Trattamenti innovativi per il riuso degli effluenti dei depuratori

Relatore: ing. Danilo Spasiano, PhD

Interreg Greece-Italy
European Regional Development Fund
EUROPEAN UNION
Re- Water

de' remi facemmo ali
1990
POLITECNICO DI BARI
Politecnico di Bari

Ingegneria Civile Ambiente Territorio Edile Chimica
DICATECh

Contaminanti emergenti

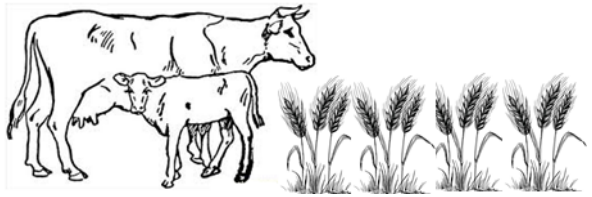
1) Sono spesso recalcitranti, ovvero molto **resistenti ai processi decomposizione che normalmente avvengono in natura**.

2) Per le loro caratteristiche di persistenza e, in molti casi, di tossicità sono composti particolarmente **nocivi per la salute umana e per l'ambiente**.

3) Il pericolo consiste nella **crescente concentrazione negli ecosistemi** terrestri e acquatici.

Qual è l'origine di tali inquinanti?

Allevamento/agricoltura

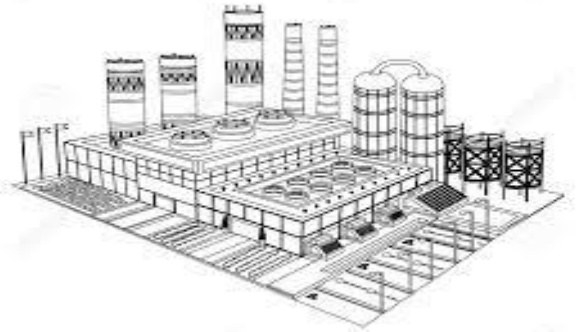


- Farmaci (antibiotici, antiinfiammatori);
- Metaboliti dei farmaci;
- Pesticidi;
- Erbicidi…

Agglomerati urbani

- Farmaci (antibiotici, antiinfiammatori);
- Metaboliti dei farmaci;
- Prodotti per la cura personale
- Prodotti per la pulizia della casa…

Industrie



- Solventi;
- Ritardanti di fiamma;
- Diossine, IPA, PCB…

Contaminanti emergenti

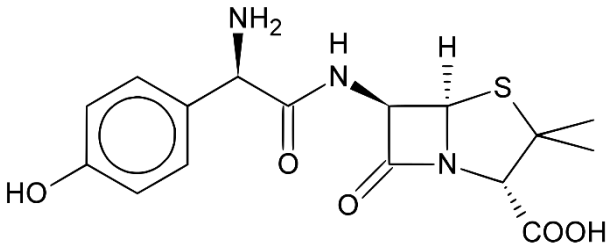
Concentrazione di inquinanti organici recalcitranti nei liquami (ng/L) e negli effluenti (ng/L) degli impianti di depurazione.

Selected ECs	Asia			North America			Europe		
	Influent	Effluent	Reference	Influent	Effluent	Reference	Influent	Effluent	Reference
Antibiotics									
Amoxicillin	<MQL–6516	<MQL–1670	[1–4]	n.r	<MQL	[5]	<MQL	<MQL–190	[6–8]
Azithromycin	1537–303,500	60.1–980	[1, 9]	61–2500	57–1300	[10]	77–1139	38–784	[11–13]
Ceftazidime	<MQL	<MQL	[1]	–	–	–	–	–	–
Chloramphenicol	<MQL–2430	<MQL–1050	[1, 4, 14, 15]	–	–	–	<MQL–319	<MQL	[16, 17]
Chlortetracycline	2333–15,911	<MQL–1986	[1, 4]	<MQL–310	<MQL–420	[10, 18]	n.r	<MQL	[8]
Ciprofloxacin	15.5–6453	<MQL–524.1	[1, 9, 19, 20]	<MQL–246,100	<MQL–620	[10, 18, 21, 22]	<MQL–13,625	<MQL–5692	[6, 11, 13, 23]
Clarithromycin	26–1854	4.79–637.1	[1, 20]	<MQL–8000	130–7000	[10, 22]	0.4–647	25–359	[11–13]
Clindamycin	23.8–26.6	2.94–4.24	[1]	–	–	–	<MQL–101	10–180	[16, 24]
Enrofloxacin	<MQL	<MQL	[25]	5.9–250	3.5–270	[10, 18]	<MQL–18	<MQL–636	[8, 26]
Erythromycin	111.4–403.3	70–186.6	[1]	–	–	–	<MQL–2130	<MQL–290	[6, 13, 23]
Erythromycin-H_2O	226–20,600	194.5–14,400	[1, 4, 20, 27]	<MQL–3900	<MQL–838	[10, 18, 22]	24–6755	15–2841	[11, 12, 17]
Lincomycin	<MQL–19,401	3.92–21,278	[1, 20, 28, 29]	<MQL–360	4.9–510	[10, 18, 21, 30]	<MQL–281	<MQL	[6]
Meropenem	264.8–433.6	27–67.9	[1]	–	–	–	–	–	–
Minocycline	730.9–3808	<MQL	[1]	<MQL	<MQL	[10]	–	–	–
Ofloxacin	54.8–1274	13.3–7870	[4, 20, 25, 31]	470–1000	<MQL–506	[21, 22]	n.r	71–8637	[8]
Oxytetracycline	<MQL–30,049	<MQL–2014	[1, 4, 20, 25]	<MQL–47,000	<MQL–4200	[10, 18, 21]	<MQL–7	<MQL–5	[32]
Sulfamethazine	<MQL–1814	<MQL–260.8	[1, 4, 20, 29]	<MQL–300	<MQL–363	[10, 18, 22]	<MQL–680	<MQL	[13, 16, 32]
Sulfamethoxazole	3.0–1389	<MQL–562	[1, 4, 20, 29, 33]	<MQL–4200	<MQL–1800	[5, 10, 18, 22]	<MQL–11,555	<MQL–544	[6, 11–13, 24, 34]
Tetracycline	<MQL–12,340	<MQL–1536	[1, 4, 20, 25, 27]	<MQL–48,000	<MQL–3600	[10, 18, 22]	<MQL–790	<MQL–850	[8, 13, 32]
Trimethoprim	19.5–570	3.7–772	[1, 14, 19, 20, 27]	<MQL–6796	<MQL–37,000	[5, 10, 18, 21]	<MQL–4342	<MQL–3052	[6, 11–13, 17, 24, 34]
Tylosin	<MQL	<MQL	[1, 4]	<MQL–1500	21–720	[10, 18]	<MQL	<MQL–173	[8, 16]
Vancomycin	962–43,740	<MQL	[1]	–	–	–	n.r	<MQL–8514	[8]
Antimicrobials									
Miconazole	<MQL–597	<MQL	[20, 25]	5.2–43	1.6–27	[10]	<MQL–337.9	<MQL–35.7	[35, 36]
Thiabendazole	<MQL–1.29	<MQL	[20, 25]	6.8–220	6.2–140	[10]	–	–	–
Triclocarban	341.1–8880	8.4–5860	[1, 28, 37]	340–4644	64–617	[10, 38]	97–140	n.r	[39]
Triclosan	1.3–2500	49.1–263.9	[1, 28, 37, 40]	14–6817	3.1–360	[10, 38, 41]	<MQL–5260	<MQL–430	[13, 34, 39]

Selected ECs	Asia			North America			Europe		
	Influent	Effluent	Reference	Influent	Effluent	Reference	Influent	Effluent	Reference
NSAIDs									
Acetaminophen	67–147,700	<MQL–2568	[19, 20, 25, 33, 42]	21,000–500,000	<MQL–62,000	[9, 10, 38]	<MQL–482,687	<MQL–24,525	[6, 13, 17, 34, 43]
Codeine	<MQL–242	<MQL–208	[25, 28]	77–5700	80–3300	[10]	150–32,295	9.7–15,593	[17, 23, 44]
Diclofenac	13–445	<MQL–69.2	[20, 25, 29, 40, 42]	140–2450	<MQL–359	[30, 41, 45]	<MQL–4869	<MQL–5164	[13, 17, 34, 43, 46]
Fenoprofen	<MQL–2260	<MQL–23.4	[25, 42]	<MQL	<MQL–405	[45, 47]	n.r	<MQL–280	[48]
Ibuprofen	34.8–55,975	<MQL–1890	[19, 20, 25, 28]	2500–45,000	16–14,600	[5, 10, 30, 41, 45]	<MQL–83,500	<MQL–24,600	[13, 17, 34, 43, 46]
Indomethacin	<MQL–449.4	<MQL–61.4	[25, 42]	<MQL–640	<MQL–507	[41, 45, 47]	<MQL–297	<MQL	[6]
Ketoprofen	<MQL–286	<MQL–183	[25, 28, 29]	60–150	40–90	[41]	<MQL–5700	<MQL–1620	[6, 13, 17, 48]
Naproxen	<MQL–7762	<MQL–159	[19, 25, 29, 42]	1700–25,000	<MQL–3500	[9, 10, 45, 47, 49]	<MQL–611,000	<MQL–33,900	[6, 13, 17, 34, 43]
Salicylic acid	167–16,900	<MQL–1426	[20, 42]	2820–27,800	<MQL–320	[41, 47]	<MQL–164,400	<MQL–10,100	[6, 17, 34, 43]
Beta-blockers									
Atenolol	<MQL–294,700	<MQL–518.6	[9, 19, 25, 29, 42]	500–2642	<MQL–14,200	[5, 9]	<MQL–33,106	<MQL–7602	[6, 13, 17, 24]
Metoprolol	<MQL–79,500	<MQL–268	[9, 14, 25, 29]	16–154	15–212	[9]	<MQL–4148	<MQL–5762	[6, 13, 17, 23, 24]
Propranolol	<MQL–9.56	<MQL–8.3	[14, 25]	–	–	–	<MQL–1962	<MQL–615	[6, 13, 17, 23, 24]
Anticonvulsants									
Carbamazepine	<MQL–18,500	<MQL–900	[20, 25, 28, 29, 42]	<MQL–440	28–551	[5, 9, 30, 45, 50]	<MQL–3110	<MQL–4596	[6, 13, 17, 24, 34, 46, 51]
Gabapentin	4825.5–15,359	213–8855	[42]	n.r	1000 ± 900	[52]	6442–25,079	7651–56,810	[17, 24]
Sulpiride	64.9–15,358.8	70.7–322.4	[14, 42]	n.r	33–137	[53]	113–1100	110–294	[54]
Artificial sweeteners									
Acesulfame	560–13400	5840–9147	[20, 40, 55]	90–2270	600–4330	[56]	12,000–43,000	15,000–46,000	[57]
Cyclamate	<MQL–66,400	<MQL–160	[20, 28, 55]	–	–	–	10,000–65,000	<MQL–450	[57]
Saccharin	9310–389,000	<MQL–2370	[20, 28, 55]	1860–25,100	220–700	[56]	7100–18,000	<MQL–1800	[57]
Sucralose	1100–6520	1300–5490	[20, 28, 40, 55]	17,500–46,100	18,700–48,900	[56]	2000–9100	2000–8800	[57]
Lipid regulators									
Bezafibrate	16.8–159	<MQL–51.4	[14, 58]	–	65–359	[45]	100–7600	<MQL–4800	[13, 23, 24, 34, 46, 59]
Clofibric acid	<MQL–65	<MQL–44.9	[14, 20, 25, 29, 42, 58]	<MQL	<MQL–44	[45, 47]	<MQL–265.9	<MQL–91	[6, 17, 23, 34]
Gemfibrozil	<MQL–453.4	<MQL–535.2	[14, 20, 25, 29, 58, 60]	<MQL–36,530	<MQL–1493	[45, 47]	<MQL–17,055	<MQL–5233	[6, 23, 34, 43]

Selected ECs	Asia			North America			Europe		
	Influent	Effluent	Reference	Influent	Effluent	Reference	Influent	Effluent	Reference
Hormones									
Estrone	<MQL–132.5	<MQL–51.2	[29, 42, 61, 62]	8–52	<MQL–56	[5, 38, 41, 47]	2.4–670	<MQL–95	[13, 46, 63]
Estriol	<MQL–802	<MQL–30.2	[29, 42, 61]	<MQL–217	<MQL	[38, 64]	<MQL–660	<MQL–275	[13, 46, 63]
17α-ethinylestradiol	<MQL–26.1	<MQL–13.1	[61]	<MQL–242	<MQL	[30, 64]	0.4–70	0.5–106	[13, 46]
X-ray contrast media									
Iohexol	63.8–124,966	2100–8700	[20, 37, 40, 42]	n.r	8623–9237	[65]	18,000 ± 2000	1200 ± 100	[66]
Iopromide	47.7–12,200	<MQL–7140	[20, 37]	–	–	–	<MQL–7500	<MQL–9300	[13, 59]
Iopamidol	82.8–45,611	<MQL–6520	[20, 37, 42]	–	–	–	4300 ± 900	4700 ± 1000	[67]
UV filters									
Octocrylene	<MQL	<MQL–153	[42, 68]	–	–	–	100–1200	<MQL–300	[69]
Oxybenzone	<MQL–2616.8	<MQL–772	[28, 42, 68, 70]	–	–	–	<MQL–7800	<MQL–700	[23, 69]
Stimulant									
Caffeine	759–60,500	13–51,700	[14, 19, 20, 28, 60]	5809–82,882	<MQL–37,200	[5, 9, 38]	102–113,200	30–13,900	[6, 43, 71]
Anti-itching									
Crotamiton	<MQL–1500	<MQL–1000	[25, 42, 72]	–	–	–	<MQL–140	<MQL–100	[51]
Insect repellent									
DEET	124–2341.9	21.6–324.8	[14, 20, 42, 58, 60, 72]	200–42,334	13–1663	[9, 30]	<MQL–6900	n.r	[73]
Plasticizer									
Bisphenol A	55.6–5850	<MQL–123	[25, 72]	595–2469	2–450	[9, 41]	<MQL–2376	16–1840	[46, 73]

Possibili soluzioni



Carboni attivo

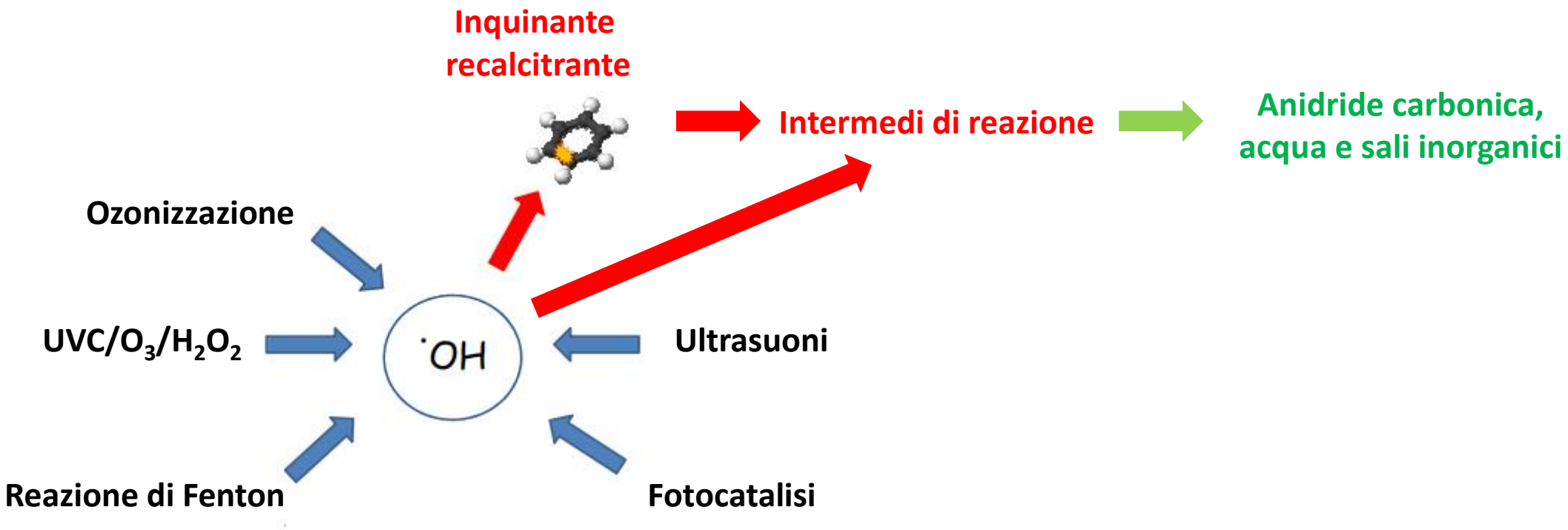
Carboni attivo esausto da rigenerare o da smaltire

Osmosi inversa

Produzione di un ritenato da trattare prima dello smaltimento

Processi di rimozione avanzata

Processi di rimozione avanzata



Fenton

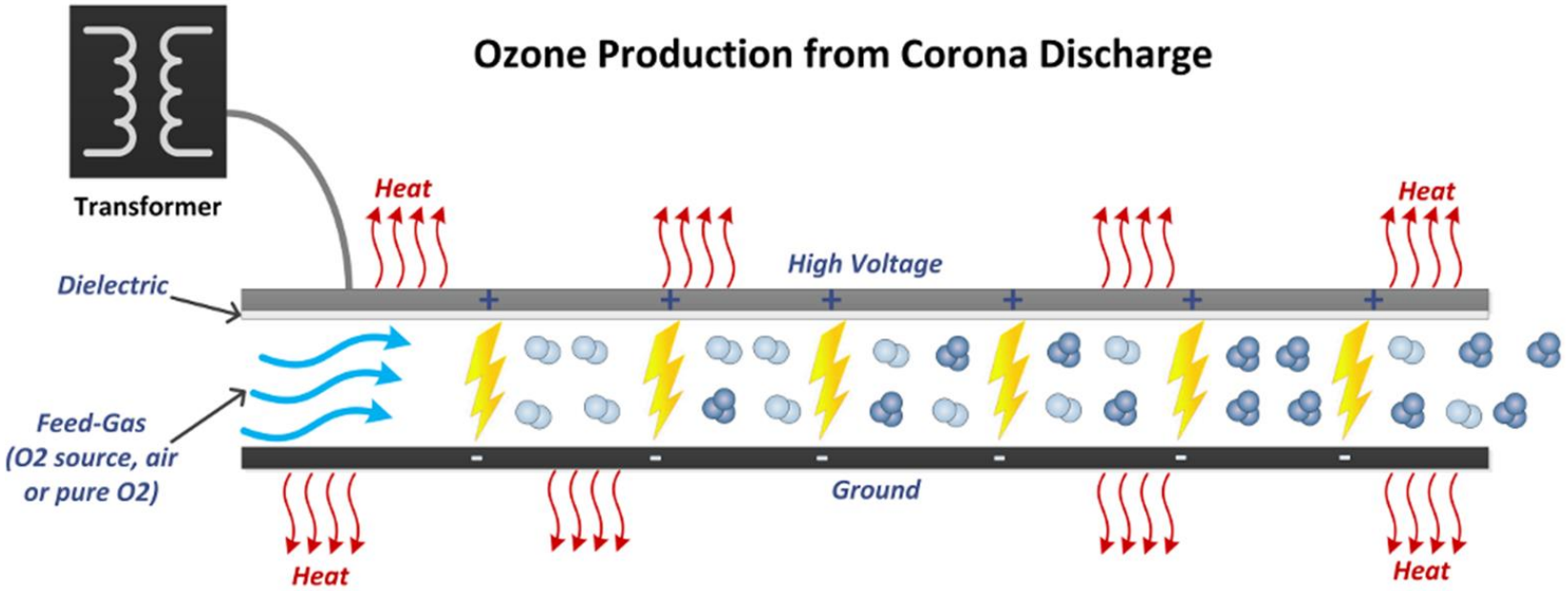
Una reazione scoperta (casualmente) nel 1894 da Henry John Horstman Fenton:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^{\bullet}$$

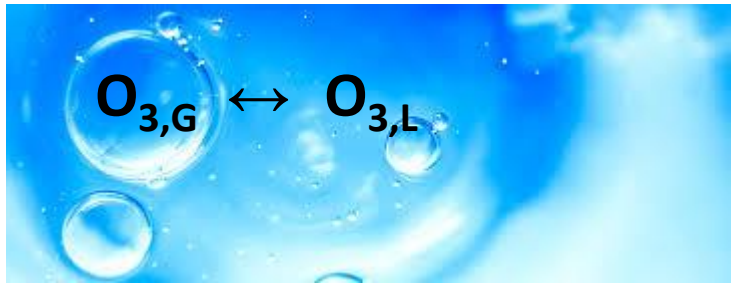
Reazione di Fenton

Si hanno
Elevati consumi di Fe(II), H_2O_2
Produzione di fanghi (Idrossidi di Fe(II) e Fe(III))

Ozonizzazione



Ozone is formed via an electrical discharge that is diffused over an area using a dielectric to create a corona discharge. Oxygen passed through this corona discharge is converted to ozone.

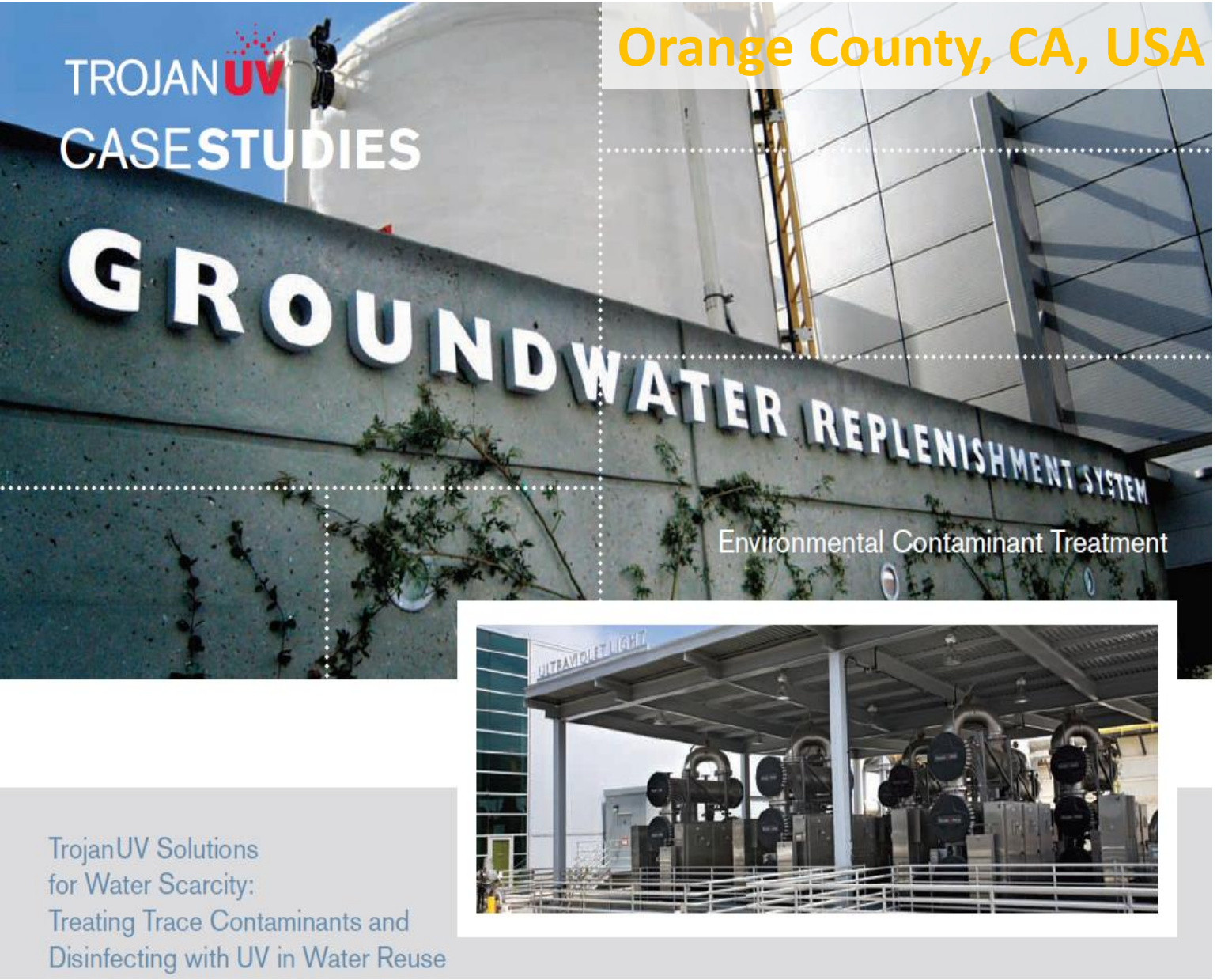


Se il pH della soluzione è maggiore di 5

$$O_{3,L} + HO^- \rightarrow \ldots \rightarrow H_2O_2$$

$$O_{3,L} + H_2O_2 \rightarrow HO^\bullet$$

UVC/H_2O_2



$$UVC + H_2O_2 \rightarrow H_2O_2^*$$

$$H_2O_2^* \rightarrow 2\ HO^\bullet$$

<u>Bastano 3-4 mg/L di acqua ossigenata</u>

Processi di rimozione avanzata alternativi

Processi di rimozione convenzionali:

UVC/H_2O_2
O_3
O_3/H_2O_2
O_3/UVC
$O_3/H_2O_2/UVC$
….

Tali processi richiedono l'impiego di ozono, acqua ossigenata e radiazione UVC. Consumiamo reagenti la cui produzione richiede un grosso dispendio di energia e risorse.

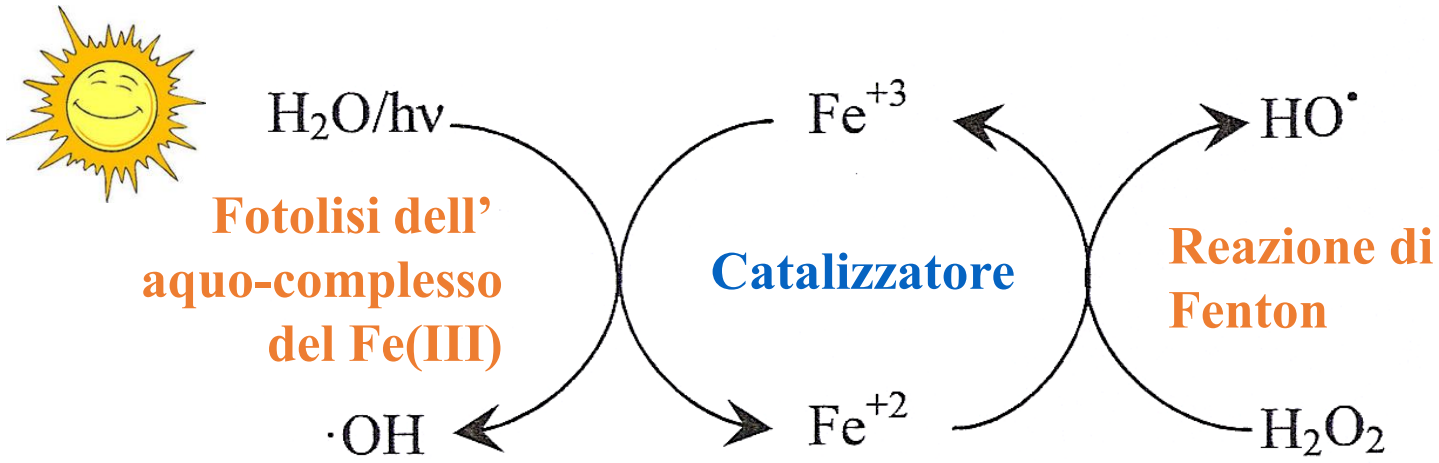
Negli ultimi anni si stanno studiando altri processi di rimozione avanzata più «ecosostenibili»

Tali processi sono caratterizzati da un basso consumo di reagenti e da un impiego di fonti di energia rinnovabile.

Un esempio sono i **<u>processi di rimozione avanzata fotocatalitici</u>**

foto-: usano una radiazione… in particolare, la radiazione solare;
-catalitici: usano un catalizzatore.

Foto-Fenton

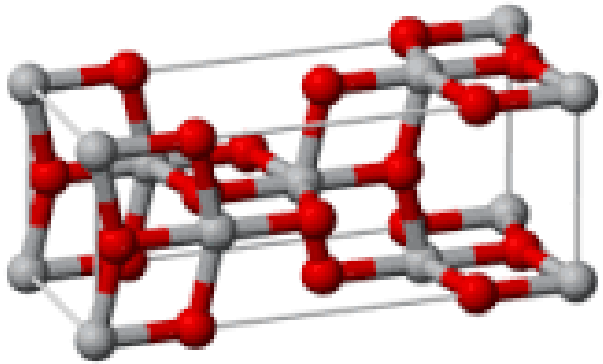


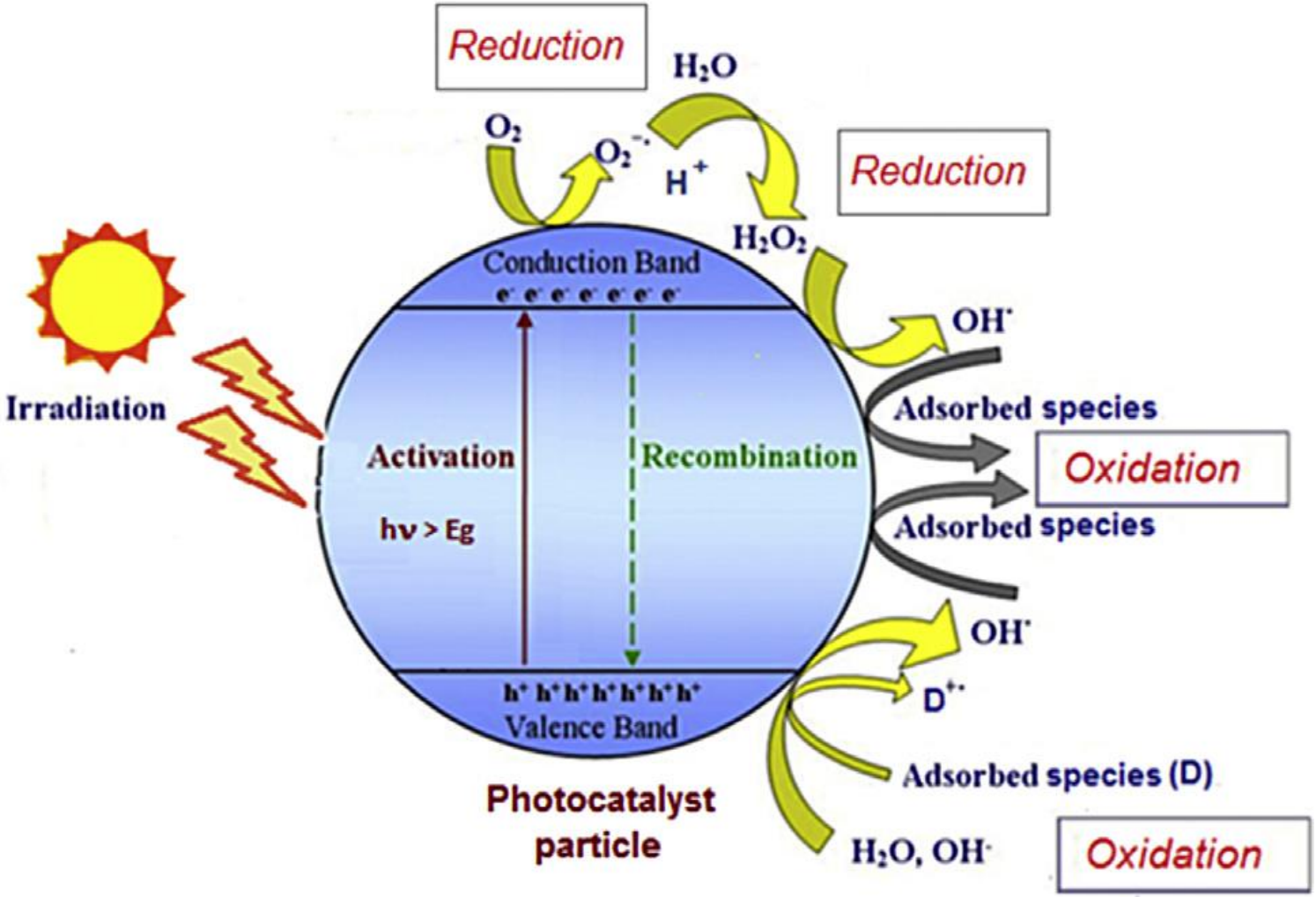
Il ferro è un catalizzatore perché non si consuma!

Il processo sfrutta una fonte di energia rinnovabile: la radiazione UV solare.

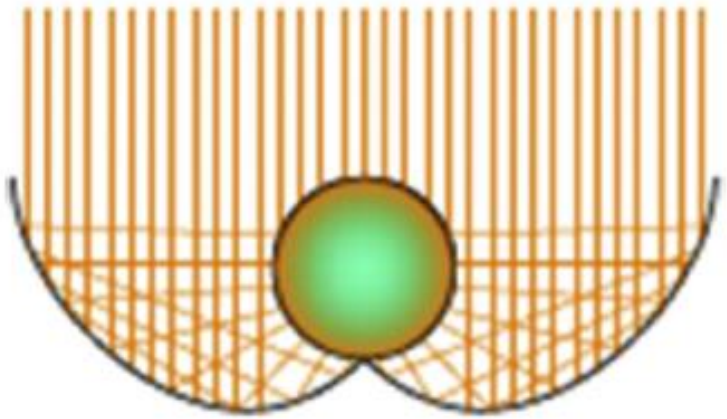
Biossido di titanio

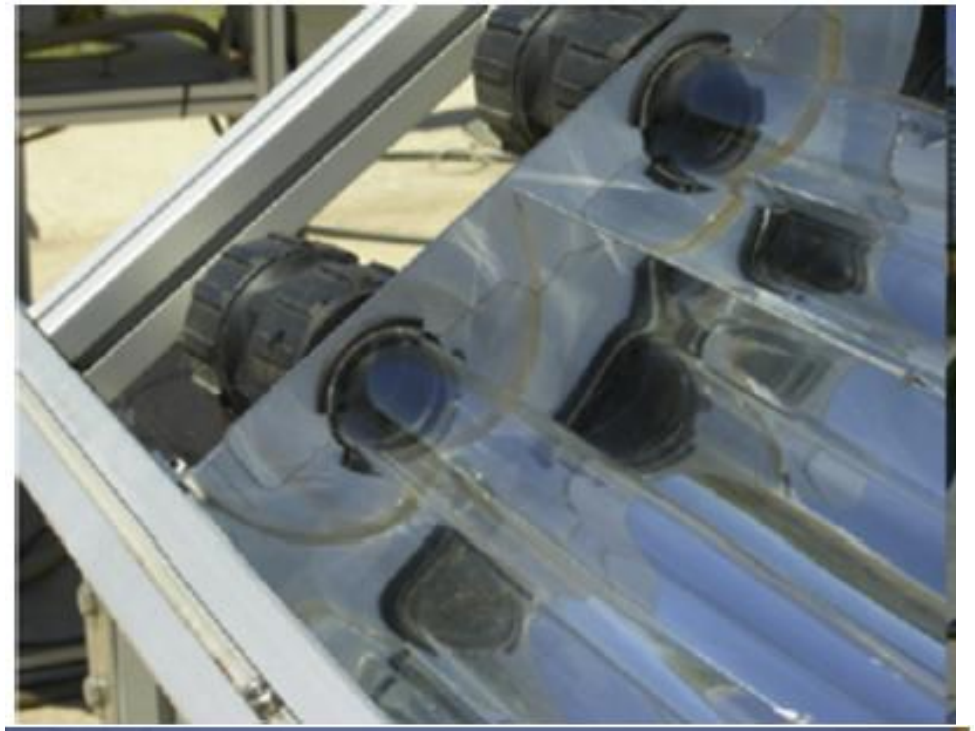
Molti dei problemi legati all'adozione del foto-Fenton possono essere superati usando il biossido di titanio come catalizzatore e la radiazione solare UV-visibile.





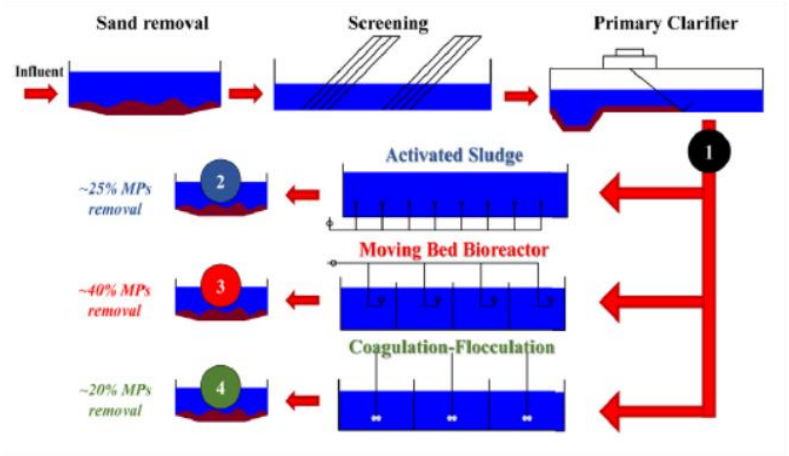
Fotoreattori con ‘compound parabolic collector’





Effect of advanced oxidation processes on the micropollutants and the effluent organic matter contained in municipal wastewater previously treated by three different secondary methods

Stefanos Giannakis [a], Franco Alejandro Gamarra Vives [a], Dominique Grandjean [b], Anoys Magnet [c], Luiz Felippe De Alencastro [b], César Pulgarin [a,*]



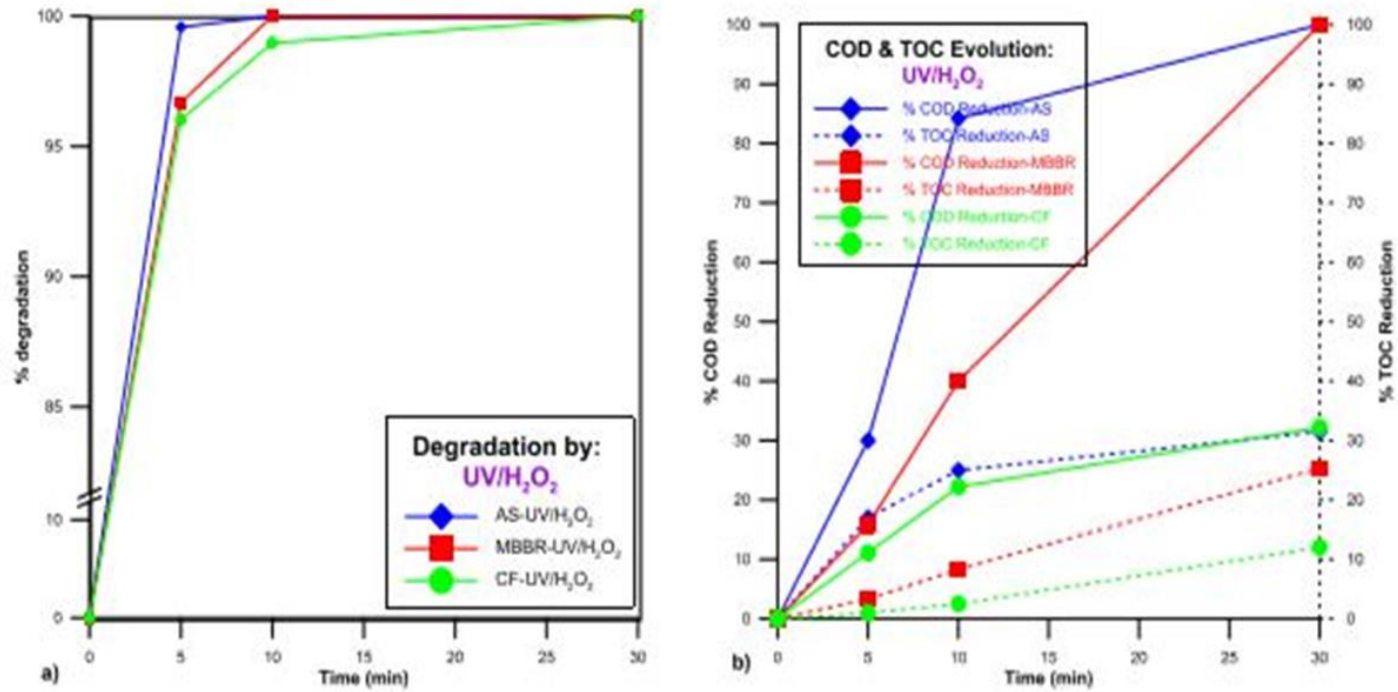
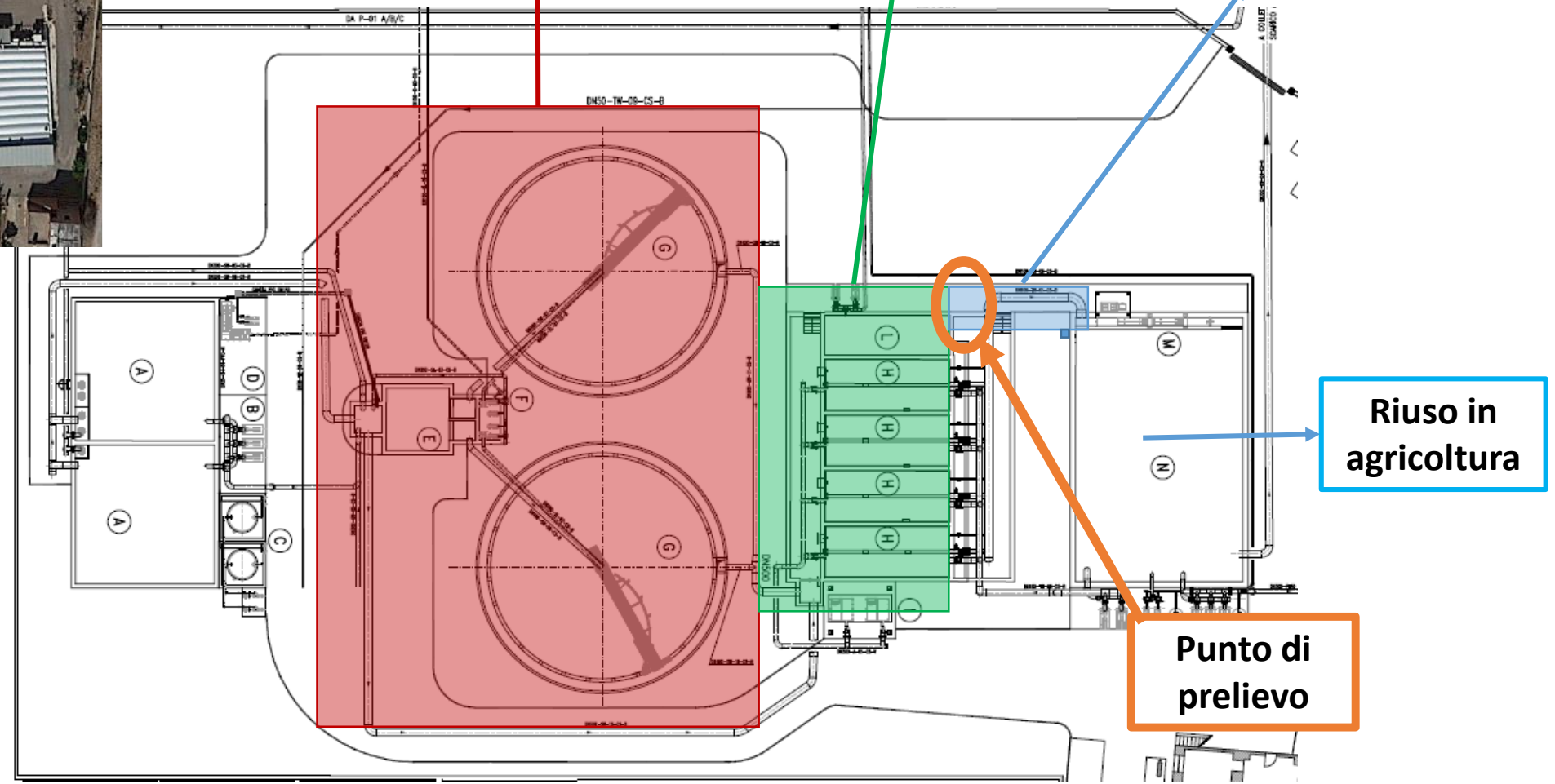


Fig. 3. UV/H_2O_2 treatment results. a) % degradation vs. time. b) % COD & TOC reduction vs. time.

Perché monitorare tutti i contaminanti recalcitranti quando potrebbe bastare monitorare il COD?

Trattamento terziario del depuratore di Gallipoli



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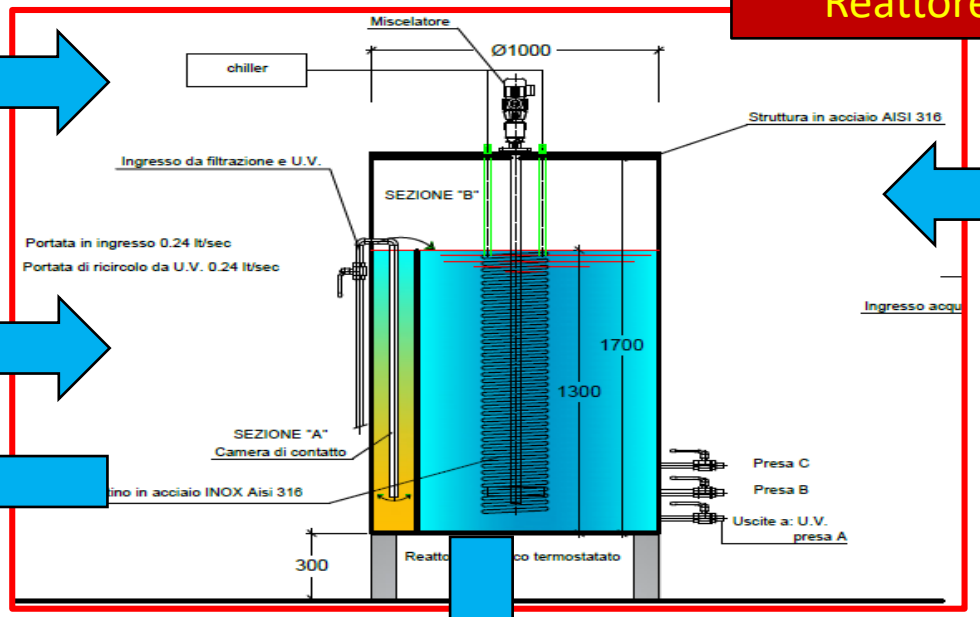
Re- Water

Stazione dosaggio H_2O_2

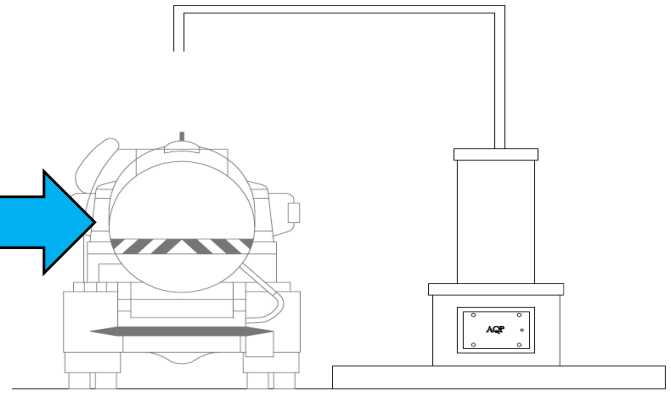
Stazione disinfezione UV

Reattore CSTR



Arrivo dalla filtrazione dell'affinamento

ACCUMULO



Grazie per la cortese attenzione

danilo.spasiano@poliba.it

Concentrazione di inquinanti organici recalcitranti nei fanghi di depurazione (ng/g ST)

Selected ECs	Sludge (ng/g dw)	Reference
Antibiotics		
Amoxicillin	<MQL	[1]
Azithromycin	<MQL–666	[2, 3]
Ceftazidime	–	–
Chloramphenicol	<MQL	[6]
Chlortetracycline	<MQL–1908	[6–8]
Ciprofloxacin	1400–4800	[9, 10]
Clarithromycin	<MQL–537	[2, 6]
Clindamycin	<MQL–6.54	
Enrofloxacin	7.6–11560	[8, 11]
Erythromycin	6–79	[6, 8]
Erythromycin-H_2O	–	–
Lincomycin	<MQL–4967	[8, 12, 13]
Meropenem	–	–
Minocycline	1000–2000	
Ofloxacin	1480–5760	[6, 10, 11]
Oxytetracycline	<MQL–3790	[7, 11]
Sulfamethazine	<MQL–54.58	[7]
Sulfamethoxazole	<MQL–84.4	[7, 11, 13]
Tetracycline	<MQL–466	[6, 11]
Trimethoprim	<MQL–13	[1, 2, 12, 13]
Tylosin	31–139	[8]
Vancomycin	<MQL	[8]
Antifungal/antimicrobials		
Miconazole	<MQL–2609	[2, 11, 12, 14]
Thiabendazole	<MQL–10.6	[11, 12]
Triclocarban	362–8460	[2, 11–13]
Triclosan	354–15600	[2, 10, 11, 13]
NSAIDs		
Acetaminophen	<MQL–586	[2, 6, 11, 12]
Codeine	<MQL–79	[11, 13] [2]
Diclofenac	<MQL–133	[2, 6, 11, 16]
Fenoprofen	<MQL	[11]
Ibuprofen	<MQL–3988	[10, 11, 13, 16]
Indomethacin	<MQL–77	[6, 11] [1]
Ketoprofen	<MQL–58.4	[6, 11–13]
Naproxen	<MQL–1022	[10, 11, 16]
Salicylic acid	<MQL–13743	[10, 16]

Selected ECs	Sludge (ng/g dw)	Reference
Beta-blockers		
Atenolol	<MQL–86	[1, 6, 11]
Metoprolol	<MQL–226	[6, 11]
Propranolol	<MQL–849	[2, 6, 11]
Anticonvulsants		
Carbamazepine	<MQL–50	[1, 2, 11, 13]
Gabapentin	–	–
Sulpiride	–	–
Artificial sweeteners		
Acesulfame	<MQL–166	[12, 13, 17]
Cyclamate	66.6–544	[13]
Saccharin	<MQL–19200	[12, 13, 17]
Sucralose	<MQL–1980	[12, 13, 17]
Lipid regulators		
Bezafibrate	17–64	[6]
Clobric acid	< MQL	[6, 11]
Gemfibrozil	< MQL–1192	[10, 11]
Hormones		
Estrone	<MQL–17.5	[16, 18]
Estriol	<MQL–49	[10, 16, 18]
17α-ethinylestradiol	<MQL–17	[10, 16]
X-ray contrast media		
Iohexol	–	–
Iopromide	80 ± 4	[20]
Iopamidol	–	–
UV filters		
Octocrylene	1060–9170	[21]
Oxybenzone	<MQL –790	[11, 12, 21]
Anti-itching		
Crotamiton	<MQL–62	[11] [6]
Insect repellent		
DEET	2–6	[6]
Stimulant		
Caffeine	<MQL–805	[2, 6, 11]
Plasticizer		
Bisphenol A	<MQL–4700	[11, 18, 22]

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Water Reuse in Europe

Relevant guidelines, needs for and barriers to innovation

Laura Alcalde Sanz, Bernd Manfred Gawlik

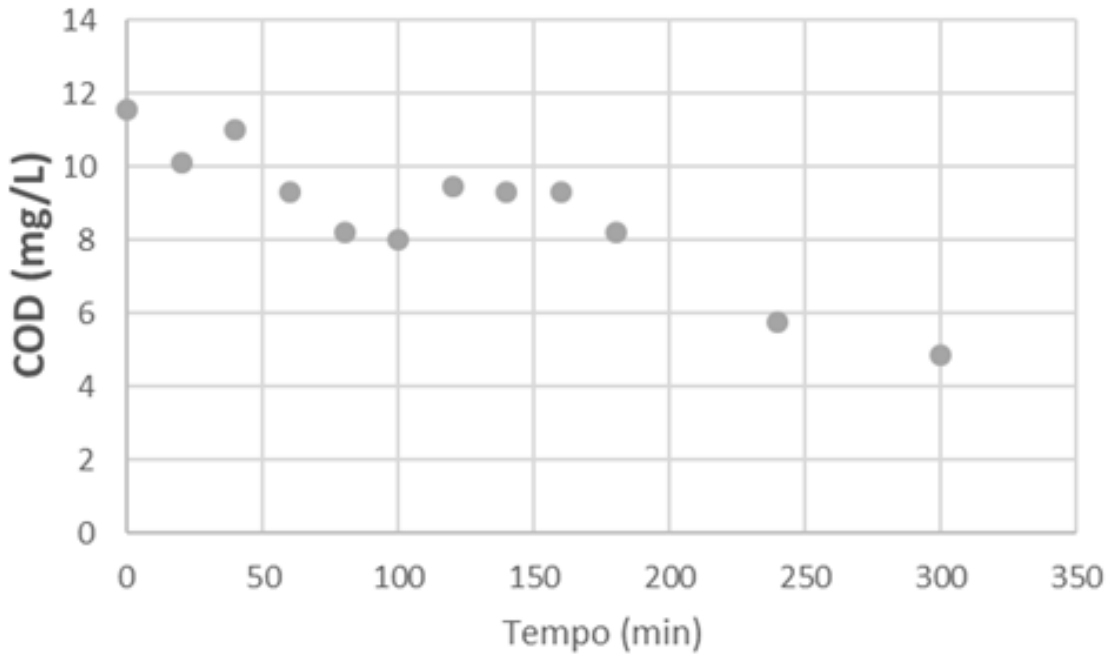
2014

Chemical agents	Adverse effects
Pharmaceuticals and metabolites (antibacterials (sulfamethoxazole), analgesics (acetominophen, ibuprofen), beta-blockers (atenolol), antiepileptics (phenytoin, carbamazepine), veterinary and human antibiotics (azithromycin), oral contraceptives (ethinyl estradiol))	
Personal care products (triclosan, sunscreen ingredients, fragrances, pigments)	Carcinogenic, teratogenic and/or mutagenic effects, risk for human health (cyanotoxins), bioaccumulation, toxicity to plants. Various effects, often unexplored.
Household chemicals and food additives (sucralose, bisphenol A (BPA), dibutyl phthalate, alkylphenol polyethoxylates, flame retardants (perfluorooctanoic acid, perfluorooctane sulfonate)	
Transformation products (NDMA, HAAs, and THMs)	
Industrial chemicals (PFCs, MTBE, solvents, …)	Carcinogenic, teratogenic and/or mutagenic effects, risk for human health (cyanotoxins), bioaccumulation, toxicity to plants. Various effects, often unexplored.
Pesticides, biocides and herbicides (e.g. atrazine, lindane, diuron, fipronil)	
Natural chemicals (hormones, phytoestrogens, geosmin, 2-methylisoborneol)	

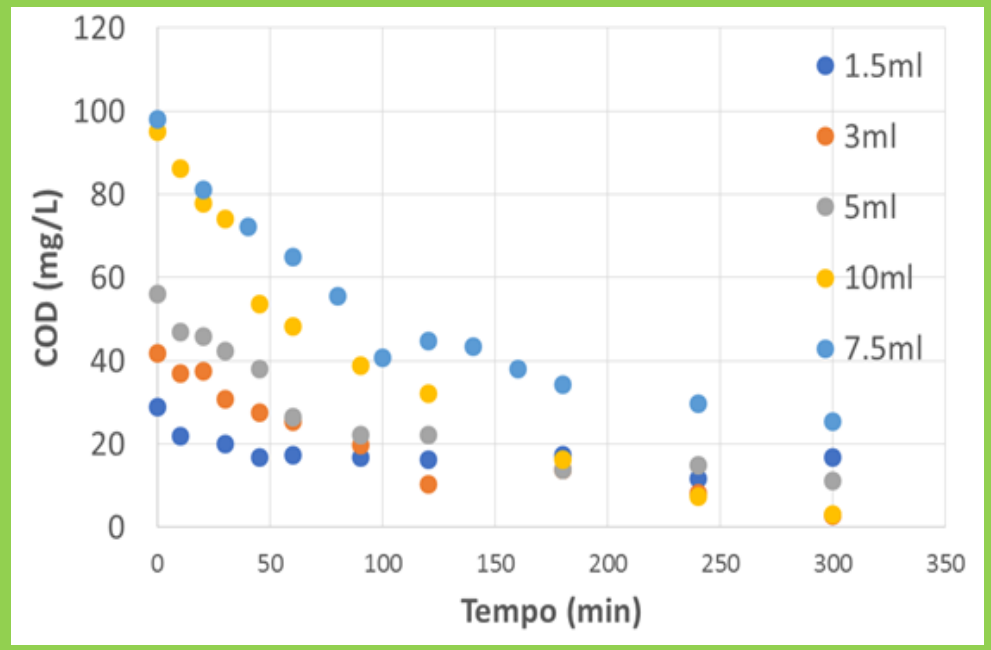
Dives in Misericordia Church in Rome (left); Cité de la Musique et des Beaux-Arts in Chambéry (right).

Risultati sperimentali

NO H_2O_2



CON H_2O_2



Modellazione del processo UV/H_2O_2

$$COD + hv \rightarrow by - products$$

$$H_2O_2 + hv \rightarrow 2HO^{\cdot}$$

$$HO^{\cdot} + H_2O_2 \xrightarrow{k_w} H_2O + HO_2^{\cdot}$$

$$2HO_2^{\cdot} \xrightarrow{k_t} H_2O_2 + O_2$$

$$COD + HO^{\cdot} \xrightarrow{k_{OH/COD}} BP + HO^{\cdot} \rightarrow by - products$$

Schema di reazioni

Viene considerato al pari di uno pseudo-componente

Dopo aver fatto le ipotesi di stato stazionario sulle specie radicaliche ($HO^{\cdot}$ and $HO_2^{\cdot}$) e dopo aver fatto un po' di passaggi matematici…

Modellazione del processo UV/H_2O_2 (2)

$$\frac{d[H_2O_2]}{dt} = -F[H_2O_2] - F[H_2O_2]\frac{k_{OH,H_2O_2}[H_2O_2]}{k_{OH,H_2O_2}[H_2O_2] + k_{OH,COD}[COD]}$$

$$\frac{d[COD]}{dt} = -F[COD] - 2F[H_2O_2]\frac{k_{OH,COD}[COD]}{k_{OH,H_2O_2}[H_2O_2] + k_{OH,COD}[COD]}$$

Dove:

$k_{OH,H_2O_2} = 2.7 \cdot 10^{-4} mM^{-1}s^{-1}$ *(Buxton et al., 1988)*

$$F[H_2O_2] = \varphi^{254}_{H_2O_2} \cdot \frac{I_0^{254}}{V_{TOT}} \cdot (1 - \exp(-2.3 \cdot L \cdot A_{TOT}) \cdot g_{H_2O_2}$$

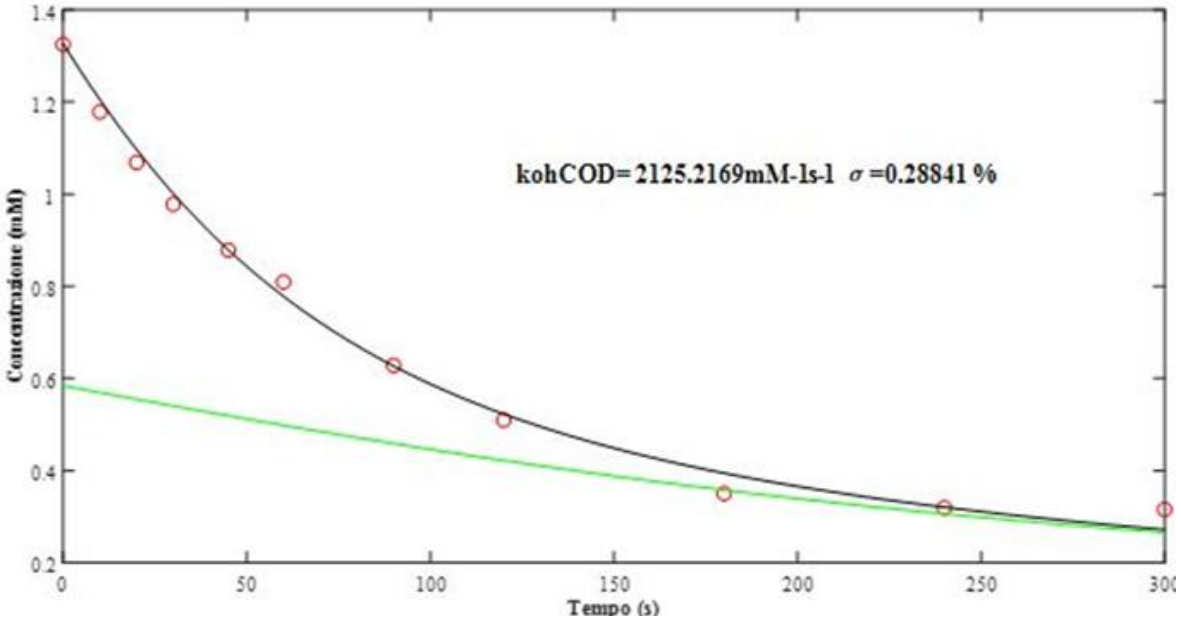
Parametro incognito

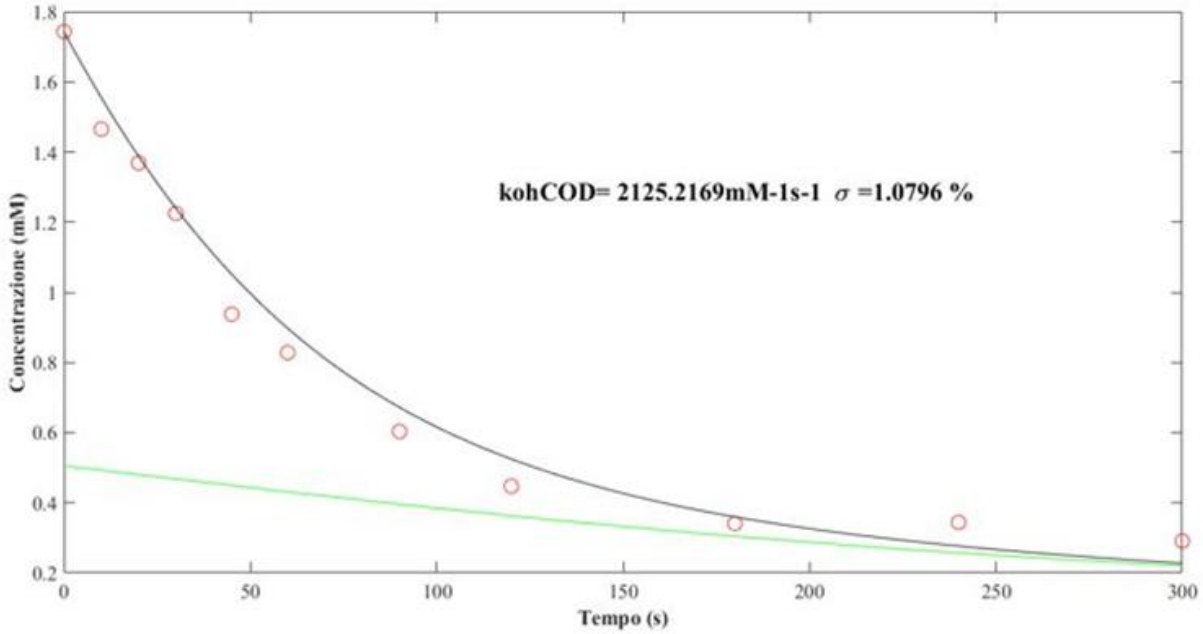
$$F[COD] = \varphi^{254}_{COD} \cdot \frac{I_0^{254}}{V_{TOT}} \cdot (1 - \exp(-2.3 \cdot L \cdot A_{TOT}) \cdot g_{COD}$$

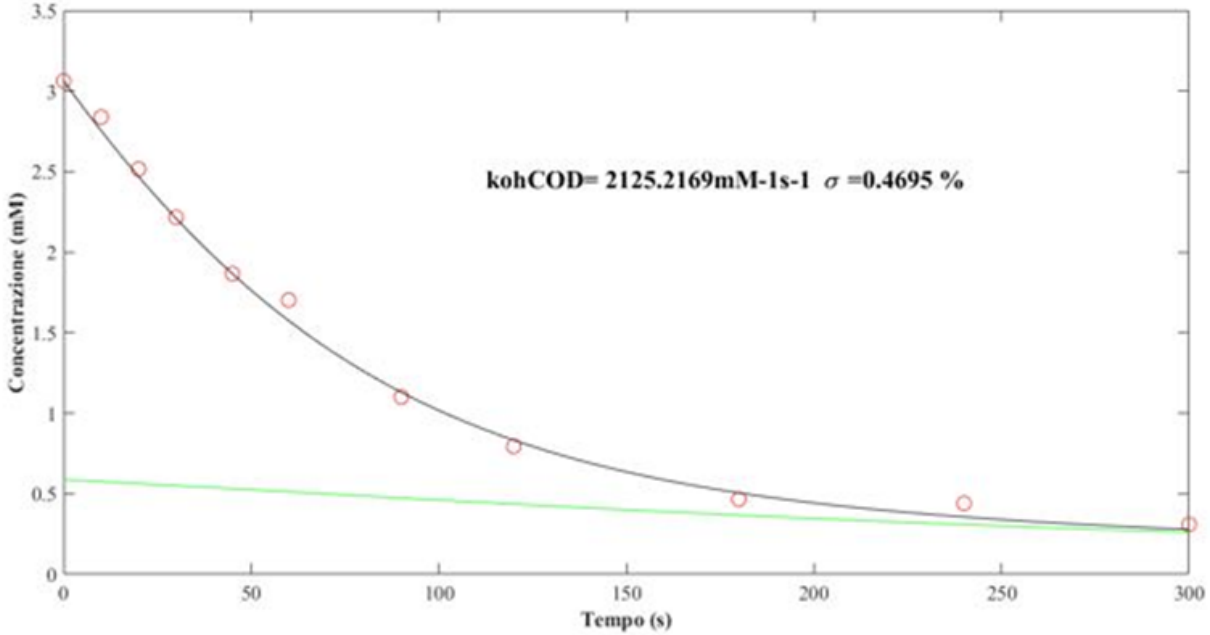
$$A_{TOT} = (\varepsilon^{254}_{H_2O_2}[H_2O_2] + \varepsilon^{254}_{COD}[COD])$$

$$g_{H_2O_2} = \frac{\varepsilon^{254}_{H_2O_2}[H_2O_2]}{A_{TOT}}$$

$$g_{COD} = \frac{\varepsilon^{254}_{COD}[COD]}{A_{TOT}}$$





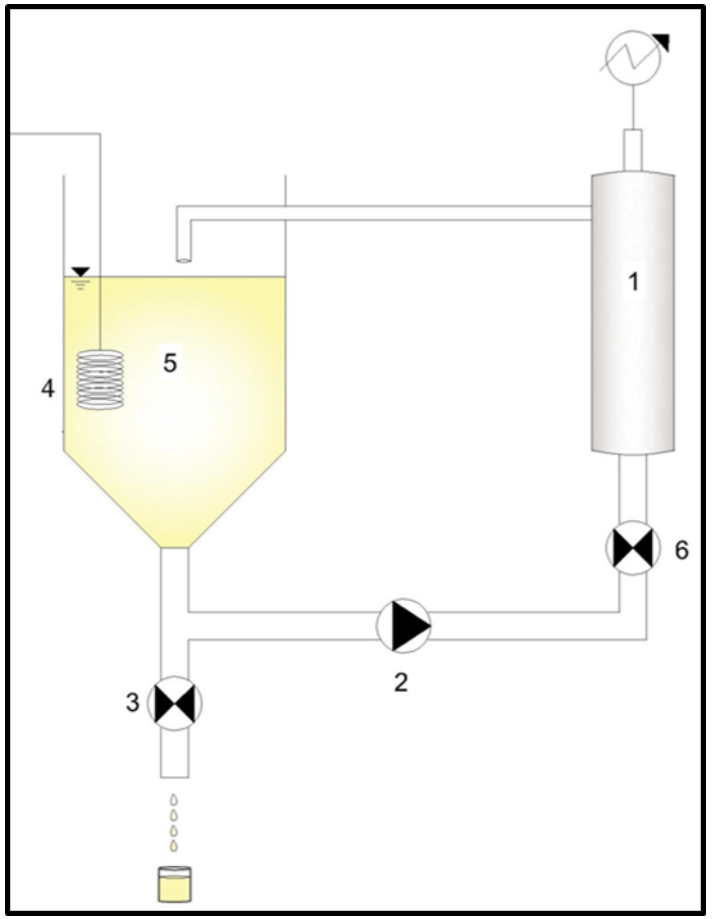


$$k_{OH,COD}=2125 mM^{-1}\cdot s^{-1}$$

La linea continua nera rappresenta la concentrazione teorica del COD totale, ovvero la somma del COD relativo ai composti inizialmente disciolti nell' effluente del depuratore (**linea verde**) ed il COD relativo alla presenza di H_2O_2

$$[COD]_{t,total} = [COD]_t + [H_2O_2]_t \cdot 0.48 \cdot \frac{34}{32}$$

Apparato sperimentale



1 Lampada UV
2 Pompa di ricircolo
3 Valvola per prelievi
4 Raffreddamento
5 Vasca di ricircolo
6 Valvola di regolazione