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*Review*

# A Systematic Review on Solar Heterogeneous Photocatalytic Water Disinfection: Advances over Time, Operation Trends, and Prospects

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**Abstract:** Access to drinking water is a human right recognized by the United Nations. It is estimated that more than 2.1 billion people lack access to drinking water with an adequate microbiological quality, which is associated to 80% of all diseases, as well as with millions of deaths caused by infections, especially in children. Water disinfection technologies need a continuous improvement approach to meet the growing demand caused by population growth and climate change. Heterogeneous photocatalysis with semiconductors, which is an advanced oxidation process, has been proposed as a sustainable technology for water disinfection, as it does not need addition of any chemical substance and it can make use of solar light. Nevertheless, the technology has not been deployed industrially and commercially yet, mainly because of the lack of efficient reactor designs to treat large volumes of water, as most research focus on lab-scale experimentation. Additionally, very few applications are often tested employing actual sunlight. The present work provide a perspective on the operation trends and advances of solar heterogeneous photocatalytic reactors for water disinfection by systematically analyzing pertaining literature that made actual use of sunlight, with only 60 reports found out of the initially 1044 papers detected. These reports were discussed in terms of reactor employed, photocatalyst used, microorganism type, overall disinfection efficiency, and location. General prospects for the progression of the technology are provided as well.

**Keywords:** sunlight; reactor design; advanced oxidation processes; water treatment; water potabilization



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## 1. Introduction

As of 2010, the access to water has been recognized as a human right by the United Nations. Water purposed for personal and domestic use should comply with sufficiency, physical availability, safeness, and affordability [1]. Nevertheless, the World Health Organization (WHO) estimates that 30% of the global population, which accounts for 2.1 billion people, lack access to water sources which meet guidelines for safe drinking water [2]. Water which does not observe those set guidelines cannot be considered as drinking water and its consumption can be hazardous; it is estimated than the intake of unsafe water is at fault for 80% of all of the world diseases [3]. Among these illnesses, infectious diseases which are caused by pathogens, mainly bacteria and virus, are recurrent [4]. Some of them are typhoid, cholera, dysentery, parasitic infections [3] or viral infections [4]. Water-borne diseases can become lethal, especially if patients do not receive medical attention; these diseases cause 1.8 million deaths each year, children being the most vulnerable group. Water quality improvement reduces this morbidity [3].

The WHO has also projected, that by year 2025, half of the world population will live in water-stressed areas. To provide safe drinking water has become one of the biggest challenges for mankind in the present century [5].

Among the WHO guidelines for drinking water, it is stated that no microorganism known to be pathogenic should be contained within water, hence, drinking water should be disinfected to ensure this guideline [6]. Water supply systems may use one or several disinfection technologies in order to ensure no pathogenic microorganisms are present, the selected technology depends on a plethora of factors, such as availability, water initial microbial quality, and final quality intended or needed, cost effectiveness, volume required or even level of automation and local level costs [7]. Table 1 lists some of the most used water disinfection processes, along with some of their advantages and disadvantages.

**Table 1.** Advantages and disadvantages of some of the most used water disinfection technologies.

| Water Disinfection Process | Advantages | Disadvantages |
| --- | --- | --- |
| Chlorination | Low cost; effective at low concentrations; residual effect, widely available [8] | Formation of toxic byproducts; modified taste and odor; ineffective against biofouling; cannot kill parasite eggs [9,10] |
| Ozonation | High biocidal efficacy over a wide antimicrobial spectrum; color, odor, and taste control [11] | Lack of residual effect as ozone is unstable in water; byproducts formation from bromide and natural organic matter [12] |
| Disinfection with colloidal silver | Well-known biocidal and disinfection properties; able to remove organic compounds; lacks the adverse effects of chlorination and ozonation [13] | Relatively high cost and time inversion; loss of effectivity over time [14] |
| Peracetic acid treatment | Low dependence on pH; high sterilization ability; reduced toxic byproducts' formation; easy implementation [15] | Costly activation (UV light or metal catalysis); not proven technical feasibility to inactivate fungi, algae of microorganisms on biofilms; scarce pilot plant applications which limit economic feasibility evaluation [16] |
| Ultraviolet radiation | No chemical addition, reduce disinfection byproduct (DBP) formation, high efficiency in inactivating chlorine-resistant organisms [17] | Energy intensive; unsuitable for places without stable energy supply, lack of a residual effect [18,19] |
| Solar disinfection (SODIS) | Non-energy intensive as it harvests solar energy; effective for several microorganisms; point of use technology [20] | Low efficiency in solar energy conversion; long exposure time; microorganism regrowth might happen if UV exposure is not high enough [21] |

Considering the increasing water stress context and the drawbacks of the known disinfection water technologies, the need to re-design or improve these processes to obtain technologies which are robust, simple to use, chemical-free, and inexpensive arises [22]. Some emerging water disinfection technologies include electrodisinfection [23], water cavitation [24] or heterogeneous photocatalysis (HP) with semiconductors, also known as photocatalysts (PC).

HP is an advanced oxidation process (AOP) which was first reported in 1972 when water splitting was observed on the surface of a titanium electrode, namely, over titanium dioxide ($TiO_2$) due to the effect of light irradiation [25]. When a PC is exposed to radiation ($h\nu$) with energy higher than its band-gap (space between the molecule conduction band and valence band) level energy, an electron from the valence band migrates to the conduction band, creating a hole with a positive charge in the valence band ($h^+$) and an extra electron with a negative charge in the conduction band ($e^-$) [26,27]. The photo-generated charges move up to the PC's surface, which then give place to redox reactions when oxygen and water are present, generating reactive oxidizing species (ROS), mainly hydroxyl radical ($HO^\bullet$) and superoxide radical ($O_2^{\bullet -}$), but also hydrogen peroxide ($H_2O_2$) [28].

HP is a potentially sustainable technology for water disinfection, as PC can be activated employing solar light, and moreover, no additional chemical substance is needed in the process, which minimizes the potential formation of DBP and environmental harmful effects [29,30]. HP has also been researched for inorganic pollutants' removal from water, such as hexavalent chromium [31] or trivalent arsenic [32], and also for recalcitrant organic compound degradation, such as dyes, phenolic compounds, pesticides and pharmaceutical active compounds [33], posing HP as potential technology for comprehensive water treatment.

$TiO_2$ has remained as the most studied PC, despite the fact it is only active under UV irradiation (which accounts for nearly 5% of the total Sun spectral irradiation), whose wavelengths are in the range of 200–400 nm ($TiO_2$ peak absorbance is around 387 nm). It also has a relative high recombination rate of the photo-generated charges. Several approaches have been researched to address these issues, such as doping $TiO_2$ with other elements, coupling it with other PC to form heterojunctions, or even with itself to form homojunctions; these strategies generally improved $TiO_2$ photocatalytic activity to some extent, although most studies have only been carried out at lab-scale [25,34–37].

Other materials have also been researched, such as the bismuth oxyhalides, which are a group of layered materials with narrow band-gaps [38,39]. It is still unclear if a narrow band-gap is the answer to use solar light more efficiently. Black $TiO_2$, a variation of $TiO_2$ exhibiting dark coloration instead of white, and sometimes also referred to as reduced, hydrogenated or oxygen-vacant $TiO_2$, was first reported in 2011 and its ability to show photocatalytic activity even under infrared light was noticed; although activity under visible light has been reported and some studies show it outperforms pristine $TiO_2$, it is believed that this happens due to an improved use of UV irradiation rather than an effective use of visible or infrared light [40,41].

Water disinfection via HP has been broadly researched for at least the last twenty years, although disinfection via UV or heat might happen simultaneously [42]; in disinfection via HP, the generated ROS cause oxidative stress to a wide variety of microorganisms, including Gram-positive and Gram-negative bacteria, DNA viruses, and RNA viruses amongst others [43]. The microorganism cell integrity is compromised as the ROS cause damage to the cell membrane, resulting in cytoplasm leakage; ROS can also obstruct cell vital functions like protein synthesis or break biomolecules covalent bonds [44–46]. A schematic representation of ROS generation at molecular level (depicting $TiO_2$) and microorganism disinfection via HP is shown in Figure 1.

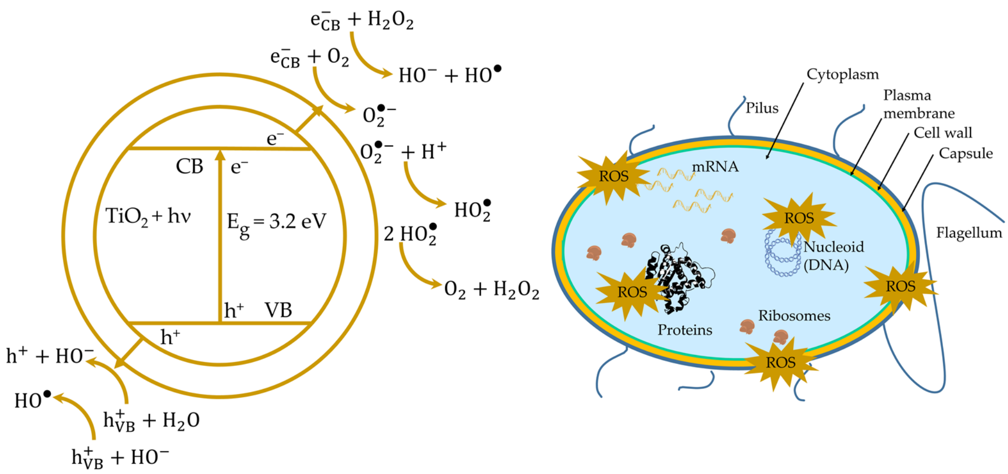


**Figure 1.** Schematic representation of ROS generation and disinfection via heterogeneous photocatalysis.

Water disinfection via solar HP depends on several factors, to name a few: nature and initial concentration of microorganisms, process duration, irradiation intensity, composition of the water matrix, turbidity, water layer depth, sunlight angle of incidence, water pH, and PC properties [45,47,48].

Despite its potential advantages, solar HP has not been deployed as a full-scale process yet; several limitations have been addressed such as low photoconversion efficiency, scarce information on energy consumption, and yields on catalyst preparation processes amongst others, but the lack of knowledge on the field of solar HP reactor design, as well as the absence of a consensus on proper design methodologies, are recognized as the main ones [49]. Several reactor designs have been proposed based on devices designed for solar thermal applications, such as the parabolic trough reactor (PTR) based on the parabolic trough solar collector, the compound parabolic reactor (CPC) based on the compound parabolic solar collector or the flat plate reactor (FPR), based on flat devices for solar energy collection [50,51].

The number of papers published on the topic of disinfection via HP has been increasing year by year during the last two decades, which reflects the interest of the scientific community in these technologies [52]. However, the amount of research focusing on reactors is generally scarce [53], and even though one of the most promoted characteristics of HP is its ability to make use of solar energy, plenty of research is performed using solar simulators instead of actual sunlight, as the controlled conditions allowed by these devices provide accurate and reproducible results [54]. PC efficiency differs between simulated sunlight and actual sunlight [55], hence, carrying out research employing actual sunlight is also relevant and needed.

The objective of the present work is to analyze scientific works focusing on the use of solar HP reactors for water disinfection. A systematic literature search was conducted following the preferred reporting items for systematic reviews and meta-analyses (PRISMA) protocol to discriminate non-pertaining works. The operations have been discussed in function of reactor type used, PC properties, type of the microorganism treated, disinfection performance, and location.

## 2. Methods for Literature Search, Inclusion Criteria, and Review

The literature review method was performed following the PRISMA four-step procedure [56,57]. The four steps are:

1. Identification of relevant papers indexed by databases.
2. Screen the papers for the determined criteria.
3. Verify papers' eligibility.
4. Incorporate the eligible papers in the systematic review.

An electronic search of articles was performed on Scopus and Web of Knowledge using the terms "photocataly*", "disinfection", and "solar", searching on title, abstract, and keywords in Scopus, and on all fields in Web of Knowledge. As it has been reported that research focused on reactors is scarce, comprising less than 2% of all papers related to HP [58], it was decided to include any work involving water disinfection regardless of the objective or publication year.

Books, book chapters, review articles, conference proceedings, and articles not published in English were excluded. Following the first exclusion, papers were screened for the inclusion criteria. Papers not focused on water disinfection (i.e., energy generation or pollutants' degradation), papers that did not make use of actual sunlight (i.e., solar simulators or UV-lamps), and papers that did not use a photocatalytic reactor (i.e., test tubes of bakers) were not included. Figure 2 illustrates the PRISMA steps taken for papers' eligibility.

Eligible papers were then analyzed for data extraction including the type of reactor employed, the PC used, type of microorganisms, microorganism concentration, disinfection efficiency, operation duration, volume of water treated, operation timeframe, and experimentation location.

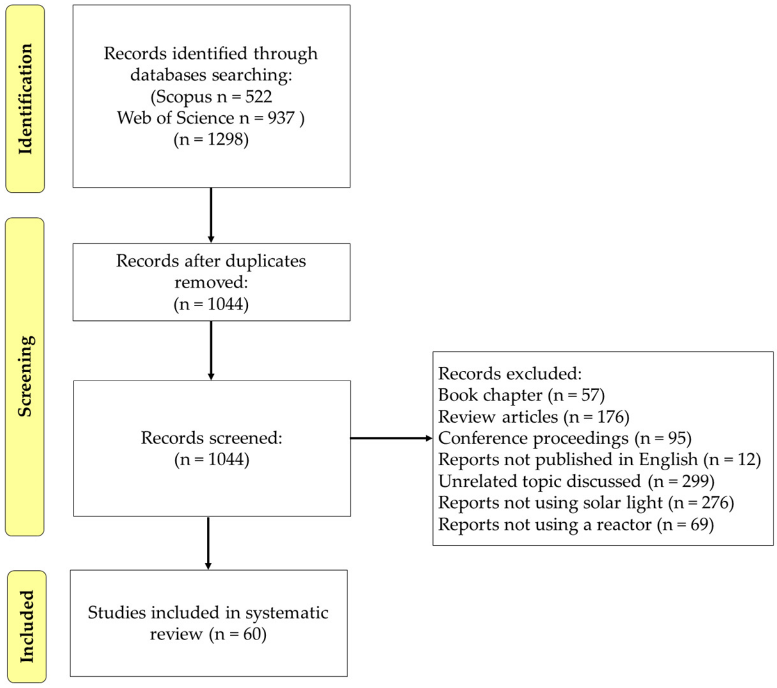


**Figure 2.** Flowchart for selection of literature.

## 3. Results and Discussion

### *3.1. Data Synthesis*

A total of 60 reports were deemed as appropriate for screening in this systematic review out of the 1043 records originally found. Table 2 shows the extracted data.

**Table 2.** Research reports focused on water disinfection via heterogeneous photocatalysis employing actual sunlight carried out in photocatalytic reactors.

| Reactor | Photocatalyst | Microorganism Treated | Initial Concentration (CFU/mL) | Log Cycle Reduction | Process Time (min) | Volume Capacity (L) | Operation Timeframe | Location | Reference |
|---|---|---|---|---|---|---|---|---|---|
| CPC type | $TiO_2$ | *Escherichia coli* | $10^5$ | −5 | 35 | 150 | Not reported | Not reported | [59] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 120 | Not reported | 14:00–16:00 | Seoul, Korea (38° S, 127° E) | [60] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^5$ | −4 | 30 | 35 | Not reported | Almeria, Spain (37° N, 2° W) | [61] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −4 | 12 | 11 | Not reported | Almeria, Spain (37° N 2° W) | [62] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 90 | 70 | 12:00–16:00 | Lausanne, Switzerland (46° N, 6° E) | [63] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^4$ | −3 | 45 | 11 | Not reported | Almeria, Spain (37° N, 2° W) | [64] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 60 | 1 | Not reported | Dublin, Ireland (53° N, 15° W) | [65] [a] |
| CPC type | $TiO_2$ | *Fecal coliforms* | $10^6$ | −6 | 360 | 20 | Not reported | Tucumán, Argentina (26° S, 64° W) | [66] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 150 | 37 | Not reported | Lausanne, Switzerland (46° N, 6° E) | [67] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 180 | 35 | Not reported | Lausanne, Switzerland (46° N, 6° E) | [68] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 30 | 35 | Not reported | Lausanne, Switzerland (46° N, 6° E) | [69] [a] |
| FPR type | $TiO_2$ | *Escherichia coli* | $10^4$ | −4 | 60 | 1 | Not reported | Not reported | [70] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^7$ | −5 | 90 | 14 | Not reported | Almeria, Spain (37° N, 2° W) | [71] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −4 | 90 | 14 | 09:00–10:30 | Almeria, Spain (37° N, 2° W) | [72] [a] |
| Rectangular type | $TiO_2$ | *Coliforms* | $10^6$ | −5 | 120 | 12.8 | 13:00–15:00 | Not reported | [73] |
| CPC type | $TiO_2$ | *Fusarium solani* | $10^3$ | −3 | 300 | 14 | 11:00–16:00 | Almeria, Spain (37° N, 2° W) | [74] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 50 | 20 | Not reported | Porto, Portugal (41° N, 8° W) | [75] [a] |
| CPC type | $TiO_2$ | *Fusarium* spp. | $10^3$ | −3 | 240 | 60 | 11:00–16:00 | Almeria, Spain (37° N, 2° W) | [76] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^5$ | −5 | 240 | 12 | 10:00–14:00 | Lares, Peru (13° S, 72° W) | [77] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | Not reported | 10 | Not reported | Almeria, Spain (37° N, 2° W) | [78] [a] |
| Offset tubular type | $TiO_2$ | *Coliforms* | $10^3$ | −3 | 240 | 300 | 11:00–15:00 | Not reported | [79] |
| Offset tubular type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 300 | 7 | 10:30–15:30 | Almeria, Spain (37° N, 2° W) | [80] |
| FPR type | $TiO_2$ | *Aeromonas hydrophila* | $10^5$ | −1.2 | 2.5 | 0.2 | Not reported | Queensland, Australia (20° S, 142° E) | [81] |

**Table 2.** *Cont.*

| Reactor | Photocatalyst | Microorganism Treated | Initial Concentration (CFU/mL) | Log Cycle Reduction | Process Time (min) | Volume Capacity (L) | Operation Timeframe | Location | Reference |
|---|---|---|---|---|---|---|---|---|---|
| FPR type | $TiO_2$ | *Aeromonas hydrophila* | $10^5$ | −1.38 | 2.5 | 0.2 | Not reported | Queensland, Australia (20° S, 142° E) | [82] |
| CPC type | $TiO_2$ | Phage ΦX174 | Not reported | −3 | 30 | Not reported | Not reported | Dublin, Ireland (53° N, 6° W) | [83] [a] |
| CPC type | $TiO_2$ | *Microcystis aeruginosa* | $10^7$ | −7 | 20 | 20 | Not reported | Porto, Portugal (41° N, 8° W) | [84] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^5$ | −4 | 300 | 10 | 11:00–15:00 | Almeria, Spain (37° N, 2° W) | [85] [a] |
| PTR type | $TiO_2$ | *Coliforms* | Not reported | −2 | 300 | 5 | Not reported | Indore, India (22° N, 75° E) | [86] |
| FPR type | N-$TiO_2$ | *Coliforms* | $10^3$ | −3 | 180 | 1.332 | 10:00–16:00 | Not reported | [87] |
| CPC type | $TiO_2$ | *Fecal coliforms* | $10^3$ | −1 | 120 | 1 | Not reported | Medellin, Colombia (6° N, 75° W) | [88] [a] |
| CPC type | $TiO_2$ | *Fusarium solani* | $10^2$ | −2 | 120 | 60 | 10:00–16:00 | Almeria, Spain (37° N, 2° W) | [89] [a] |
| Staircase reactor | $TiO_2$ | *Escherichia coli* | $10^6$ | −2 | 140 | Not reported | Not reported | Not reported | [90] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 300 | 8.5 | 10:00–15:00 | Almeria, Spain (37° N, 2° W) | [91] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | Not reported | 60 | Not reported | Almeria, Spain (37° N, 2° W) | [92] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^7$ | −4 | 300 | 15 | Not reported | Perpignan, France (42° N, 2° E) | [93] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^7$ | −2 | 90 | 6.4 | Not reported | Bangkok, Thailand (13° N, 100° E) | [94] [a] |
| PTR type | $TiO_2$ | *Fecal coliforms* | $10^6$ | −6 | 360 | Not reported | 09:00–17:00 | Madhya Pradesh, India (22° N, 75° E) | [95] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^5$ | −5 | 80 | 27 | Not reported | Cadiz, Spain (36° N, 6° W) | [96] [a] |
| Box type | $TiO_2$ | *Escherichia coli* | $10^7$ | −2 | 480 | 0.66 | 08:30–16:30 | Not reported | [97] |
| CPC type | $TiO_2$ | *Curvularia* sp. | $10^3$ | −3 | 120 | 20 | Not reported | Almeria, Spain (37° N, 2° W) | [98] [a] |
| CPC type | Ag-$BiVO_4$ | *Escherichia coli* | $10^6$ | −6 | 40 | 10 | 10:00–15:00 | Almeria, Spain (37° N, 2° W) | [99] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^6$ | −4 | 81 | 15 | Not reported | Dublin, Ireland (53° N, 6° W) | [100] [a] |
| Concave dish type | $TiO_2$ | *Heterotrophic bacteria* | $10^3$ | −4 | 240 | 1 | 10:30–14:30 | Gonabad, Iran (34° N, 58° E) | [101] |
| CPC type | $TiO_2$ | *Enterobacter cloacae* | $10^9$ | −4.9 | 180 | 3 | Not reported | Medellin, Colombia (6° N, 75° W) | [102] [a] |
| CPC type | $TiO_2$ | *Fecal coliforms* | $10^5$ | −5 | 300 | 20 | Not reported | Almeria, Spain (37° N, 2° W) | [103] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^7$ | −7 | 105 | 0.6 | 10:00–13:00 | Nsukka, Nigeria (6° N, 7° E) | [104] |

**Table 2.** *Cont.*

| Reactor | Photocatalyst | Microorganism Treated | Initial Concentration (CFU/mL) | Log Cycle Reduction | Process Time (min) | Volume Capacity (L) | Operation Timeframe | Location | Reference |
|---|---|---|---|---|---|---|---|---|---|
| Compound Triangular collector type | N-$TiO_2$ | *Escherichia coli* | $10^3$ | −1 | 180 | Not reported | 10:00–13:00 | Salerno, Italy (37° N, 14° E) | [105] [a] |
| Offset tubular type | Ag-$TiO_2$ | *Escherichia coli* | $10^5$ | −5 | 30 | 20 | 10:00–14:00 | Pondicherry, India (12° N, 79° E) | [106] |
| FPR type | $TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 60 | 1 | Not reported | Not reported | [107] [a] |
| PTR Type | Ag-$TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 180 | Not reported | 10:00–15:00 | Not reported | [108] |
| Offset tubular type | $TiO_2$ | *Coliforms* | $10^2$ | −2 | 560 | Not reported | 08:00–17:00 | Mae Salong Nok, Thailand (20° N, 99° E) | [109] |
| CPC type | ZnO | *Fecal coliforms* | $10^7$ | −7 | 15 | 1.1 | 11:00–15:00 | Tehran, Iran (35° N, 51° E) | [110] |
| FPR type | $TiO_2$ | *Fecal Coliform* | $10^6$ | −4 | 45 | 2 | Not reported | Durango City, Mexico (23° N, 104° W) | [111] [a] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^5$ | −3.5 | 360 | 2.2 | Not reported | Cadiz, Spain (36° N, 6° W) | [112] |
| CPC type | rGO-$TiO_2$ | *Klebsiella pneumonia* | $10^9$ | −9.3 | 210 | 0.39 | Not reported | Stellenbosch, South Africa (33° S, 18° E) | [113] |
| CPC type | $TiO_2$ | *Escherichia coli* | $10^7$ | −4 | 300 | 15 | 11:00–15:00 | Perpignan, France (42° N, 2° E) | [114] |
| Optofluidic capillary type | Red phosphorous | *Escherichia coli* | $10^8$ | −8 | 28 | 0.004 | Not reported | Not reported | [5] |
| Linear Fresnel type | $TiO_2$ | *Escherichia coli* | $10^4$ | −3 | 300 | 1 | 08:00–13:00 | Tsukuba, Japan (36° N, 140° E) | [115] |
| Through reactor | Fe-$TiO_2$ | *Escherichia coli* | $10^6$ | −6 | 120 | 6 | 12:00–14:00 | Patiala, India (30° N, 76° E) | [116] |
| Offset tubular type | $TiO_2$ | *Escherichia coli* | Not reported | Not reported | 420 | 120 | Not reported | Boyacá, Colombia (5° N, 72° O) | [117] |

[a] Cumulative UV dose ($Q_{UV}$) reported.

### 3.2. Type of Reactor

CPC commonly is composed of tubes made of borosilicate glass placed above parabolic reflectors made of polished aluminum, installed on an inclinable stand tilted at local latitude; water reservoirs and water pumps are also needed, and the most sophisticated ones count with radiometers, flow meters, and sensors for temperature, pH and dissolved oxygen measuring [118]. Due to its optical efficiency and its ability to use both direct and diffuse solar UV light, it has been considered as the most ideal solar reactor design available at the moment, especially for environmental applications [119], hence, its use on the majority of papers reviewed was within expectations. Figure 3 depicts a scheme of a CPC and its main components, as well as one of a FPR and one of a PTR.

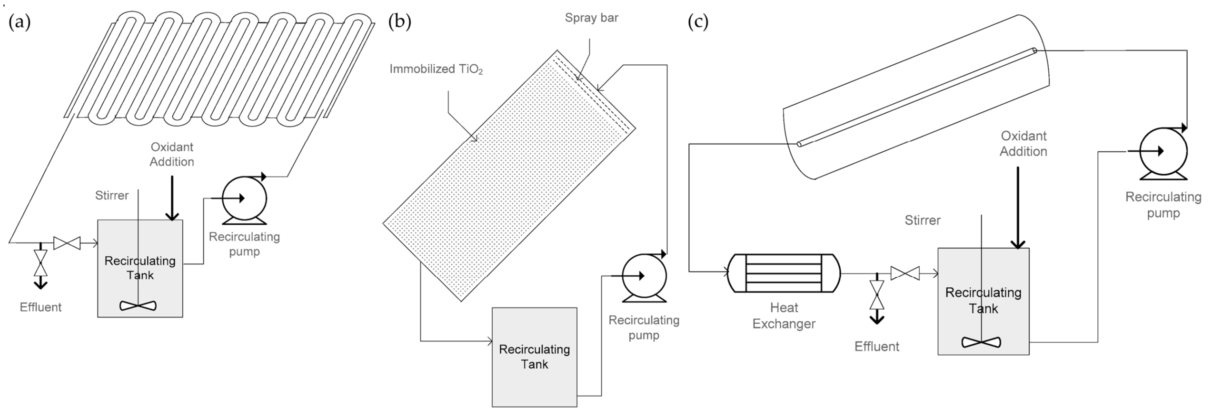


**Figure 3.** Schematic drawing of: (**a**) slurry CPC (Compound Parabolic Concentrator) system; (**b**) FPR (Flat plate reactor) system; (**c**) PTR (Parabolic Trough reactor) system (Reprinted from Ref. [120]).

FPR consists of a flat or corrugated surface over which a thin film of water flows (in most cases, in the laminar flow regime); it can be a proper approach for small-scale applications, as it offers a large surface area where the PC can be immobilized and its design is simple [121]. Although the reports reviewed in this paper employing this design were a lot less in number than those using a CPC design, it is worth mentioning that two of the papers reviewed reported steady-state operation rather than batch operation, which is one of the sought-after characteristics for reactors. An example on the operation of an FPR reactor can be seen in Figure 3 above.

Offset tubular reactors were employed in four studies. This reactor is similar to the CPC, with the only difference that it does not count with a reflector, which makes it less expensive than the CPC. As it does not need a reflector, more tubular sections could be positioned within the same space a CPC occupies, allowing the treatment of a higher volume of water, as a work which compared both reactor designs suggested [122]. A potential disadvantage in comparison to the CPC is the lower temperature the water might reach. Temperature in the range of 20–80 °C does not affect $TiO_2$ photo-excitation, and although dissolved oxygen concentration in water decreases as water temperature increases, ionic products of water ($OH^-$ and $H_3O^+$) do increase in the temperature range of 20–80 °C, which promotes $HO^\bullet$ generation [92]. In addition to damage caused by ROS to the microorganism, inactivation also happens due to UV light effect alone and temperature increase [123].

Three more papers reported the use of a PTR, in which main components include a parabolic reflector mounted over a rotating platform to concentrate sunlight; its use has dropped as it has numerous insuperable disadvantages: its components are relatively expensive, the high sunlight concentration leads to an excessive heating which hinders photocatalytic activity, and it is only able to use direct sunlight [115]. Likewise, a scheme showing an example of the operation of a PTR can be seen above in Figure 3. Table 3 shows the summarized advantages and disadvantages of the most used photocatalytic reactor types.

**Table 3.** Summarized advantages and disadvantages of the most used photocatalytic reactor types.

| Type of Reactor | Brief Description | Advantages | Disadvantages |
|---|---|---|---|
| Compound parabolic concentrator (CPC) | Transparent cylindrical receptors are placed onto two joined half-parabola shape reflectors [120] | Uniform light irradiation for cylindrical receptors; able to use both direct and diffuse solar irradiation; able to work at low solar concentration rates [50,120,124] | Aluminum reflector is imperative, which implies a higher cost; limited optical efficiency due to light being reflected multiple times [50,125] |
| Flat plate reactor (FPR) | Water flows over a tilted plate whence the PC has been immobilized [48] | Relatively lower price as no reflectors are needed [50] | Atmosphere can prevent the system to work in an appropriate way; high pressure is needed to pump water onto large surfaces [124] |
| Parabolic trough reactor (PTR) | A parabolic light-reflecting surface concentrates solar irradiation in a transparent receptor; usually operated in the turbulent flow regime; equipped with a sun tracking system [120] | Easily adaptable and developed technology; able to use both direct and diffuse solar irradiation [50,124] | Sun tracking systems implies a higher cost; aluminum reflector is imperative, which implies a higher cost; solar concentration factor above one sun inhibits photocatalytic activity; increase in temperature reduces dissolved $O_2$ concentration, which slows down photocatalytic activity; unable to use diffuse solar irradiation, rendering it impractical on overcast or cloudy days; a relatively large area needed for installation [50,120] |

The remaining eight reactors can be considered as empirical approaches (as no design parameters are included), that due to relative youngness of the field, results in research teams frequently employing their own singular design, as pointed out more than a decade ago [48].

It has already been stated that the biggest challenge to translate HP to commercial applications remains as the lack of an efficient reactor design suitable for treating large volumes of water [126]. CPC remains as the most studied reactor, with no update, which again points out the need of more research focused on reactors. Additionally, reactor innovation can be made in diverse ways; some process intensification attempts have been reported which involve combining solar HP with other processes, such as solar pasteurization [42] or ozonation [127]. Additionally, more research is needed to understand the interesting synergy between the different microorganism inactivation mechanisms which take place within a solar HP reactor (UV light inactivation and thermal inactivation).

### 3.3. Photocatalyst Used

$TiO_2$ was the most used PC in the papers analyzed, with a total of 51 studies. Around 1600 papers examining its usage in disinfection have been published over the last 20 years, with an increase from 5 papers in 2000 to 165 papers in 2019, which is an indicator of the growing research interest in heterogeneous photocatalytic disinfection, leading to scaled-up applications [30], just as the ones analyzed in the present work. $TiO_2$ is known for its noteworthy photocatalytic activity, due in part to its specific surface area of around 46.06 $m^2\ g^{-1}$ and a surface energy of 80 $mJ{\cdot}m^{-2}$, but also for its optical and electronic properties, high chemical stability, low cost, non-toxicity, and environmental friendliness [128–130].

On the other hand, $TiO_2$ has limitations, including fast electron-hole recombination even though adding $H_2O_2$ results in an almost inhibited recombination by scavenging $e^-$ [131], and other drawbacks include slow charge carrier transfer, elevated recycling cost, and photocatalytic activity under UV irradiation only, due to its wide band-gap of 3.2 V, limiting its efficiency for solar applications, as UV irradiation account for less than 5% of the received solar energy [132]. ZnO, a PC used in one of the papers reviewed, also has a band-gap value of 3.2 V, and its drawbacks are almost identical to those of $TiO_2$ [47].

To overcome these issues, $TiO_2$ modification by doping and coupling has been researched, i.e., Ag-$TiO_2$, Fe-$TiO_2$, N-$TiO_2$, and rGO-$TiO_2$, as used in some of the papers reviewed. Research towards PC with narrower band-gap and photocatalytic activity under visible light irradiation, such as the ones used in the papers reviewed, Ag/BiVO4 and red phosphorus, is the other relevant research trend [133].

When $TiO_2$ surface is modified by metal doping, a Schottky's barrier arises when irradiated with UV light, causing the metal Fermi levels to be lower than those of $TiO_2$ CB, increasing the metal ability to accept electrons, inhibiting charge recombination. When doping with a noble metal, surface plasmon resonance happens, which allows for electrons to be transferred directly to $TiO_2$ CB [134].

Fe-$TiO_2$ has shown a better performance than $TiO_2$, which is attributed to Fe atoms acting as electron-hole traps, slowing down charge recombination and enhancing photocatalytic activity as a result; it also enhances specific surface area [135,136].

Ag-$TiO_2$ has been researched since 1984 and it has several advantages over conventional $TiO_2$, such as a narrower band-gap of 2.77 eV, a higher specific surface area (239 $m^2\ g^{-1}$) and the plasmonic effect which increase visible light response. Ag-$TiO_2$ has been gaining attention from the scientific community, which is also aimed at overcoming its share of drawbacks, such as photocatalytic activity gradual loss and Ag leaching [132].

Nitrogen is the most common non-metal used as a doping agent for $TiO_2$, predominantly because of its small ionization energy and its atomic size comparable with that of oxygen. Doping with oxygen confers $TiO_2$ photocatalytic activity under visible light, although the exact mechanism for this enhancement is still elusive and not totally understood; N-$TiO_2$ has been used for several applications [137,138], including solar disinfection in a photocatalytic reactor.

Graphene-based and $TiO_2$ composites have also been researched, which has resulted in an increased photocatalytic performance [139]. Composited of $TiO_2$ and reduced graphene oxide (rGO) higher photocatalytic activity is attributed to a synergism involving a higher number of photocatalytic active sites, a superior light collection (due to the hierarchical structural interface between unidimensional $TiO_2$ and bidimensional rGo) and an enhanced charge separation rate [140].

PC based on bismuth have been widely researched for water disinfection applications, as they are non-toxic, chemically stable, visible light active, synthesized with ease, reusable, and relatively economic. Bismuth-based PC disadvantages include low light absorption, high charge recombination, and a slow charge migration [141]. One of the most promising PC is bismuth vanadate ($BiVO_4$), whose properties can be improved when doped with metals (such as in one of the works reviewed in this paper) [142]. Although 15% Ag doped $BiVO_4$ was effective for water disinfection, it was not able to outperform conventional $TiO_2$ [99].

Red phosphorous belongs to a different kind of PC, which are elemental PC; it is an allotrope inert to chemical reactions and of elevated thermodynamic stability; its band-gap in the range of 1.4–2.0 allows it to be active under visible light, its production is non-expensive as the raw material is of low cost, and it is non-toxic; it is a promising material for photocatalytic disinfection, although there is still non-consensus about its resistance to oxidation [143,144].

### *3.4. Microorganism Type*

The vast majority of works reviewed in this paper (a total of 38) analyzed water disinfection employing *E. coli*, which is within expectations due to the interest of the scientific community in it, being a research subject in evolutionary, biological genetic, and molecular studies [145]. *E. coli* is regarded as an indicator microorganism for the presence of bacteria by the WHO, as it has been characterized extensively and its presence, which indicates fecal contamination, is common in untreated water sources; however, it is inactivated with more ease than other microorganisms, hence, its absence does not guarantee that any other fecal coliform or microorganism are absent as well [146].

Among the reviewed works, five and six papers analyzed disinfection of total coliforms and fecal coliforms, respectively, which is completely appropriate within the interest of performing experiments in conditions as close to reality as possible, as it is well-known that disinfection rate is different between microorganisms of different species, and might even vary between strains of the same species [147]. Methods to quantify coliforms in the works reviewed included test paper and plate count [88,109].

One of the reviewed papers analyzed total heterotrophic bacteria disinfection, and four more analyzed disinfection employing a determined species. Several bacteria genres are included within the heterotrophic bacteria, such as *Aeromonas*, *Citrobacter*, *Enterobacter*, *Helicobacter*, *Klebsiella*, and *Serratia* among others. Some of these bacteria can cause health issues to humans with suppressed immunologic response, and are also associated with a low organoleptic water quality [148,149].

The last paper analyzing bacteria disinfection focused on *Microcystis aeruginosa*, which is a cyanobacteria that causes immense harm to ecosystems due to the release of cyanobacterial toxins and algal organic matter; its rapid growth greatly affects the efficiency of drinking water treatments [149]. Previous work has reported that *Microcystis aeruginosa* regrowth after photocatalytic is inhibited, as cell density is less than 85% compared to control experiments [150]. Since HP is also able to degrade organic compounds, it can offer a comprehensive alternative for cyanobacteria disinfection, as the detrimental cyanobacterial toxins can be degraded as well [126,151].

In the case of fungi disinfection, three papers focused on the genre *Fusarium*, which is a fungus that can cause a condition called fusariosis. Fusariosis symptoms depend on the affected area and the host's immunological response, but they can include nail, skin, and eye infection [152]. According to estimations, the species *Fusarium solani* is associated with 50% of all the fusariosis reported cases [153].

One of the papers examined *Curvularia* spp. disinfection; although it is infrequent, this genre can cause several types of human mycoses, including: fungal keratitis, onychomycosis, peritonitis, invasive sinusitis, subcutaneous disease, and systemic infections among others [154].

Viral disinfection was also reported. A paper analyzed disinfection of the ΦX174 virus, which is a phage that commonly infects *E. coli*, hence, its presence in water is also considered an indicator of fecal pollution [155].

Summarizing, solar HP reactors have been studied for water disinfection involving several types of microorganisms, and although the exact mechanism does surely differ from one microorganism to another, the oxidation caused by the ROS surely plays an important role.

### 3.5. Disinfection Performance

Most of the papers reviewed reported a high disinfection rate of several orders of magnitude. However, this data alone is not enough to properly assess process efficiency. It is well known that there are no established and unanimous figures of merit to evaluate the efficiency of HP processes, as the amount of diverse studies is vast, which include emerging technologies and processes combinations, giving as a result a massive challenge to critically assess HP efficiency, which is also multidimensional, as operational costs, sustainability, feasibility and yields, among other parameters, such as microorganism nature, initial concentration, water matrix and pH, to name a few, should, preferably, be considered [47,156].

Proposed benchmarks to evaluate HP efficiency include disinfection rate constant, photocatalytic space-time yield, photonic yield, quantum yield, and population log reduction among others; however, the methodologies to determine these figures of merit can differ from one study to another, hence, the IUPAC recommends to treat these figures of merit as only apparent, as a consensus on how to properly determine a benchmark for comparison among different reports is still lacking [30,53,157]. This issue has been known for decades and in 2001, the IUPAC published a technical report which suggested the use of several figures of merit to evaluate AOPs (including solar HP) [158]. Although the use of some of these benchmarks (i.e., collector area per order) is still reported on in recent scientific literature, none of the reports reviewed in the present work made use of any of them.

Thirty-two papers reported the use of $Q_{UV}$, which is the cumulative UV energy during an irradiation time per unit of volume of water; as this figure of merit considers solar irradiation intermittencies throughout the day, it can be considered accurate for efficiency assessments for the time being [159], considering the complex and variable composition of the water microbial consortium.

Ten out of the 69 analyzed papers reported an order of magnitude reduction smaller than 3, which indicates that in the vast majority of the reviewed works, a disinfection rate of at less 99.9% was achieved, which complies with the minimal health risk standard set by the WHO [160].

### *3.6. Volume Treated*

In 2011, the WHO estimated that a person needs between 50 and 100 L of water per day to meet their basic needs which include, but are not limited to, drinking water, food preparation, and sanitation. However, in water scarcity scenarios, the WHO recommends a minimum of 7.5 L of drinking water per capita per day [161–164]. The lack of reactor designs able to treat large volumes of water is one of the main reasons which restrains HP commercial and industrial application [165]; nevertheless, some of the papers reviewed in this present work report on treating water volumes in the range of 100–300 L, which could very well meet drinking water requirements of households or small public facilities [166], although actual application depends on more factors, such as level of automation or cost effectiveness, to mention some [7].

Future research should also focus on exploring solar HP reactors or systems able to disinfect water in a steady-state operation rather than batch operation; the volume output still needs to be increased, and even though scaling-up might pose a challenge due to light distribution, scaling-out or numbering-up might offer a feasible alternative, providing land for installation is available; finally, any system should be tested in conditions as close to reality as possible [167,168].

### *3.7. Experimentation Location*

The reviewed works were performed around the globe in latitudes as northernmost as 53° N and southernmost as 38° S, as well as in longitudes as westernmost as 104° W and easternmost as 142° E, which indicates water disinfection via HP could be used in varied locations. However, UV radiation reaching Earth's surface varies around the world and through time, and is dependent on many factors, such as presence of clouds, atmosphere ozone concentration, sunlight reaching the surface oblique angle, aerosol particles concentration, sun elevation, and surface reflectivity, among others [169].

Disinfection via solar HP is related to SODIS, which is being globally promoted. Guidance has been made public to facilitate the worldwide implementation of a standardized procedure [170,171], although, at the moment, there are no available predictive approaches for SODIS expected efficiency worldwide [172], hence, the scenario is analogous for disinfection via solar HP.

### *3.8. Discussion, Considerations, and Prospects*

Based on the present literature review and considering the relatively small number of works that make use of real solar irradiation, there has not been any considerable advance in reactor design in more than two decades, with batch-operated CPC reactor being the most common operation. One of the papers reported coupling solar HP with an electrochemical process [114], which can be considered a process intensification approach [173], rather than an improvement in reactor design by itself.

HP reactor design is challenging, as it involves complex interactions between the PC and microorganisms with the light [174], which can explain the minimal innovation in solar HP reactor designs. It is worth mentioning that recent research on reactor modeling has generated important information regarding reactor design in function of the radiant energy absorbed, shedding light on the relevance of PC chemical properties, PC loading and the high relevance of the Damköhler number (the ratio of the rate of a chemical reaction to diffusive mass transfer rate) [175,176].

HP has also shown efficiency in antibiotic resistant microorganism disinfection, which is not always achieved by conventional technologies such as chlorination [177–179].

Another important reason for HP research on water treatment is that the presence of emerging pollutants (such as: active pharmaceutical ingredients or personal care products) has been reported in groundwater, surface water, and tap water; even if they are commonly found in trace concentrations, its occurrence poses a threat to human health and HP-based technologies can degrade the pollutants or their organic compounds' precursors, mitigating their formation [180–182]. In addition to the intrinsic effect these substances can cause on their own account, when water undergoes disinfection by chlorination, organic compounds can react with chlorine and give rise to the formation of DBP; drinking water with trace concentrations of disinfection byproducts can have a chronic adverse effect on human health [183]. As HP is theoretically able to mineralize organic compounds [184], its implementation within a water potabilization process could potentially mitigate disinfection byproducts' formation [185].

It has been reported than there is a disproportion on the amount of studies focusing on fundamental science regarding HP compared to that regarding applied science; more than 129,000 papers have been published on the HP topic, although usually these studies focus on the application of photocatalytic reactions employing a benchmark PC to a specific process, or in the performance of a new PC applied to a benchmark process [186]. More multidisciplinary endeavors are needed to keep improving HP efficiency.

## 4. Conclusions

The present work provides a systematic review making use of the PRISMA methodology, which served the purpose of discriminating non-relevant works, including only papers focused on water disinfection employing HP making use of actual sunlight, with a total of 60 papers found. This information sheds light on the need of performing research employing real sunlight as well, as photocatalysis efficiency differs when simulated sunlight is used.

The found papers were analyzed in terms of several operational parameters, identifying the following trends:

1. CPC reactor is the most used type of reactor and its design has not received any major modification in decades.
2. $TiO_2$ remains the most researched PC despite being unable to use visible light. The use of modified $TiO_2$ for allowing its visible light activity was more reported than the use of other PCs.
3. The reports indicated good disinfection efficiency, but the use of proper benchmarks is not a standardized practice.
4. Most of the works reported the working volume, with some of them treating enough water for households or small public buildings.
5. Water disinfection via solar HP has been performed in many places around the globe, but proper models to predict disinfection efficiency in different locations are still lacking.

More research is needed in several disciplines to keep improving HP efficiency, aiming to its large-scale application in the future.

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