



Photocatalytic removal of diazinon from aqueous solutions: a quantitative systematic review

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Abstract

Diazinon is a widely used pesticide that can be effectively degraded in aqueous solutions via photocatalytic oxidation. This quantitative systematic review was conducted to shed light on the various aspects of photocatalytic diazinon removal based on evidence. A systematic search was performed in Scopus, PubMed, Web of Science, Embase, and Ovid databases with keywords including diazinon, photocatalysis, and their equivalents. The search was limited to original articles in English published between January 1, 2010, and March 25, 2021. The results were expressed by descriptive statistics including mean, SD, median, and percentiles, among others. The initial electronic and manual search retrieved 777 articles, among which 41 studies comprising 49 trials were qualified for data synthesis. The reported diazinon degradation rate ranged from 2 to 100%, with a mean \pm SD of $59.17 \pm 28.03\%$. Besides, ZnO/UV, WO₃/UV, TiO₂/UV, and TiO₂/Vis, in sequence, were the most widely used processes with the highest efficacies. Solution pH in the range of 5–8, catalyst dose below 600 mg/L, diazinon initial concentration below 40 mg/L, and contact time of 20–140 min could be the optimum conditions. Diazinon degradation obeyed the first-order kinetic model with k_{obs} between 0.0042 and 1.86 min⁻¹ and consumed energy of 38.93–350.36 kWh/m³. Diazoxon and IMP were the most detected by-products of diazinon degradation although bioassay data were scarce. Based on the results, photocatalytic processes are very efficient in removing diazinon from aqueous solutions although more elaborate studies are needed to assess the mineralization rate and effluent toxicity.

Keywords Advanced oxidation process · Diazinon · Kinetics · Photocatalyst · Photocatalytic oxidation · Toxicity

Introduction

Photocatalytic oxidation processes are drawing much attention for the removal of toxic and refractory pollutants. These processes benefit from catalysts of metal oxides (ZnO, TiO₂, WO₃, CeO₂, Fe₂O₃), metal sulfides (CdS, PbS, CuS),

metal–organic frameworks (MOFs), polymer-based, and so on, in either pristine or modified forms with nano- or micro-sizes to accelerate photo-driven reactions (Choi et al. 2017; Zhang et al. 2020, Samy, Ibrahim et al. 2021). A photocatalytic process is based on the photo-stimulation of semiconductors using ultraviolet or visible light. By absorbing the light with energy enough to exceed the bandgap energy of

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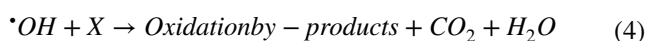
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the semiconductor catalyst, the catalyst generates a positive hole (h_{vb}^+) in its valence band due to electron detachment (Rezaei and Mohseni 2017; Hosseini and Mohebbi 2018). The reaction of the photo-created holes (h_{vb}^+) with H_2O or OH^- produces $\bullet OH$ radicals that strongly oxidize organic pollutants. Besides, the detached electron, which is transferred to the conduction band (e_{cb}^-), participates in the reduction of O_2 to $\bullet O_2^-$, which is another agent capable of oxidizing organic species (Cadiou et al. 2020, Liu, Guo et al. 2020). Reactions driving the photocatalytic degradation of an organic pollutant are as follows (Yap et al. 2019):



As mentioned, the photocatalyst and the light source are two main role players in the photocatalytic reactions to initiate. Simple photocatalysts such as TiO_2 , ZnO , and WO_3 are desirably used for this purpose, as they are usually associated with low-cost and labor-saving preparation (Biglari et al. 2017, Hadei, Mesdaghinia et al. 2021). However, many researchers are intended to incorporate some compounds into the crystalline structure of the photocatalysts to overcome some problems with simple photocatalysts or enjoy photocatalysts with new features. Preventing from the quick recombination of electrons and holes (Kumara et al. 2021), narrowing the bandgap and hence expanding the light spectrum for photocatalytic activation (Gong, Wu et al. 2021), increasing the separation properties of the photocatalyst from the bulk solution (Wang, Gao et al. 2021), and promoting the photostability of the photocatalyst (Goulart, Santos et al. 2021) are some reasons for modifying simple photocatalysts. Therefore, binary photocatalysts such as MoS_2/ZnS (Joy et al. 2017), tertiary photocatalysts such as $TiO_2-Au-CdS$ (Zhao et al. 2016), and doped photocatalysts such as $Fe-TiO_2$ (Borji et al. 2014; Tabasideh et al. 2017), $Ag-TiO_2$ (Parastar et al. 2013), and $Er-ZnO$ (Chemingui et al. 2021) have been developed.

The light source provides the energy required for the excitation of the photocatalyst in photocatalysis reactions. The

light needs to provide valence band electrons with ample energy to move to the conduction band. The UV light at the wavelength of 254–365 nm is most widely used for photo-activating the catalysts (Dong et al. 2005; Daiqi and Xiaoshan 2010), as it emits highly energetic photons capable of exciting a wide variety of semiconductors with various bandgaps including TiO_2 (band gap energy 3.2 eV), ZnO (3.2 eV), ZnS (3.7 eV), WO_3 (2.8 eV), and V_2O_5 (2.7 eV). A large portion of the operating costs of a photocatalytic system is devoted to the energy required by the light source. In recent decades, a significant contribution has been made to invent visible light-harvesting catalysts so they can be activated with less energy cost under visible light which is also safer than UV. Some of them include Pt-doped TiO_2 (Nakaoka et al. 2010), N-doped TiO_2 (Salarian et al. 2016), WO_3 -doped ZnO (Maleki, Moradi et al. 2020), and Cd_3OSO_4 (Mohammadzadeh Kakhki and Ahsani 2020), which can also be activated in solar systems.

Like any other technology, there are some pros and cons for photocatalytic oxidation processes. Concerning advantages, they run under ambient temperature and pressure; employ some photo-stable, low-cost, readily available, and effective catalysts; and are capable of complete oxidation of refractory organics without the generation of unwanted by-products including sludge (Gunasekar et al. 2013; Favier et al. 2019, Li, Ivanenko et al. 2019, Noor, Sajjad et al. 2020). Among the disadvantages, we can point to the need for some high-cost light sources, e.g., UV and xenon lamps, the occurrence of some competing side-reactions, the fast recombination of electron–hole in some photocatalysts, the need for downstream separation of the catalyst, and the loss of photo-activity of the catalyst in successive use which necessitates the continuous addition of the fresh catalyst (Yousaf, Nadeem et al., Katzenberg et al. 2020). Despite the limitations, the photocatalytic oxidation of persistent organic pollutants has shown to be promising in the detoxification of effluents containing pesticides and pharmaceutical residues, providing a cleaner environment.

Diazinon with the formula of $C_{12}H_{21}N_2O_3PS$ is a commonly used insecticide relying on phosphorus as one of its main constituents. This organophosphorus insecticide is classified by the World Health Organization (WHO) as “probably carcinogenic to humans” (class II) chemicals used to control pests mostly in agricultural fields (Kalantary et al. 2014; Fritschi et al. 2015). Although its usage is widely restricted in some regions such as the USA and the European Union, it is still one of the popular insecticides around the world (Khun et al. 2021). Diazinon has been tracked in water resources including freshwater, seawater, and even wastewater treatment plant effluents and some foods (Hassan, Rahaman et al. 2019). It is toxic to aquatic organisms at a concentration of 350 ng/L (Kalantary et al. 2014). Although there are limited data on the carcinogenic effects of diazinon

on humans, there is some evidence about the adverse effects of diazinon on the liver and kidneys (Fritschi et al. 2015, Hassan, Rahaman et al. 2019). Due to its relative water solubility, non-polarity, moderate mobility, and persistence in soil, diazinon makes a great deal of concern for surface and groundwater contamination (Brady et al. 2006; Saraji et al. 2018). Therefore, its removal from aqueous solutions before discharging to the environment is of utmost importance.

Based on a literature review, there are tens of studies showing that photocatalytic oxidation is capable of complete or nearly complete removal of diazinon. These studies have tried relatively fixed operational parameters but in wide ranges. For instance, contact time has changed from 3 to 360 min and diazinon concentration from 1 to 500 mg/L. Besides, studies have utilized different catalysts to examine their efficacy for photo-activation and diazinon removal. Therefore, a comprehensive study may be helpful to summarize the data of previous studies and make firm conclusions, with the hope that results would help better understand the status quo and design more effective studies in the future. As a result, this quantitative systematic review was conducted to explore the operational parameters, their levels, and effects on selected outcomes in the photocatalytic removal of diazinon using various catalysts and light sources.

Materials and methods

Study protocol and search strategy

The recommendations of the Cochrane Handbook for Systematic Reviews of Interventions (Higgins and Green 2008) were followed for the conduct of this study. The protocol of the study was submitted in the International Prospective Register of Systematic Reviews (ID = CRD42020166279). The Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement (Moher, Liberati et al. 2009) was used to organize the current report.

To find studies that used photocatalytic processes for removing diazinon from aqueous solutions, a systematic search was performed in Scopus, PubMed, Web of Science, Embase, and Ovid databases. The search strategy contained the MeSH and non-MeSH terms including diazinon or equivalents (dimpylate or neocidol) and photocatalysis, photocatalytic, or photocatalyst in titles, abstracts, or keywords. Full search strategy in Scopus, for instance, was as follows: photocatalysis OR “photo catalysis” OR photocatalytic OR “photo catalytic” OR photocatalyst OR “photo catalyst” AND diazinon OR dimpylate OR neocidol. To avoid missing relevant papers possibly absent in the electronic search, the reference lists of the eligible studies were also checked out manually.

Eligibility criteria and study selection

Original articles in the English language investigating the photocatalytic removal of diazinon from aqueous solutions reporting on the diazinon degradation rate were included in the study. To obtain the most recent achievements, the search was limited to papers published between January 1, 2010, and March 25, 2021. Studies using auxiliary oxidants or purging gases, diazinon mixture with another pollutant, or those with non-extractable data were excluded from the study. The papers were reviewed and screened by two authors (FBA and MD) for the eligibility criteria, and any discrepancies in the study selection were discussed and resolved by the third author (A. h. M).

Quality assessment

The quality assessment of the selected studies was done by a researcher-made scale. This was done to avoid the risk of bias due to unqualified studies. The scale was constructed for the methodological assessment of interventional photochemical studies. The comments of five experts were sought to validate the scale. As shown in Supplementary File Table S1, the scale included 10 items with yes/no answers on various methodological aspects. Studies with at least five yeses were used for data extraction.

Data extraction

Two researchers (FBA and MD) independently read the selected papers and extracted the following data: authors' name, article title, journal name, publication year, origin country, light source, catalysts, operational parameters, and their levels, degradation rate, mineralization rate, energy consumption, and reusability data. To affirm the accuracy of data, the extracted data were scrutinized, and any disagreement was discussed to reach a consensus. If there was some ambiguity in expressing data in a paper, we contacted the authors via email.

Data analysis

Quantitative data on operational variables and outcome measures were analyzed and presented with descriptive statistics including mean, standard deviation (SD), mode, minimum, maximum, the first percentile (Q1), median, the third percentile (Q3), the 90th percentile, the 95th percentile, and the 99th percentile. The statistics were computed separately for various photocatalytic processes grouped based on the types of the base photocatalyst and light source (UV/Vis). The relationships between operational variables and diazinon removal rates were examined using histograms drawn for removal rates above the 75th percentile (Q3) in

each category. ImageJ software was used to extract data from graphs and OriginPro 2015 for drawing the graphs.

Results

Figure 1 illustrates the PRISMA 2009 flow diagram of the study selection process. In summary, the electronic search retrieved 765 articles (453 in Scopus, 292 in Web of Science, 4 in PubMed, 10 in Embase, and 6 in Ovid). Besides, 12 studies were found in the reference list of relevant articles. Of 777 studies, 71 were examined in full-text. However, 30 studies were removed because of various reasons (Fig. 1). Finally, 41 articles were subject to quality assessment. Based on the quality assessment, the scores ranged from 5 to 10, indicating all the studies were qualified for data synthesis.

Table 1 summarizes the results of 41 relevant studies (comprising 49 trials) published since 2010 to investigate the effectiveness of photocatalytic processes for diazinon removal from aqueous solutions. There were 29 articles from Iran, four from Egypt, three from Indonesia, one from Thailand, one from Bangladesh, one from Japan, one from Vietnam, and one from

China. Of 49 trials, UV lamps/LED were the sources of light in 30 (61.22%) trials, visible light in 15 (30.61%) trials, and sunlight in four (8.16%) trials. Thus, about 61% of the trials were conducted in the UV light spectrum and 39% in the visible light spectrum. A wide variety of catalysts were used for the photocatalytic removal of diazinon. We categorized them based on the main component exhibiting the photocatalytic property: TiO₂-based ($n=22$ studies; 53.66%), ZnO-based ($n=10$; 24.39%), WO₃-based ($n=2$; 4.88%), metal–organic framework (MOF)-based ($n=2$; 4.88%), VO₄-based ($n=1$; 2.44%), chalcogenide metal-based ($n=1$; 2.44%), MgO-based ($n=1$; 2.44%), hydroxyapatite-based ($n=1$; 2.44%), and polymer-based ($n=1$; 2.44%). Only 14 (34.15%) studies used pure compounds as photocatalysts, while the majority of them ($n=27$; 66%) used a modification method for manipulating the photocatalytic materials. The reasons for modifications included expanding the photocatalytic activity into the visible light spectrum, increasing the capability of the process in removing diazinon, and facilitating the separation of photocatalyst residue among others. Initial solution pH, initial diazinon concentration, catalyst dose, and contact time were the most widely used operational variables whose effects were studied on the outcomes including diazinon degradation rate,

Fig. 1 PRISMA 2009 diagram of the study selection

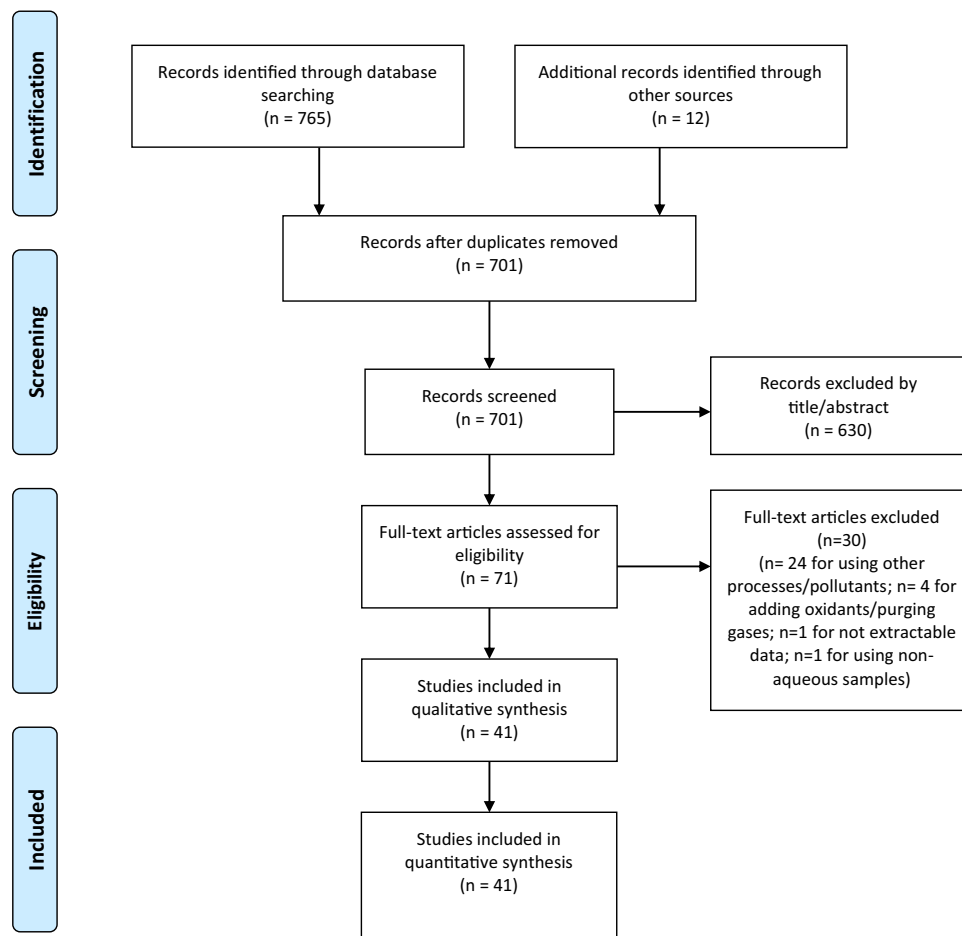


Table 1 Summary of studies assessing photocatalytic removal of diazinon from aqueous solutions from 2010 to date

Studies No	Trials No	Light source	Catalyst name	Base catalyst ¹	Maximum degradation rate (%)	Maximum mineralization rate (%)	Reusability ² [degradation rate % (cycles)]	k_{obs} (min ⁻¹) ³	L-H model parameters	E_{EO} (kWh/m ³) ⁴	Quality score (out of 10)	Study
1	1	Vis	Pt-TiO2	TiO2	100	88	NA	$k_{obs} = 0.170$ $R^2 = UN$	NA	NA	8	Nakaoka et al. (2010)
2	2	UV	Fe3O4/hydroxyapatite	Hydroxyapatite	75	NA	95 (7)	NA	NA	NA	8	Yang et al. (2010)
3	3	UV	ZnO	ZnO	86.74	NA	NA	$k_{obs} = 1.86$ $R^2 = 0.8954$	NA	NA	8	Fadaei and Dehghani (2012)
4	4	UV	FeFNS-TiO2	TiO2	96.3	67.9	NA	$k_{obs} = 0.016$ $R^2 > 0.99$	NA	NA	8	Hossaini et al. (2014)
5	5	UV	TiO2	TiO2	97.3	44.2	NA	$k_{obs} = 0.102$ $R^2 = 0.994$	NA	NA	7	Jafari et al. (2016)
6	6	UV	ZnO	ZnO	93.3	NA	NA	$k_{obs} = 0.103$ $R^2 = 0.993$	NA	NA	7	Moussavi et al. (2014)
7	7	Sunlight	TiO2	TiO2	40.9	79.6	NA	$k_{obs} = 0.0042$ $R^2 = 0.985$	NA	NA	5	Gar Alalm et al. (2015)
8	8	UV	ZnO-TiO2	TiO2	86.4	NA	85 (5)	$k_{obs} = 0.1234$ $R^2 = 0.99$	$K = 0.084$ L/mg $k_c = 0.054$ mg/L/min	303.8	8	Jonidi-Jafari et al. (2015)
9	9	UV	WO3	WO3	99.88	NA	100 (5)	$k_{obs} = 0.1233$ $R^2 = 0.9951$	$K = 0.068$ L/mg $k_c = 0.006$ mg/L/min	38.93	8	Mohagheghian et al. (2016)
10	10	UV	ZnO	ZnO	98.92	NA	100 (6)	$k_{obs} = 0.078$ $R^2 = 0.907$	$K = 0.067$ L/mg $k_c = 0.926$ mg/L/min	61.54	8	Shirzad-Siboni et al. (2015)
11	11	Sunlight	TiO2	TiO2	38.2	NA	NA	NA	NA	NA	6	Alalm et al. (2015)
12	12	UV	CNS-ZnO	ZnO	54.9	31.8	NA	$k_{obs} = 0.0072$ $R^2 = 0.9675$	NA	NA	8	Hossaini et al. (2017)
13	13	Vis	TiO2/Fe2O3	TiO2	89.21	NA	NA	NA	NA	NA	7	Mirmasoomi et al. (2017)
14	14	Vis	N-TiO2	TiO2	85	63	80 (7)	NA	NA	NA	9	Salarian et al. (2016)
15	15	UV	Fe-TiO2	TiO2	98.58	NA	NA	NA	NA	NA	8	Baneshi et al. (2017)
16	16	UV	TiO2	TiO2	55	NA	NA	$k_{obs} = 0.01826$ $R^2 > 0.97$	NA	NA	8	Hassan et al. (2017)
17	17	UV	TiO2/AC	TiO2	95	NA	NA	$k_{obs} = 0.03756$ $R^2 > 0.97$	NA	NA	9	Jonidi-Jafari et al. (2017)
18	18	UV	Ni-ZnO	ZnO	99.96	NA	85 (5)	$k_{obs} = 0.0161$ $R^2 = 0.7933$	$K = 0.163$ L/mg $k_c = 0.156$ mg/L/min	350.36	9	Jonidi-Jafari et al. (2017)
19	19	UV	Fe3O4-WO3	WO3	96.4	96.4	96 (5)	$k_{obs} = 0.1008$ $R^2 = 0.9729$	$K = 0.085$ L/mg $k_c = 0.2016$ mg/L/min	47.62	10	Mohagheghian et al. (2017)
20	20	UV	Cu-ZnO	ZnO	96.79	NA	96 (5)	$k_{obs} = 0.0279$ $R^2 = 0.9607$	$K = 0.176$ L/mg $k_c = 0.217$ mg/L/min	303.8	10	Shirzad-Siboni et al. (2017)
21	21	UV	TiO2	TiO2	75	NA	NA	NA	NA	NA	5	Usman et al. (2017)
22	22	UV	Fe-TiO2	TiO2	75	NA	NA	NA	NA	NA	8	Tabasideh et al. (2017)

Table 1 (continued)

Studies No	Trials No	Light source	Catalyst name	Base catalyst ¹	Maximum degradation rate (%)	Maximum mineralization rate (%)	Reusability ² (degradation rate % (cycles))	k_{obs} (min ⁻¹) ³	L-H model parameters	E_{EO} (kWh/m ³) ⁴	Quality score (out of 10)	Study
22	23	UV	CeVO4	VO4	78.38	NA	NA	$k_{obs}=0.0191$ $R^2=UN$	NA	NA	7	Jansanthea et al. (2018)
23	24	UV	TiO2/Fe3O4	TiO2	94.15	99.8	NA	NA	NA	NA	8	Toolabi et al. (2019)
24	25	Vis	B/TiO2-SiO2/CoFe2O4	TiO2	100	100	96 (3)	$k_{obs}=0.0242$ $R^2=0.82$	NA	NA	8	Zangeneh et al. (2018)
25	26	Vis	N-TiO2/Graphene/Au	TiO2	32.4	NA	NA	NA	NA	NA	9	Ayoubi-Feiz et al. (2019)
26	27	Vis	N-TiO2/Graphene/Ag	TiO2	30	NA	NA	NA	NA	NA	NA	Ayoubi-Feiz et al. (2018)
27	28	Vis	N-TiO2/graphene	TiO2	28.3	NA	NA	NA	NA	NA	9	Ayoubi-Feiz et al. (2018)
28	29	UV	MgO	MgO	99.46	NA	NA	$k_{obs}=0.0237$ $R^2=0.9864$	NA	NA	8	Ahmadifard et al. (2019)
29	30	UV	g-C3N4/Fe3O4/Ag	Polymers	100	82.09	NA	$k_{obs}=0.067$ $R^2=0.9982$	NA	NA	6	Ghods et al. (2020)
30	31	Vis	Fe3O4-COOH@ZIF-8/Ag/Ag3PO4	MOF	99.7	NA	97.3 (5)	NA	NA	NA	7	Joubani et al. (2020)
31	32	UV	WO3-ZnO	ZnO	87	NA	NA	$k_{obs}=0.0205$ $R^2=0.98$	$K=0.023$ L/mg $k_c=0.67$ mg/L/min	112	8	Maleki et al. (2020)
32	33	Sunlight	WO3-ZnO	ZnO	83	NA	NA	NA	NA	NA	7	Molla et al. (2019)
33	34	Sunlight	TiO2	TiO2	100	98	NA	$k_{obs}=0.068$ $R^2=0.97$	NA	NA	5	Phuong et al. (2019)
34	35	Vis	Fe-TiO2/Bent-Fe	TiO2	80.6	NA	69.3 (2)	NA	NA	NA	6	Rahimi-Nasrabadi et al. (2019)
35	36	Vis	Co2TiO4/CofTiO3/Poly-amine	TiO2	98.3	NA	NA	NA	NA	NA	9	Rezaei et al. (2019)
36	37	UV	ZnO-SiO2-Fe3O4	ZnO	11.2	NA	NA	NA	NA	NA	7	Sheydaei et al. (2019)
37	38	Vis	N-TiO2/Ag/Ti	TiO2	30	NA	NA	NA	NA	NA	8	Mohammadzadeh Kakhki and Ahsani (2020)
38	39	Vis	Cd3OSO4	chalcogenide	90	NA	NA	$k_{obs}=0.0115$ $R^2=0.95$	NA	NA	6	Khoiriah et al. (2020b)
39	40	Vis	C,N-TiO2	C,N-TiO2	86.93	96	NA	$k_{obs}=0.0646$ $R^2=0.9895$	NA	NA	NA	NA
40	41	Vis	TiO2	TiO2	56.3	NA	NA	NA	NA	NA	NA	NA

Table 1 (continued)

Studies No	Trials No	Light source	Catalyst name	Base catalyst ¹	Maximum degradation rate (%)	Maximum mineralization rate (%)	Reusability ² [(degradation rate % (cycles))]	k_{obs} (min ⁻¹) ³	L–H model parameters	E_{EO} (kWh/m ³) ⁴	Quality score (out of 10)	Study
38	42	UV	C,N-TiO2	TiO2	76	37	NA	NA	NA	NA	7	Khoiriah et al. (2020a)
	43	UV	TiO2	TiO2	22	NA	NA	NA	NA	NA		
	44	UV	C-TiO2	TiO2	35	NA	NA	NA	NA	NA		
	45	UV	N-TiO2	TiO2	38	NA	NA	NA	NA	NA		
39	46	UV	ZnO	ZnO	95.4	NA	NA	NA	NA	NA	10	Khaghani and Zare (2020)
40	47	UV	GO-ZnO-Mn	ZnO	85	NA	NA	NA	NA	NA	7	Mohammadia et al. (2020)
	48	Vis	GO-ZnO-Mn	ZnO	35	NA	NA	NA	NA	NA		
41	49	UV	CNTs/MOF	MOF	99.7	NA	92.1 (5)	$k_{obs}=0.127$ $R^2=0.99$	NA	NA	9	Samy et al. (2021)

¹The base catalyst is the material that is purely used as the catalyst or undergoes manipulation to make the doped or composite catalyst

²Reusability data show the degradation rate of diazinon in the presence the catalyst is used in several cycles (in parenthesis)

³The constant rate based on the first-order kinetics model (l/min)

⁴The number of kWh of electrical energy needed for one order of magnitude reduction in diazinon concentration in one cubic meter of the solution

NA, data not available; MOF, metal–organic framework; Vis, visible light; Sun, sunlight; UV, ultraviolet; E_{EO} , electrical energy per order; L–H model, Langmuir–Hinshelwood model

mineralization rate, and energy consumption. We found that 21/49 (42.8%) trials claimed maximum diazinon removal rates of above 90% and 8/13 (61.5%) trials reported maximum mineralization rates of above 75%. Concerning the reusability of the photocatalyst, a vast majority of the studies reported no significant change in the efficacy of the photocatalyst for diazinon removal after 2 to 7 cycles of use.

Efficacy of photocatalytic removal of diazinon

The removal efficiency was the most intended outcome for researchers to seek in studies of photocatalytic removal of diazinon ($n=2427$ data). Hassan and Elhadidy et al. (2017) proposed the following reactions for the degradation of diazinon in the presence of TiO_2 :

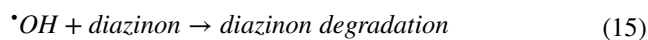
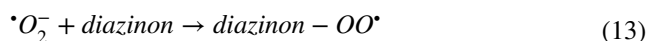
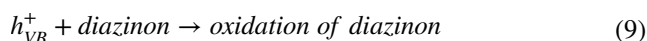
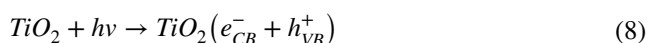


Figure 2A shows a box plot summarizing the efficacy of various types of photocatalytic processes for diazinon abatement. As can be seen, overall, the mean \pm SD of the degradation rate was $59.17 \pm 28.03\%$, with a median and IQR of 60% and 48.73%, respectively. Besides, 25% of the removal percentages were above 84.73% (Q3 value), and 10% of them were above 96% (90th percentile). Based on the process, the highest mean removal efficiency was obtained for MgO/UV ($94.4 \pm 3.08\%$), followed by MOF/UV ($84.3 \pm 15.9\%$), and the least removal efficiency was attained for ZnO/visible light ($14.9 \pm 8.6\%$). However, these results cannot be conclusive, as they were obtained from limited studies and/or trials, as shown in Supplementary File Table S2, and could be the subject of bias. Among the most widely used processes (those used in at least two studies), the highest mean removal rate belonged to ZnO/UV with $62.6 \pm 26.01\%$, followed by TiO_2 /visible light with $59.8 \pm 23.7\%$ and TiO_2 /UV with $54.8 \pm 26.9\%$. However, these three processes were adequate to bear maximum removal rates of 100%, most probably depending on operational variables. Figure 2B shows the histogram of the diazinon degradation rate above the third percentile, which is 84.73%, for various photocatalytic processes. This histogram depicts that ZnO/UV, WO_3 /UV, TiO_2 /UV, and TiO_2 /Vis, in sequence, are probably the most efficient photocatalytic processes for diazinon removal

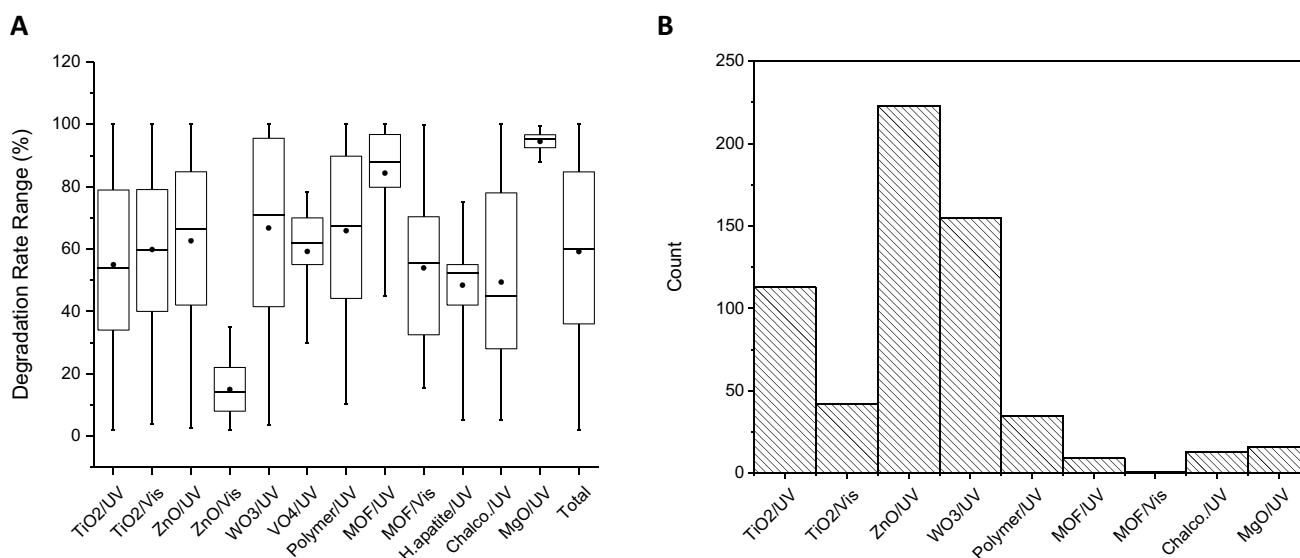


Fig. 2 **A** Box plot of variations of degradation rate in experimental studies of photocatalytic removal of diazinon, showing minimum, Q1, median, Q3, maximum, and mean (●) values; **B** histogram of degradation rates above the third percentile (84.73%) based on various

photocatalytic processes, showing ZnO/UV, WO_3 /UV, TiO_2 /UV, and TiO_2 /Vis as the best processes, in sequence, applied for diazinon abatement

because they are more likely to generate degradation rates of above 84.73%.

The mineralization rate is usually determined by measuring total organic carbon (TOC) in the treated sample. A 100% mineralization rate indicates no organic carbon content in the treated effluent. The mineralization reaction of diazinon in the presence of oxygen as the oxidizing agent is proposed as follows (Nakaoka et al. 2010):



In photocatalytic processes, the complete oxidation of diazinon is achieved, if any, at very long irradiation durations, which seems economically undesirable. We found only two processes for which the mineralization rates were reported ($n=124$ data) (Supplementary File Table S2). The mean mineralization rate was $47.5 \pm 27.5\%$ for TiO_2/UV and $60.6 \pm 28.2\%$ for TiO_2/Vis . Pure TiO_2 is a semiconductor with a wide bandgap of 3.2 eV, which is only activated with the energy of photons from UV light (Drygała 2021). However, modified TiO_2 may have a narrower bandgap, with the possibility of being activated with lower energies in the spectrum of visible light. This may also have led to its higher ability to mineralize diazinon. However, one should note that heterogeneities between subgroups in this study, e.g., concerning initial diazinon concentration, may explain some differences in the obtained results.

Operational parameters

Solution pH

Initial solution pH is an influential factor that affects the performance of the photocatalytic degradation of diazinon. Figure 3A summarizes the variations of solution pH based on the photocatalytic process applied in various studies for

assessing the effect of pH on diazinon removal. As can be seen, pH varied in a relatively constant range in different photocatalytic processes. As a whole, the pH varied from 2 to 12, with a mean and SD of 6.86 and 2.69 and a median and IQR of 7 and 4, respectively. However, 99% of the solution pH values were adjusted below 11 (Supplementary File Table S3). Concerning diazinon removal efficiency, the optimal pH was obtained in the neutral condition in most studies (Hossaini et al. 2014, 2017; Moussavi et al. 2014; Jonidi-Jafari et al. 2015; Shirzad-Siboni et al. 2015; Salarian et al. 2016; Baneshi et al. 2017; Jonidi-Jafari et al. 2017; Shirzad-Siboni et al. 2017, Ahmadifard, Heydari et al. 2019, Ghodsi, Esrafilii et al. 2020, Maleki, Moradi et al. 2020), an acidic condition in some cases (Nakaoka et al. 2010, Yang et al. 2010, Gar Alalm et al. 2015, Jafari et al. 2016, Mohagheghian et al. 2016, Zangeneh et al. 2018, Phuong, Chu et al. 2019, Rahimi-Nasrabadi, Ghaderi et al. 2019, Khaghani and Zare 2020), and alkaline condition in a few

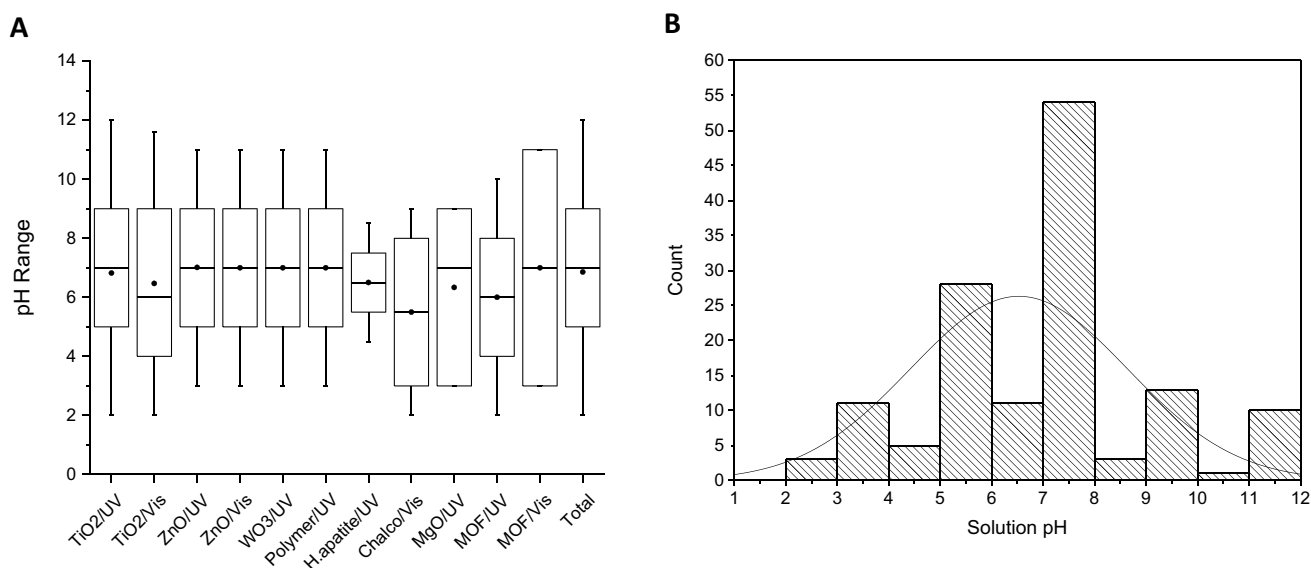


Fig. 3 **A** Box plot of variations of solution pH in experimental studies of photocatalytic removal of diazinon, showing minimum, Q1, median, Q3, maximum, and mean (●) values; **B** histogram of applied

solution pH in photocatalytic experiments ($n=139$) giving a diazinon degradation rate above the third percentile (> 74%), showing pH 5.0–8.0 as the optimum pH

studies (Fadaei and Dehghani 2012, Molla, Furukawa et al. 2019). This could also be seen in Fig. 3B, which depicts the histogram of pH for diazinon removal efficiencies above the third percentile (74%) ($n = 139$). As seen, in about 69% ($n = 96/139$) of the cases, a removal of > 74% occurred in a pH range of 5–8, while pHs between 8 and 12 accounted for around 19% of the high removal percentages.

The variation in removal efficiency in different pH values is mostly related to pKa of diazinon, pH_{Zpc} of the used catalyst, and other operational conditions. As the pKa of diazinon is 2.6, the functional groups on the surface of diazinon are negatively and positively charged above and below pH 2.6, respectively. On the other hand, the pH_{Zpc} of the catalyst plays an important role in the status of the surface charge of the catalyst. It is expected that at pHs below pH_{Zpc}, the surface of the catalyst is negatively charged and vice versa. Therefore, considering the pKa of diazinon and the pH_{Zpc} of the catalyst, it is believed that the best removal efficiency happens between the pH_{Zpc} of the catalyst and pKa of diazinon (Gar Alalm et al. 2015, Jafari et al. 2016, Hossaini et al. 2017, Ahmadifard, Heydari et al. 2019, Khoiriah et al. 2020a, b). This improvement occurs because of the increase in electrostatic attraction between diazinon molecules and active sites of the catalyst due to a rise in attractive forces within this pH range (Gar Alalm et al. 2015, Ghodsi, Esrafilii et al. 2020). This accelerates the adsorption of diazinon on the catalyst surface, which is necessary for the direct oxidation of diazinon. As much as electron–hole pairs at the surface of the catalyst recombine in a shorter time, this mechanism will be more essential for the degradation of diazinon (Jafari et al. 2016, Ahmadifard, Heydari et al. 2019). On the other hand, it is shown that by increasing pH from neutral to alkaline values, a decreasing trend is observed in diazinon removal percentages (Hossaini et al. 2014, Jonidi-Jafari et al. 2015, Jafari et al. 2016, Mohagheghian et al. 2016, Baneshi et al. 2017, Hossaini et al. 2017, Jonidi-Jafari et al. 2017, Ghodsi, Esrafilii et al. 2020, Khoiriah et al. 2020a, b). In some cases, this may be attributed to the homogeneity of charges on the catalyst surface and diazinon molecules (both with negative charges) above the pH_{Zpc} of the catalyst. As a result, a repulsive force is generated, and the removal efficiency of diazinon decreases (Gar Alalm et al. 2015; Jonidi-Jafari et al. 2015, Ghodsi, Esrafilii et al. 2020).

Mohagheghian et al. investigated the removal efficiency of diazinon using WO₃/UV. It was shown that by increasing pH from 3 to 11, the diazinon removal decreased from 99.88 to 19.8%. They believed that this decreasing trend could be related to the pH_{Zpc} of WO₃ that is negative in alkaline conditions, so the adsorption of diazinon on the catalyst surface was prevented (Mohagheghian et al. 2016). Nakaoka et al. investigated the photodegradation of diazinon by platinumized TiO₂ at pH values 2–11. They reported that by increasing

pH up to 7, there was a growing trend in removal efficiency. However, the trend sharply dropped after neutral pH up to pH 11. Therefore, they postulated that OH⁻ ions increased by increasing pH, so enhancing the production of •OH free radicals through the interaction between OH⁻ ions and photo-generated holes. Therefore, the competition between diazinon and hydroxyl radicals for the surface sites of the catalyst causes a decrease in the removal efficiency in alkaline pH values (Nakaoka et al. 2010).

Catalyst dose

The photocatalyst is one of the most determining constituents of photocatalytic processes. What can make a photocatalyst preferable to another includes parameters such as suitable distribution of particle sizes, high surface areas, high chemical stability, and ability to absorb sunlight/UV. There are different methods and materials used for photocatalyst preparation. Semiconductors that can act as photocatalysts can be made in single, composite, modified, or doped forms (Moussavi et al. 2014, Hossaini et al. 2017, Ahmadifard, Heydari et al. 2019, Ayoubi-Feiz et al. 2019, Ghodsi, Esrafilii et al. 2020, Joubani et al. 2020). One of the most widely used photocatalysts for diazinon removal is TiO₂. It is widely used either in the single, doped, or binary forms (Nakaoka et al. 2010; Hossaini et al. 2014; Gar Alalm et al. 2015; Jonidi-Jafari et al. 2015; Jafari et al. 2016; Baneshi et al. 2017; Hassan et al. 2017; Mirmasoomi et al. 2017; Usman et al. 2017; Ayoubi-Feiz et al. 2018, Sheydaei, Karimi et al. 2019). Another commonly used photocatalyst for diazinon removal is ZnO (Fadaei and Dehghani 2012, Moussavi et al. 2014, Shirzad-Siboni et al. 2015, Hossaini et al. 2017, Jonidi-Jafari et al. 2017, Shirzad-Siboni et al. 2017, Rezaei, Dehghanifard et al. 2019, Maleki, Moradi et al. 2020, Mohammadia et al. 2020). Moreover, some nanostructured composite photocatalysts have been recently used for diazinon removal (Yang et al. 2010, Mohagheghian, Ayagh et al. 2017, Jansanthea et al. 2018, Zangeneh et al. 2018, Rahimi-Nasrabadi, Ghaderi et al. 2019, Ghodsi, Esrafilii et al. 2020).

The examined studies utilized catalysts at different doses ranging from 1 to 6,000 mg/L. Based on Fig. 4A, the mean ± SD in the whole experiment was 578.7 ± 765.7 mg/L, with a median and IQR of 250 and 700 mg/L, respectively. Besides, 95% of the experiments were conducted in the presence of a 2000 mg/L catalyst dose (Supplementary File Table S4). Concerning the effect of the catalyst dose, the results showed that the removal efficiency of diazinon is enhanced by increasing the catalyst dose. This can be due to enhancing the available surface area, which provides more surfaces for diazinon molecules to be adsorbed. As a result, by enhancing available active sites, the chance of electron–hole pair formation increases, so more reactive radicals will be produced in the solution. However, the extra use

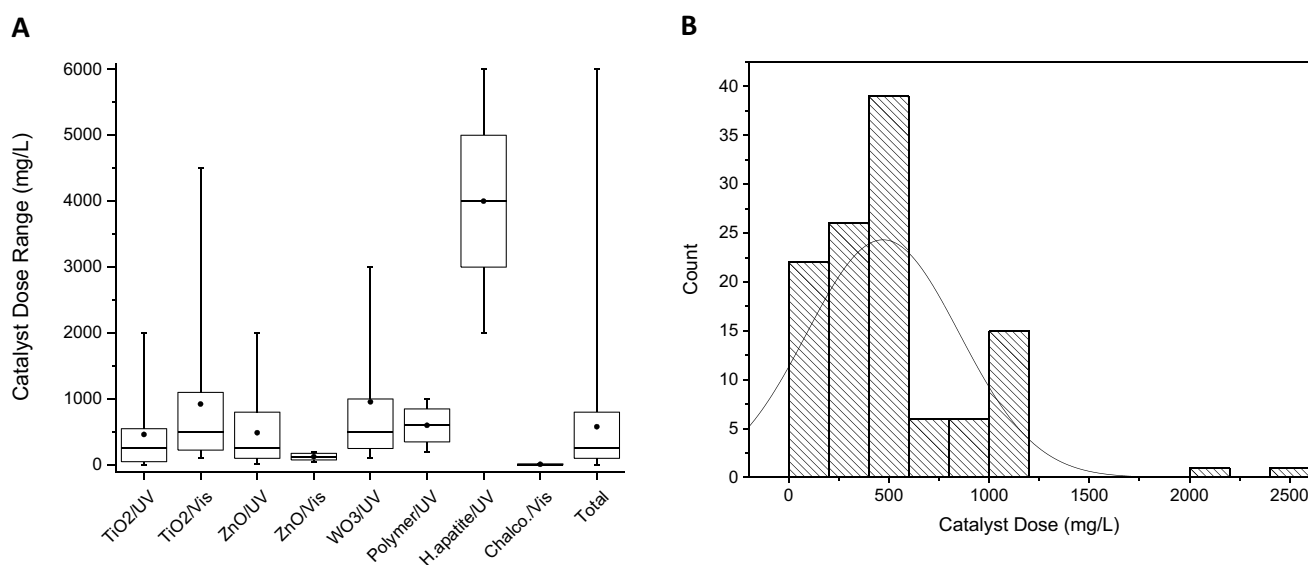


Fig. 4 **A** Box plot of variations of catalyst dose in experimental studies of photocatalytic removal of diazinon, showing minimum, Q1, median, Q3, maximum, and mean (●) values; **B** histogram of

applied catalyst dose in photocatalytic experiments ($n=116$) giving a diazinon degradation rate above the third percentile ($>78\%$), showing doses <600 mg/L as the optimum catalyst dose

of the photocatalyst can grow the turbidity of the solution, which, in turn, can inhibit the light penetration or enhance light scattering. Furthermore, the increasing dose can cause agglomeration of particles and consequently decrease the available active sites of the photocatalyst (Yang et al. 2010, Jonidi-Jafari et al. 2015, Shirzad-Siboni et al. 2015, Jafari et al. 2016, Salarian et al. 2016, Shirzad-Siboni et al. 2017, Zangeneh et al. 2018, Molla, Furukawa et al. 2019, Phuong, Chu et al. 2019, Toolabi et al. 2019, Ghodsi, Esrafilii et al. 2020, Khoiriah et al. 2020a, b). In research conducted by Salarian and Hami et al. (2016), nitrogen-doped TiO_2 nanosheets were produced through the hydrothermal method and used for diazinon removal under solar radiation. It was observed that when the amount of N- TiO_2 increased up to 2.7 g/L, the degradation of diazinon was accelerated, and after that, a decreasing trend appeared. They stated that the reason for the enhanced degradation of diazinon could be due to the highly available active sites on TiO_2 , increased absorption of light photons on active sites, and high penetration of light into the solution. However, they inferred that the subsequent falling trend in removal efficiency was because of the enhanced turbidity of the solution induced by overdosing the catalyst and consequently light scattering. Figure 4B also provides information in favor of these explanations. The figure shows the histogram of the catalyst dose for diazinon removal efficiencies above the third percentile of 78% ($n=116$). Notably, the majority of the high removal rates (86/116; 74.13%) were obtained approximately at doses below 600 mg/L, and the minority of them could be efficient at doses above this value. This confirms that (1) increasing catalyst dose can lead to increased removal only in a certain

range and (2) photocatalytic processes are very efficient even at low catalyst doses although this largely depends on the type of the catalyst and its structural and chemical quality to be efficiently activated at the presence of light.

Initial diazinon concentration

In the examined studies, the range of initial concentration of diazinon varied widely from 1 to 500 mg/L, with a mean \pm SD of 32.4 ± 35.84 mg/L and median and IQR of 20 and 30 mg/L, respectively (Fig. 5A). However, 99% of the initial concentrations adjusted were equal to or below 100 mg/L (Supplementary File Table S5). Although these high concentrations are very less likely to occur in natural waters or even industrial wastewaters, in experimental studies, it is necessary to change the levels of diazinon concentration in an adequately wide range. Thus, one can elucidate the efficacy of the treatment in response to varying concentrations of the pollutant. Besides, wastewaters usually contain a mixture of various organic pollutants, summing up to a few tens of milligram per liter concentrations, which may compete for $\bullet\text{OH}$ that act unspecific to the target pollutants. One way for simulating these high concentrations is to add to the diazinon concentration in the experiments. Taking a glance at Supplementary File Table S5, it is seen that the mode concentration of diazinon was 10 mg/L, implying that researchers tended to keep a balance between real and experimental conditions.

Analyzing the results of different studies, an increase in the initial concentration of diazinon led to a decrease in removal efficiency. Besides, Fig. 5B shows the distribution

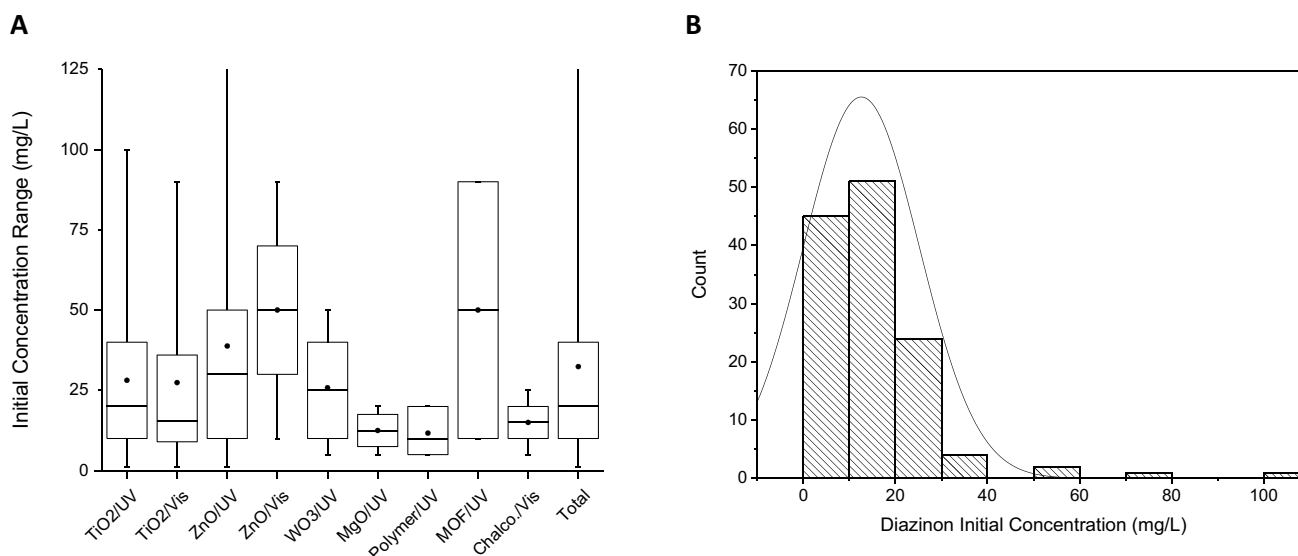


Fig. 5 **A** Box plot of variations of diazinon initial concentration (mg/L) in experimental studies of photocatalytic removal of diazinon, showing minimum, Q1, median, Q3, maximum, and mean (●) values; **B** histogram of applied diazinon initial concentration in photo-

catalytic experiments ($n=128$) giving a diazinon degradation rate above the third percentile $>85.5\%$, showing diazinon initial concentration <40 mg/L as optimum

of initial concentrations for removal efficiencies above the third percentile of 85.5% ($n=128$). As can be seen, almost all of the high removal percentages ($124/128$; 96.87%) were observed at the presence of low concentrations of diazinon (≤ 40 mg/L), which is also indicative of the inverse relationship between diazinon removal and its initial concentration. This can be related to the more diazinon molecules adsorbed on the catalyst surface at higher concentrations; thus, the active sites on the catalyst and consequently the production of reactive radicals would be limited. Moreover, the high density of diazinon molecules in the solution can inhibit light photons to reach the catalyst surface. Maleki and Moradi et al. (2020) investigated the effects of tungsten oxide-doped zinc oxide nanoparticles immobilized on glass substrates as a catalyst on photocatalytic removal of diazinon. The results showed that after 60 min, the removal efficiency reduced from 88.6 to 44.4% by raising the concentration of diazinon from 10 to 200 mg/L (Maleki, Moradi et al. 2020). Jafari and Moussavi et al. (2016) conducted the study of the degradation of diazinon using UVC and UVC/TiO₂ processes. In this study, they investigated the effect of three different initial concentrations of diazinon on the photocatalytic process (5, 10, and 20 mg/L) at a constant dose of TiO₂ (100 mg/L) and pH = 7. The results showed that the highest removal rate (especially at initial contact times) was obtained at the initial concentration of 5 mg/L, and after 40 min, the complete degradation occurred (Jafari et al. 2016). Fadaei and Dehghani (2012) investigated the removal of two organophosphorus pesticides (diazinon and malathion) using zinc oxide as the catalyst. To determine

the effect of the initial concentration of diazinon on the photocatalytic process, the initial concentrations of 100, 300, and 500 $\mu\text{g/L}$ were tested. They reported that by increasing initial concentration from 100 to 500 $\mu\text{g/L}$, the degradation rate dropped from 69.88 to 55.2%. They attributed this reduced degradation rate to the (1) formation and adsorption of degradation by-products on the catalyst surface, which ended in occupying active sites on the catalyst surface, (2) decreased rate of light absorption due to increased adsorption of diazinon molecules on the catalyst surface, (3) enhancement of solution turbidity through the increase of diazinon concentration which inhibited light photons to penetrate the solution, (4) absorption of light photons in the path length by diazinon molecules rather than the catalyst, which could reduce catalytic efficiency, and (5) adsorption of diazinon molecules, instead of OH⁻ ions, on the catalyst surface, thereby reducing the generation of •OH radicals and the degradation rate, as well (Fadaei and Dehghani 2012).

Contact time

In the reviewed studies, the contact time varied between 3 and 360 min (Fig. 6A). The mean \pm SD in overall experiments was 67.4 ± 47.04 min, with a median and IQR of 60 and 60 min, respectively. However, 90% of the experiments were conducted at contact times below 120 min (Supplementary File Table S6), indicating the photocatalytic process acts relatively fast for diazinon removal. Figure 6B provides more information about the relationship of contact time in the photocatalytic process with the diazinon removal rate. As

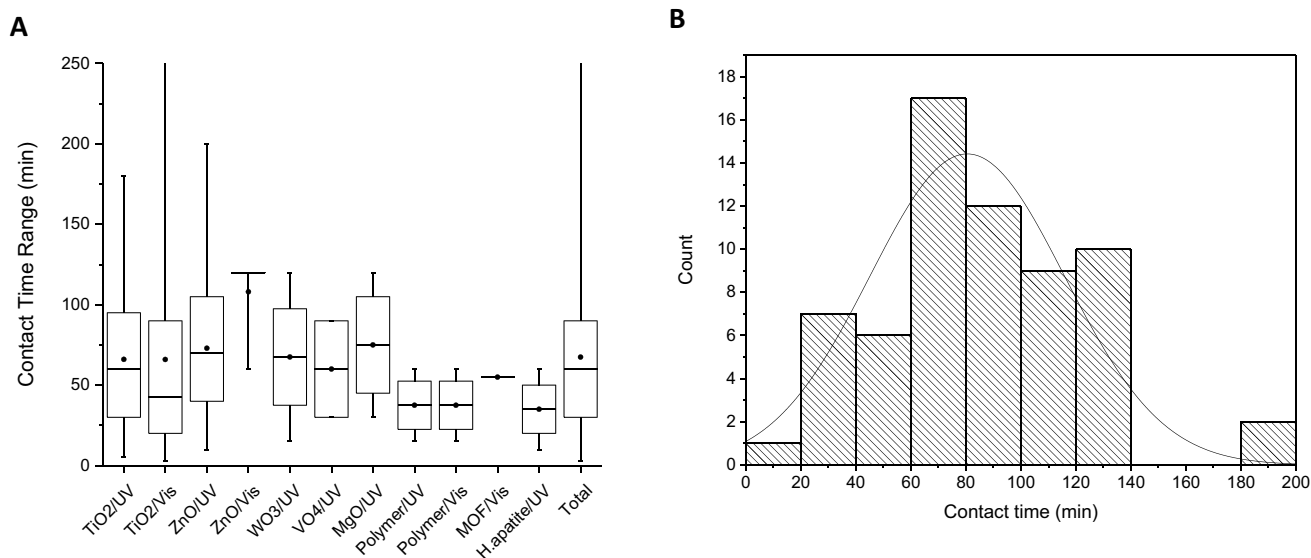


Fig. 6 **A** Box plot of variations of contact time (min) in experimental studies of photocatalytic removal of diazinon, showing minimum, Q1, median, Q3, maximum, and mean (●) values; **B** histogram of applied

contact time in photocatalytic experiments ($n=64$) giving a diazinon degradation rate above the third percentile (>85%), showing contact time of 20–140 min as optimum

can be seen, removal efficiencies above the third percentile of 85% ($n=64$) were more likely to obtain at contact times from 20 to 140 min (61/64; 95.3%). However, this finding could be biased due to the effect of other operational parameters, especially diazinon initial concentration.

As known, when the contact time increases, more electrons will be excited at the catalyst surface, so increasing the chance of reactive radical production. This increase in **contact time** can enhance the possibility of collision between diazinon molecules and reactive radicals, leading to improved degradation and efficiency (Toolabi et al. 2019). In some cases, this increase in removal rate by increasing the contact time stops in a certain period, and after that, there is a constant trend or slight increase in the decomposition rate (Baneshi et al. 2017; Toolabi et al. 2019, Ghodsi, Esrafilii et al. 2020). In a study conducted by Baneshi and Rezaei et al. (2017) on the photocatalytic degradation of diazinon by Fe-doped TiO_2 , the effect of contact time was investigated from 5 to 90 min. The results showed an increasing trend in diazinon removal up to 60 min (98.57%), and after that, the removal rate was negligible; so 60 min contact time was assigned as optimum in the study (Baneshi et al. 2017). Molla and Furukawa et al. (2019) investigated the effect of irradiation time on diazinon removal using nanosized-photocatalyst TiO_2 in water under sunlight irradiation. The results show a sharp increase in removal efficiency up to 10 min. However, the complete decomposition occurred at 60 min (Molla, Furukawa et al. 2019). Hossaini and Moussavi et al. (2017) conducted a study on diazinon removal through CNS-ZnO/LED photocatalytic process. It was observed that by increasing contact time to 100 min, diazinon decomposition

increased to 53.8%. However, after 100 min up to 200 min, this increase became slow and degradation reached 54.9%. Therefore, in this study, the optimum contact time for diazinon removal was determined as 100 min (Hossaini et al. 2017).

Kinetic studies

Zero-, first-, and second-order kinetic models have been utilized for assessing the kinetics of diazinon removal in heterogeneous photocatalytic systems. The linear forms of the equations for zero-, first-, and second-order kinetic models are shown in Eqs. (17) to (19):

$$C_t = C_0 - k_0 t \quad (17)$$

$$\ln \frac{C_0}{C_t} = k_{\text{obs}} t \quad (18)$$

$$\frac{1}{C_t} = \frac{1}{C_0} + k_2 t \quad (19)$$

In these equations, C_0 is the initial diazinon concentration (mg/L), C_t is the diazinon concentration at time t (mg/L), t is the reaction time (min), k_0 is the zero-order reaction constant (mg/L/min), k_{obs} is the first-order reaction constant (1/min), and k_2 is the second-order reaction constant (L/mg/min). Kinetic studies are usually conducted under fixed (optimum) conditions of initial diazinon concentration, solution pH, catalyst dose, etc. but the reaction time varies up to a few hours. Finally, the experimental data are fitted with

the kinetic models to find the best fitting model. To do so, plots are drawn with C_t against t for the zero-order model, $\ln C_0/C_t$ against t for the first-order model, and $1/C_t$ against t for the second-order model. The plot with the highest R^2 value (closest to 1) shows the best describing model, and the intercept and the slope of the plot are utilized to calculate the rate constant.

Herein, we found 23 studies that investigated the kinetics of photocatalytic diazinon abatement. Of them, only six studies (Jonidi-Jafari et al. 2015, Jonidi-Jafari et al. 2017, Mohagheghian, Ayagh et al. 2017, Shirzad-Siboni et al. 2017, Ghodsi, Esrafilii et al. 2020, Khoiriah et al. 2020a, 2020b) tried all the three kinetic states in order to prove the kinetics of the reaction, all of which showed that the first-order kinetic model best explains the experimental results. The other studies ($n=17$ studies) only tried the first-order model presumably based on the assumption that diazinon degradation at low concentrations under photocatalytic conditions follows the first-order reaction laws (Jafari et al. 2016). Based on Table 1, the highest reaction rates were 1.86 min^{-1} (Fadaei and Dehghani 2012), 1.7 min^{-1} (Nakaoka et al. 2010), 0.127 min^{-1} (Samy, Ibrahim et al. 2021), 0.1234 min^{-1} (Jonidi-Jafari et al. 2015), and 0.1233 min^{-1} (Mohagheghian et al. 2016), in sequence, which seems to be inversely related to the reaction time in the kinetic experiments (3 min, 30 min, 60 min, 120 min, and 120 min, respectively).

The Langmuir–Hinshelwood (L–H) model has been developed to describe the heterogeneous photocatalytic systems in which the substrate (herein diazinon) is adsorbed on the photocatalyst following a Langmuir isotherm, and the reaction takes place based on the rules of the first-order model (Ohtani 2011). Besides, the adoption equilibrium state should remain during photocatalytic degradation. In such a system, the rate of adsorption is always faster than the rate of degradation, and the photo-absorption process limits the rate of substrate abatement. The following equation is used to express the L–H model (Shirzad-Siboni et al. 2015):

$$-\frac{dC}{dt} = \frac{k_c K C}{1 + K C_0} = k_{obs} C \quad (20)$$

where k_c is the reaction rate constant of the surface-adsorbed substrate with electron–holes (mg/L/min) and K is the adsorption equilibrium constant (L/mg). To obtain the parameters, this equation can also be written as follows (Mohagheghian, Ayagh et al. 2017):

$$\frac{1}{k_{obs}} = \frac{1}{k_c K} + \frac{C_0}{k_c} \quad (21)$$

By plotting reciprocal k_{obs} (obtained from the first-order reaction at different initial diazinon concentrations) against initial diazinon concentration, one can calculate K and k_c

values from the intercept and the slope of the plot, respectively. This was done by seven studies (Jonidi-Jafari et al. 2015, Shirzad-Siboni et al. 2015, Mohagheghian et al. 2016, Jonidi-Jafari et al. 2017, Mohagheghian, Ayagh et al. 2017, Shirzad-Siboni et al. 2017, Maleki, Moradi et al. 2020), which reported K values from 0.023 (Maleki, Moradi et al. 2020) to 0.176 L/mg (Shirzad-Siboni et al. 2017) and k_c values ranging from 0.006 (Mohagheghian et al. 2016) to 0.926 mg/L/min (Shirzad-Siboni et al. 2015).

Energy consumption

Energy consumption is among various factors that can affect the selection of photocatalytic processes, as it affects the cost-effectiveness of the process. In the examined studies, the energy consumption was expressed as the number of kWh of electrical energy needed for one order of magnitude reduction (i.e., 90% removal) in diazinon concentration in 1 m^3 of the solution, known as electrical energy per order (E_{EO}). Therefore, it is directly related to the power of the light source and the duration of irradiation to reach a 90% removal rate and is applied only for artificial light sources. For this purpose, k_{obs} from the first-order kinetic model can be utilized to compute the E_{EO} values. Seven studies (Jonidi-Jafari et al. 2015, Shirzad-Siboni et al. 2015, Mohagheghian et al. 2016, Jonidi-Jafari et al. 2017, Mohagheghian, Ayagh et al. 2017, Shirzad-Siboni et al. 2017, Maleki, Moradi et al. 2020) used the following equation to calculate the E_{EO} values as kWh/m³/order (Maleki, Moradi et al. 2020):

$$E_{EO} = \frac{38.4 \times P}{V \times k_{obs}} \quad (22)$$

where P is the power of the light source (kW) and V is the solution volume (L). According to Table 1, the E_{EO} was between 38.93 (Mohagheghian et al. 2016) and 350.36 kWh/m³ (Jonidi-Jafari et al. 2017). It seems that the lowest E_{EO} values correspond to lower initial diazinon concentrations, as such solutions need less power and time to reach a 90% removal rate.

By-products

The formation of by-products is always one of the most challenging aspects of photocatalytic processes. Since photocatalytic processes are used to decompose toxic pollutants, it is expected that the effluent has less toxicity than the influent. However, it is not always true. Seven studies (Nakaoka et al. 2010, Jonidi-Jafari et al. 2017, Shirzad-Siboni et al. 2017, Ahmadifard, Heydari et al. 2019, Toolabi et al. 2019, Khaghani and Zare 2020, Samy, Ibrahim et al. 2021) investigated the formation of by-products of diazinon

in photocatalytic degradation. The identified by-products are listed in Table S7.

The by-products produced at different reaction times may show the degradation pathway of the original compound. In photocatalytic processes, there are different pathways and consequently different intermediates for diazinon degradation depending on the experimental condition (Khaghani and Zare 2020). Diazoxon is mainly produced due to the desulfuration of diazinon via the substitution of oxygen by sulfur in the P=S bond through $\bullet\text{OH}$ radical attack or by direct diazinon oxidation. In some studies, diazoxon is reported as a more toxic compound than diazinon (Ahmadifard, Heydari et al. 2019, Toolabi et al. 2019). Another common intermediate is 2-isopropyl-6-methyl-4-pyrimidinol (IMP), which is suggested to be produced through the break of the pyrimidine ring (P-O) in diazinon, C-O bond in diazoxon, or even via the hydrolysis of diazinon. This compound is reported to be less toxic than primary diazinon (Čolović et al. 2010; Nakaoka et al. 2010, Ahmadifard, Heydari et al. 2019, Molla, Furukawa et al. 2019). The hydroxylation of the propyl group in diazinon is another phenomenon, which can cause further by-products such as hydroxydiazinon, 2-hydroxydiazinon, 1-hydroxy isopropyl diazinon, and O,O-diethyl-O-(2-isopropyl-6-ethyl-5-hydroxypyrimidine-4-yl) (Nakaoka et al. 2010, Ahmadifard, Heydari et al. 2019, Molla, Furukawa et al. 2019, Khaghani and Zare 2020). These intermediates can turn into diazinon aldehyde and diazinon methyl ketone through further oxidation (Nakaoka et al. 2010). Besides, the oxidation of the C-O bond of diazinon can yield diethyl thiophosphate (DETP) or triethyl thiophosphate (TETP). The further degradation of these products can lead to triethyl phosphate or diethyl phosphonate production. These two products can also be produced through the hydrolysis of diazoxon. In the next step, triethyl phosphate or diethyl phosphonate can be converted into phosphoric acid and finally, carbon dioxide, phosphate, and water (Ahmadifard, Heydari et al. 2019, Molla, Furukawa et al. 2019, Samy, Ibrahim et al. 2021).

Hossaini and Moussavi et al. (2017) bioassayed the toxicity of by-products of diazinon degradation by *Vibrio fischeri*. Their results showed that the photoluminance of bacteria reduced by 55.3% and 8.9% in the influent and the sample treated by the CNS-ZnO/LED process, respectively, indicating the degradation of diazinon to less toxic compounds (Hossaini et al. 2017). Khaghani and Zare (2020) surveyed the toxicity of malathion and diazinon in a UV/nano-Zn process. They detected diethyl phosphate (DEP), diethyl thiophosphate (DETP), 2-isopropyl-6-methyl-4-pyrimidinol (IMP), hydroxydiazinon, O-analog diazinon (diazoxon), and some other compounds as the by-products of diazinon degradation. Moreover, the results showed that the by-products produced through diazinon decomposition were more in number and toxicity than those produced after malathion

degradation. In this study, the EC_{50} values were 2.24 for *Nitrobacter* and 2.82 mg/L for *Nitrosomonas*, indicating that *Nitrobacter* was more sensitive than *Nitrosomonas* when exposed to photocatalytic by-products (Khaghani and Zare 2020).

One of the limitations of this study was the inclusion of studies conducted merely on diazinon, which limits the generalizability to other pesticides and refractory pollutants. Besides, there was high heterogeneity between studies concerning variables affecting the photocatalytic removal of diazinon, which may have been a source of bias. Despite limitations, this study provided data on the most important operational variables affecting photocatalysis reactions, as well as degradation rates, kinetics, required energy, and by-products in various photocatalytic processes, which may be used in setting up laboratory or even large-scale treatment systems. Future systematic reviews may provide information on non-operational variables and fixed conditions, e.g., reactor volume, mixing intensity, and light source power, as well as effluent toxicity, in the photocatalytic removal of diazinon or other persistent pollutants.

Conclusion

This quantitative systematic review could retrieve 41 studies assessing the photocatalytic removal of diazinon from aqueous solutions in 49 trials. About 63% of the trials were conducted in the UV light spectrum and 37% in the visible light spectrum. Besides, 14 studies utilized pure compounds as photocatalysts, while 27 of them used a modified catalyst. TiO_2 -based materials were the most frequent catalysts ($n=22$ studies), followed by ZnO-based catalysts ($n=10$ studies). All of the studies reported data on the diazinon degradation rate, which ranged from 2 to 100%, with a mean \pm SD of $59.17 \pm 28.03\%$. Based on the process, MgO/UV and MOF/UV were the most efficient processes for diazinon removal, and ZnO/Vis was the least efficient one. However, among the most widely used processes with the highest efficacies were ZnO/UV, WO_3 /UV, TiO_2 /UV, and TiO_2 /Vis, in sequence. Solution pH in the range of 5–8, catalyst dose below 600 mg/L, diazinon initial concentration below 40 mg/L, and contact time between 20 and 140 min could be the optimum conditions for diazinon degradation using photocatalytic oxidation. The degradation of diazinon obeys the first-order kinetic model with k_{obs} between 0.0042 and 1.86 min^{-1} , which depends on reaction time. The consumed energy ranged from 38.93 to 350.36 kWh/m^3 , which varied as a function of initial diazinon concentration. Diazoxon and IMP are the most detected by-products of diazinon degradation, although bioassay data are scarce. Based on the results, photocatalytic processes are very efficient for removing diazinon from aqueous solutions,

although more elaborate studies are needed to assess the mineralization rate and toxicity of the effluent.

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Declarations

Ethics approval This study was approved by the Research Ethics Committee of Faculty of Health, Tehran University of Medical Sciences (IR.TUMS.SPH.REC.1400.196).

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