés and Brillas, 2012).

For this reason, treatability of hospital wastewaters containing antibiotics were investigated by advanced oxidation processes, in this study. Also, Taguchi's L25 orthogonal array design was applied to design of advanced oxidation processes for simplifications of the analysis. Three different generations (1st, 2nd and 3rd) of cephalosporin antibiotics: cefaclor, cephradine and cefoperazone were chosen to investigate in hospital wastewater.

2. Experimental

2.1. Chemicals and standards

Antibiotic standards used in this study are, cephradine (European Pharmacopoeia Reference Standard, C090000), cefaclor (Fluka, 72579-50MG) and cefoperazone (Sigma-Aldrich, C4292-1G). Methanol (gradient grade for HPLC, Sigma-Aldrich, 34885-2,5L-R) and formic asid (Sigma-Aldrich) were the purity of ≥ 99.9% and 98-100%, respectively, which were used in mobile phase for the HPLC analysis.

2.2. Wastewater characteristics

Composite wastewater samples were taken from a hospital discharge, (Duzce University Research Hospital, Turkey) for one year, monthly.

Table 1. The physical and chemical characteristics of hospital wastewater

Parameter	Unit	Value
pH	-	9,1 ± 0,1
Conductivity	μg cm^{-1}	1464,5 ± 1,2
DO*	mg l^{-1}	2,33 ± 0,02
BOD_5*	mg l^{-1}	155,5 ± 2,3
COD*	mg l^{-1}	290,5 ± 3,1
TOC*	mg l^{-1}	77,105 ± 0,8
DOC*	mg l^{-1}	66,84 ± 0,9
SS*	mg l^{-1}	80 ± 1,7
TKN*	mg l^{-1}	226,7 ± 2,3
Total Phosphorous	mg l^{-1}	4,41 ± 0,3

*COD: Chemical oxygen demand; TOC: Total organic carbon; BOD_5: 5-day biochemical oxygen demand; DOC: Dissolved organic carbon; SS: Suspended solids; TKN: Total kjeldahl nitrogen; DO: Dissolved oxygen

Samples were stored in opaque PE containers at 4 °C to prevent degradation. Cefradine concentration range was found between 0,09-0,28 μg l^{-1} and cefaclor was 0-0,039 mg l^{-1}, while cefoperazone could not be detected. The physical and chemical characteristics of wastewater were given in Table 1.

2.3. Analytical methods

TOC and DOC measurements were made by using TOC-L analyser (Shimadzu) and ASI-L autosampler (Shimadzu). Conductivity, pH and DO measurements were made by using multimeter (Hach-Lange, HQ40D). Analytic determination of antibiotics in hospital wastewater were carried out by HPLC-PDA detector (High performanced liquid chromatography - photo diode array detector) (Shimadzu). Inertsil C-18 column (150 x 4.6 mm; 5 mm) was used as mobile phase column. Mobile phase solution was prepared by water including 0,1 % formic asid and methanol, MeOH:H2O=40:60 (v/v). Analysis were carried out at isocratic mode. Flow rate was chosen as 0,5 ml min-1. PDA detector wavelength range was selected as 254 and 270nm. Column oven temperature was 35°C. 6 ml solid-phase extraction cartridges (SUPELCO, Supelclean ENVI-8), Na2EDTA (Disodium ethylenediaminetetraacetate dihydrate) (Merck) and HCl (EMSURE) were used for solid phase extraction.

MnO_2 powder (Merck) was used to remove residual H_2O_2, by controlling with test strips (Merck). To remove $Fe(OH)_3$ and MnO_2, supernatants were filtered through 0,45μm milipore membranes. BOD_5, COD, SS, total phosphorous, total nitrogen and pH were measured according to the Standard Methods (APHA, 1998). All of the measurements were carried out in triplicate.

2.4. Advanced oxidation processes

Photochemical oxidation system, used for the H_2O_2/UV (hydrogen peroxide/ultraviole), and H_2O_2/O_3/UV (hydrogen peroxide/ozonation/ultraviole) processes was designed and manufactured to be used as both batch and constant. Experiments were carried out in constant mode. Low-pressure TUV-8 model UV lamp was used (Philips) in the reactor. Maximum wavelength of the lamp was 254 nm. It was placed into the reactor (2,8 L) in pure quartz cover (40 cm length and 4 cm diameter). The reactor body was made of 316-Ti stainless steel and inner diameter was 41 cm by 9.8 cm length.

Opal 200 model ozone generator was used for the ozonation processes with ozone production capacity of 200 mg hr^{-1} by airflow. Ozonation experiments were made by addition of wastewater into the glassware (0,5 L). Flow rate of the generator was fixed at 60 L hr^{-1} hourly for all of the experimental works. All connections were made by Teflon tubing.

2.5. Taguchi's Experimental Design

Taguchi's method is a robust statistical tool, capable of finding out significant parameters for ideal processes/experiments from many other parameters (Chiang and Hsieh, 2009). The method include orthogonal arrays and allow studying with minimum number of experiments by using several factors (Sharma *et al.*, 2005). Also, it reduce the variability of the process, and consequently, reduce the operating costs (Barros *et al.*, 1995). In this study, "Taguchi's Method" statistical principles were applied to design and optimization of parameters effecting advanced oxidation processes. Experimental designs were made according to the Taguchi's L25 orthogonal array design. "Minitab 17" statistical analysis program was used for the data input and design. Taguchi's L25 orthogonal design parameters for AOPs were shown in Table 2.

Table 2. Factors and levels of the processes according to Taguchi's L25 orthogonal design

Process	Factor	Level 1	Level 2	Level 3	Level 4	Level 5
Fenton	A: pH	2	3	4	5	6
	B: Fe^{2+} (mg l^{-1})	25	50	75	100	125
	C: H_2O_2 (mg l^{-1})	25	50	75	100	125
UV/H_2O_2	E: UV (min)	30	60	90	120	150
	C: H_2O_2 (mg l^{-1}l)	25	50	75	100	125
$UV/O_3/H_2O_2$	F: UV/O_3 (min)	30	60	90	120	150
	C: H_2O_2 (mg l^{-1})	25	50	75	100	125

3. Results and discussion

Cephalosporine antibiotic active compounds (cefradine and cefaclor) were degraded completely within minutes for all of the advanced oxidation processes. COD and TOC removals were interpretated by Main Effects Plots for the AOPs.

Fenton

Main Effects Plots for COD and TOC were given in figure 1. and figure 2. for Fenton. Depending on the increase in pH, COD removals decreased first and then increased suddenly, later decreased again, and finally rose. It was resulted from the changes in COD removal efficiencies that fenton reactions are very sensitive to changes in pH. Depending on the changes in H_2O_2 concentration, COD removal efficiencies were decreased with the H_2O_2 addition. When the concentration reached to 100 mg l^{-1}, COD removal rates rose significantly and then dropped again. Changes in Fe^{+2} concentration did not effect the COD removals significantly, while a slight increase was observed in removals at 50 ppm.

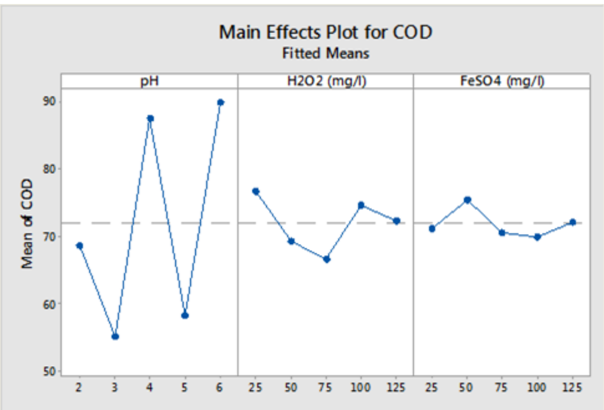

Figure 1. Main Effects Plot for COD for the Fenton

TOC removal rates showed a sudden increase when pH rose to 5 and then decreased again. TOC removals were effected positively from the increase in peroxide concentration. The removal efficiencies remained constant for 50-75 mg l^{-1}, then dropped again, and when H_2O_2 concentration reached to 125 mg l^{-1}, efficiency rose suddenly. TOC removals were effected positively from increase in Fe^{+2} concentration but it was observed a sudden decrease at 125 ppm.

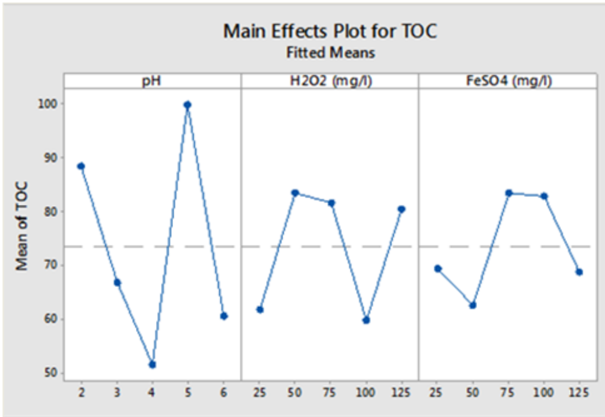

Figure 2. Main Effects Plot for TOC for the Fenton

The changes in COD and TOC removal rates were affected adversely from each other in Fenton reactions. TOC removal rates decreased with COD removal increase, while increased with COD removal decrease.

For the Fenton processes, 91,8% COD and 70,8% TOC removal rates were observed as the best results at the conditions of pH 4 with 25 mg l^{-1} H_2O_2 and 75 mg l^{-1} $FeSO_4$. 63,16% COD and 75% TOC degradation were observed at pH 6 with 25 mg l^{-1} H_2O_2 and 125 mg l^{-1} $FeSO_4$. Furthermore, 91,7% COD removal was obtained at pH 6 with 125 mg l^{-1} H_2O_2 and 100 mg l^{-1} $FeSO_4$, and 91,16% COD degradation was obtained at pH 6 with 25 mg l^{-1} H_2O_2 and 125 mg l^{-1} $FeSO_4$. Affam and Chaudhuri (2013) investigated fenton oxidation of amoxicillin and cloxacillin aqueous solution (H_2O_2/COD: 2, H_2O_2/Fe^{2+}: 76, reaction time: 90 min and pH 3) and they found removal efficiencies % 78,98 and 72,96 for COD and TOC, respectively.

UV/H_2O_2

Main Effects Plots for COD and TOC were given in figure 3. and figure 4. for the UV/H_2O_2 processes, respectively. Depending on the increase in UV irradiation time, the

COD removal efficiencies decreased first, eventually reached to the optimum after 90 min and then dropped again. Depending on the changes in H_2O_2 concentration, the removal efficiencies of COD and TOC were constant at 30 and 60 min, while reached to optimum after 90 min of irradiation, then started to decrease. TOC was reduced by additon of H_2O_2 first, then increased when it reached to 100 mg l^{-1}, finally dropped again.

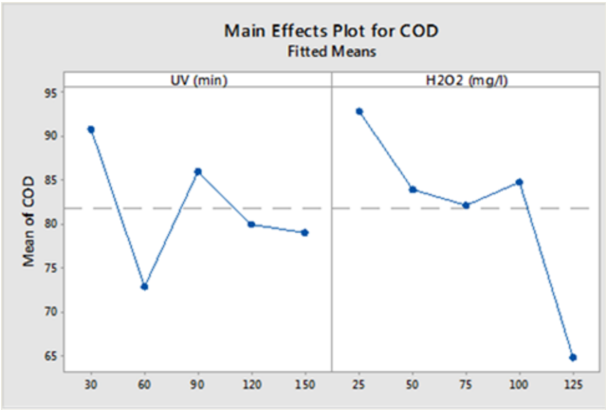

Figure 3. Main Effects Plot for COD for the UV/H_2O_2

At UV/H_2O_2 processes 91,8% COD and 91% TOC removals were obtained as the best results after 30 min UV reaction with 125 mg l^{-1} H_2O_2. 91,3% COD and 84,7% TOC removals were observed after 30 min with 75 mg l^{-1} H_2O_2 and 90% COD and 84,7% TOC removals were obtained after 30 min with 100 mg l^{-1} H_2O_2. Jung *et al.,* (2012) investigated oxidation of amoxicillin by UV/H_2O_2 and they found low mineralization degree and observed maximum 50% TOC removal after 80 min.

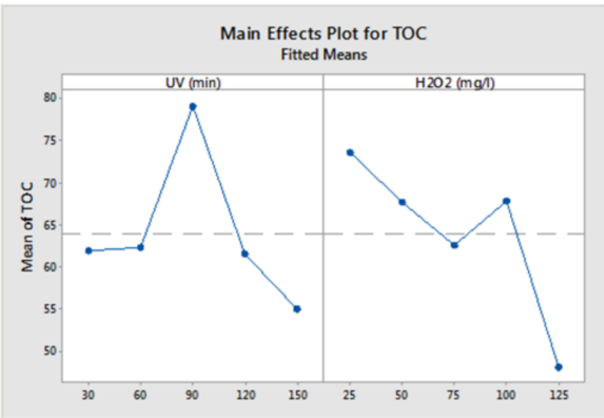

Figure 4. Main Effects Plot for TOC for the UV/H_2O_2

UV/H_2O_2/O_3

Main Effects Plots for COD and TOC were given in figure 5 and figure 6 for the UV/H_2O_2/O_3 processes respectively. The removal efficiencies of COD and TOC decreased by the addition of H_2O_2. When the concentration reached to 100 mg l^{-1}, efficiencies rose significantly, then fell again and finally remained stable. Depending on the increase in UV/O_3 reaction time, the COD and TOC removal efficiencies decreased first, eventually reached to the optimum after 90 min and then dropped again.

For the UV/O_3/H_2O_2 processes, 89% COD and 64,5% TOC removal rates were observed after 90 min reaction with 25 mg l^{-1} H_2O_2 as the best results. At the same processes, 90,65% COD and 60,83% TOC removal rates were observed after 120 min with 75 mg l^{-1} H_2O_2, while 87,6% COD and 65,6% TOC removals were obtained after 60 min with 25 mg l^{-1} H_2O_2.

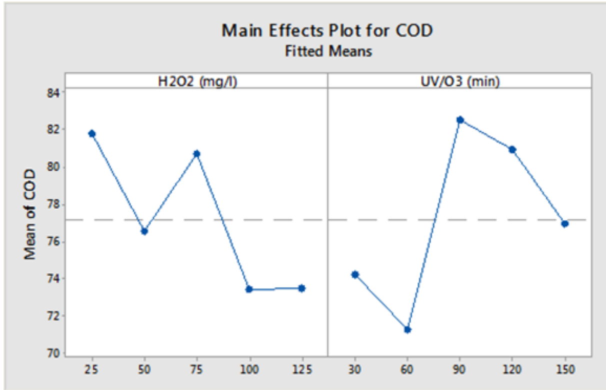

Figure 5. Main Effects Plot for COD for the UV/H_2O_2/O_3

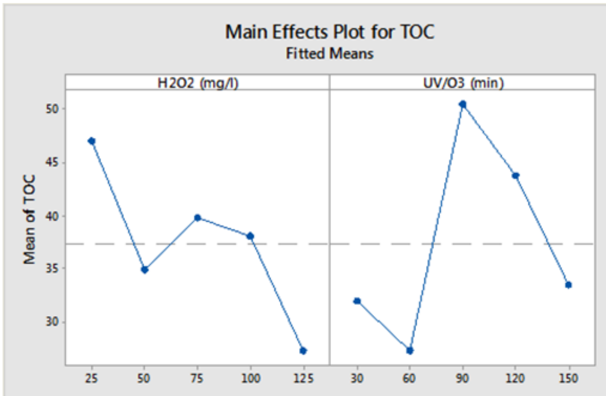

Figure 6. Main Effects Plot for TOC for the UV/H_2O_2/O_3

Analysis of variance (ANOVA)

Analysis of variance (ANOVA) was applied, for evaluating significance of the parameters (Rosa *et al.*, 2009). ANOVA analysis was shown in Table 3 for treatability of hospital wastewater by the advanced oxidation processes. According to the results, variance with 95% trust, and p-value lower than 5%; pH (F= 15.12, P-value= 0% for COD and F= 1.77, P-value= 0.186% for TOC) has significance in COD and TOC removals for Fenton processes. H_2O_2 (F= 1.165, P-value=0.365%) was found important for TOC removals for the same processes. For the UV/H_2O_2 processes, H_2O_2 (F= 1.722, P-value=0.185% for COD and F= 1.433, P-value=0.186% for TOC) has significance in COD and TOC removals and UV (F= 0.901, P-value=0.482%) was found important for TOC removals for the same process. As for the O_3/UV/H_2O_2 processes, O_3/UV reaction time (F= 1.490, P-value=0.243% for COD and F=1.137, P-value=0.368% for TOC) was found as an important parameter effecting the removal rates.

4. Conclusions

In this study, it was studied advanced oxidation treatment processes for treatability of hospital wastewaters containing antibiotics. Taguchi's L25 orthogonal array design was applied to design of processes. The main results were given below:

- The changes in COD and TOC removal rates were affected adversely from each other in Fenton reactions. TOC removal rates decreased with COD removal increase, while TOC removals improved with COD removal decrease. At UV/H_2O_2 processes depending on the changes in H_2O_2 concentration, the removal efficiencies of COD and TOC removal rates were constant at 30 and 60 min, while removals reached to optimum after 90 min of irradiation and then started to decrease. As for the O_3/UV/H_2O_2 porcesses, the removal efficiencies of COD and TOC decreased by the addition of H_2O_2 up to 100 mg l^{-1}. Depending on the increase in UV/O_3 reaction time, COD and TOC removal rates decreased first, eventually reached to the optimum after 90 min and then dropped again.
- For the Fenton processes, 91,8% COD and 70,8% TOC removal rates were observed as the best results at pH 4, with 25 mg l^{-1} H_2O_2 and 75 mg l^{-1} $FeSO_4$. At UV/H_2O_2 processes, 91,8% COD and 91% TOC removals were obtained as the best results after 30 min UV degradation with 125 mg l^{-1} H_2O_2. As for the UV/O_3/H_2O_2 processes, 89% COD and 64,5% TOC removal rates were observed after 90 min reaction with 25 mg l^{-1} H_2O_2 as the best results.
- Cephalosporine antibiotic active compounds (cefradine and cefaclor) were degraded completely within minutes for all of the processes.
- According to the ANOVA results, pH was of great importance in COD removal for the Fenton processes. For the UV/H_2O_2 processes, H_2O_2 has significance in COD and TOC removals. As for the O_3/UV/H_2O_2 processes, O_3/UV reaction time was found as an important parameter in terms of efficiency
- Consequently, Taguchi's Method was found useful for environmental applications and simplifications of advanced oxidation processes for treatment of hospital wastewaters and calculations.

Table 3. Analysis of variance for hospital wastewater degradation

	Factors	Sum of Squares		DF*		Mean Sum		Fisher Test		P-Value*	
		COD	TOC	COD	TOC	COD	TOC	COD	TOC	COD	TOC
Fenton	H_2O_2	718.134	3.767.18	4	4	179.533	941.795	0.392	1.165	0.811	0.365
	$FeSO_4$	112.244	2.166.36	4	4	28.061	541.590	0.056	0.592	0.993	0.674
	pH	6.078.09	5.111.13	4	4	1.519.52	1.277.78	15.120	1.777	0.000	0.186
	Error	2686,7	7718	12	12	223,89	643,2				
	Total	29235,2	20302	24	24						
UV/H_2O_2	H_2O_2	2.128.38	1.863.78	4	4	532.09	465.945	1.722	1.433	0.185	0.260
	UV	1.110.10	1.277.12	4	4	277.52	319.280	0.771	0.901	0.557	0.482
	Error	4435,1	4741	16	16	277,2	296,3				
	Total	8307,1	8367	24	24						
O_3/UV/H_2O_2	H_2O_2	312.126	1.036.16	4	4	78.031	259.041	0.987	0.600	0.437	0.667
	O_3/UV	434.738	1.791.10	4	4	108.68	447.774	1.490	1.137	0.243	0.368
	Error	1146,4	6842	16	16	71,65	427,6				
	Total	1893,2	9669	24	24						

*DF: Degrees of freedom; P: Probability

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