Review

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Literature Review on Graphitic Carbon Nitride (g-C3N4) as Photocatalyst for the Remediation of Water Polluted with Contaminants of Emerging Concern

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Review

Literature Review on Graphitic Carbon Nitride ($g\text{-}C_3N_4$) as Photocatalyst for the Remediation of Water Polluted with Contaminants of Emerging Concern

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Abstract: Carbon nitrides are polymeric materials with a broad extent of applications, including photocatalysis. Among them, graphitic carbon nitride ($g\text{-}C_3N_4$), a low-cost material, is an excellent photocatalyst under visible light irradiation owing to its features such as correct band positions, high stability and non-toxicity. $g\text{-}C_3N_4$ is a metal-free material that is easily synthesized by polymerizing nitrogen-rich compounds and is an efficient heterogeneous catalyst for many reaction procedures due to its distinctive electronic structure and the benefits of the mesoporous texture. In addition, in situ or post-modification of $g\text{-}C_3N_4$ can further improve catalytic performance or expand its application for remediating environmental pollution. Water pollution from organic compounds such as pesticides and pharmaceuticals is increasing dramatically and is becoming a serious problem around the world. They enter water supplies in a variety of ways, including industrial and hospital wastewater, agricultural runoff, and chemical use. To solve this problem, photocatalysis is a promising technology. Without the use of other oxidative chemicals, $g\text{-}C_3N_4$ uses renewable solar energy to transform harmful pollutants into harmless products. As a result, much recent research has focused on the photocatalytic activity of $g\text{-}C_3N_4$ for wastewater treatment. For this reason, the main objective of this paper is to contribute a chronological overview of the bibliometrics on the $g\text{-}C_3N_4$ for the removal of pesticides and pharmaceuticals from water using the tools BibExcel, Bibliometrix and R-Studio IDE. A bibliometric analysis was performed using the Science Citation Index Expanded (WoS©) database to analyze the scientific literature published in the field over the last 10 years. The results were used to identify limitations and guide future research.

Keywords: bibliometrics; carbon nitride; heterogeneous photocatalysis; pesticides; pharmaceuticals; water treatment

1. Introduction

Carbon nitrides (CN) are polymeric materials with a wide range of applications derived from carbon materials by substituting N for the C atoms, making them attractive for a diversity of applications such as membranes, adsorbents, catalytic reactions, photocatalysis, sensors, supercapacitors, solar and fuel cells, hydrogen storage devices and biomedical [1–3]. All these applications are dependent on the exceptional electronic, optical, and chemical properties of CN in combination with its synthesis from easily available precursors and its resistance to adverse physical and chemical conditions. Like most carbon materials, CN have an extended history, dating back to 1834, when a material called "*melon*" (linear polymers of tri-s-triazines linked via secondary N) was described by Liebig in 1834 [4], although the potential value of the material in question has only been completely accepted in recent decades. This is probably due to its high thermal (stable up to 600 °C in air) [5], hydrothermal stability (insoluble either in acidic, neutral or basic solvents) [1,6] and its

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undisclosed structure. This allows the material to work in both gaseous and liquid environments and at higher temperatures, increasing its wide range of applications [7]. CN materials have been intensively researched since Liu and Cohen [8] predicted that they have the potential to be ultra-hard materials. Among various analogues, graphitic carbon nitride (g-C_3N_4) as an analog of graphite, a polymeric compound constructed via tri-s-triazine units n-type semiconductor, is considered the most stable environmental allotrope and a hot topic in materials science due to its extraordinary electronic structure. Contemporary activity has been stimulated by findings indicating that the "*graphitic*" materials may have helpful properties for catalysis and energy (converting or storing), as well as other possible applications [9]. Currently, g-C_3N_4 is making a difference in many areas of chemistry. Suffice it to say that the rate of publications on CN has increased exponentially from 2015 to date, as shown in Figure 1.

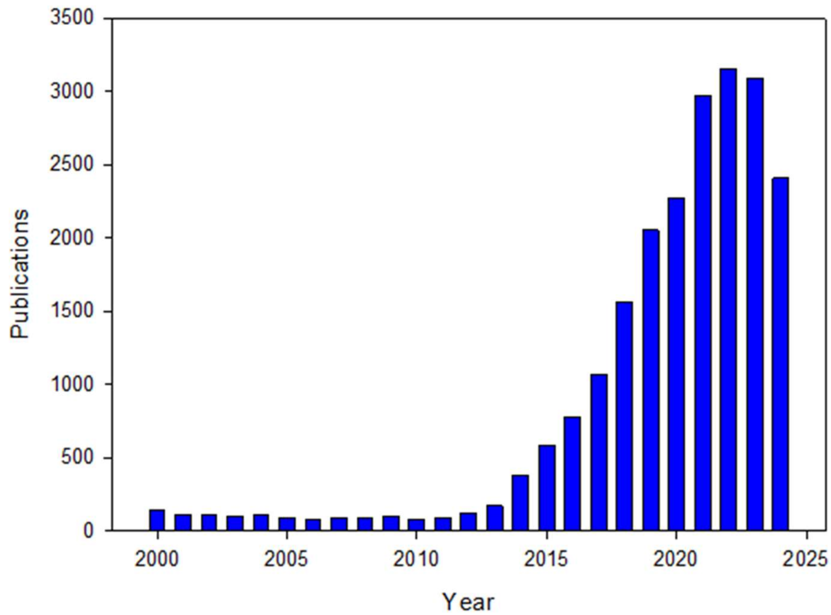

Figure 1. Number of publications including ''carbon nitride'' OR ''g-C_3N_4'' in title from 2000 to October 2024 (Source: WoSCC, dated 10 October 2024).

1.1. Nomenclature and Synthesis

The most suitable nomenclature to designate different classes of chemically and physically produced CN materials is the first issue to be addressed [10]. It is increasingly common to call them g-C_3N_4, which is usually defined as a group of compounds with a general formula close to C_3N_4 and structures constructed on heptazine units. However, this definition is misleading due to different reasons. For this reason, it is also valuable to develop an effective approach to the labeling of the different classes of CN compounds. This approach should accurately describe their chemical and structural properties as they relate to their functional performance. First, these C_3N_4 forms are best described as $C_xN_yH_z$ compounds. This is because most of the materials that have been obtained to date contain not only C and N, but also significant amounts of hydrogen (H) as a part of their constitutions. On the other hand, materials made from linked heptazine (tri-s-triazine, C_6N_7) units are questionable to be fully condensed "graphitic" structures. As an alternative, they form zig-zag polymer chains related to those present in Liebig's "*melon*", with a composition close C_2N_3H. In addition, other compounds with planar C_3N_4 layers are also formed by polytriazine imide (PTI) linked units providing hosts for embedded ions including Li^+, Cl^- and Br^- as well as extra H^+ [11]. The presence of H reveals that the real g-C_3N_4 is not completely condensed and that there are several surface defects, which can be valuable in catalysis. Due to the presence of H, and because N has one

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electron more than C, g-C_3N_4 has a variety of surface properties essential for catalysis, including basic surface functionalities, electron-rich properties, H-bonding motifs, etc. [12].

Although g-C_3N_4 is a two-dimensional covalent structure like graphene, the latter consists of pure C, while the former contains C and N. The semiconducting properties of g-C_3N_4 are significantly different from those of graphene sheets. g-C_3N_4 is a CN type, which has seven kinds of structures with different band gaps (Eg): alpha (Eg = 5.5 eV), beta (Eg = 4.8 eV), cubic phase (Eg = 4.3 eV), quasicubic (Eg = 4.1 eV), g-h-triazine (Eg = 3.0 eV), g-o-triazine (Eg = 0.9 eV), and g-h-heptazine (Eg = 2.9 eV) [13,14]. Like graphite, g-C_3N_4 also has a sheet structure containing C_3N_3 and C_6N_7 rings [15]. The rings are connected by the N at the end to form an infinite plane. g-C_3N_4 is an economical and benign material that is chemically and physically very stable. It has a narrow bandgap (Eg = 2.7 eV) agreeing to an optical wavelength (λ) of about 460 nm because its yellow color that is appropriate for visible light (λ > 400 nm) absorption and photocatalytic activity for environmental pollution remediation, among other applications [16]. Density functional theory (DFT) calculations shows that the Eg of melon is 2.6 eV, decreasing from 3.5 eV in melem (the simplest heptazine-based compound), and finally decreasing to 2.1 eV with the complete formation of condensed g-C_3N_4, since the reduction and oxidation levels are related to the positions of the valence (VB) and conduction (CB) bands [6].

Bulk g-C_3N_4 (bg-C_3N_4) can be easily obtained at high temperature by direct thermal polymerization of the precursors. Reactive nitrogen-rich and oxygen-free compounds with pre-bonded C-N core structures such as thiourea, urea, melamine, cyanamide, dicyandiamide, guanidinium chloride, guanidine thiocyanate, ammonium thiocyanate, and/or hexamethylenetetramine are the most used precursors for the chemical synthesis of g-C_3N_4 [3,14,17–20]. Based on planar heptazine or triazine cores, these precursors produce two-dimensional layered graphitic structures. The condensation pathways from precursors such as urea and ethylenediamine to cyanamide and dicyandiamide, as well as melamine and all other C/N materials, are easy and suitable synthetic routes to produce slightly deformed polymeric species, as shown in Figure 2.

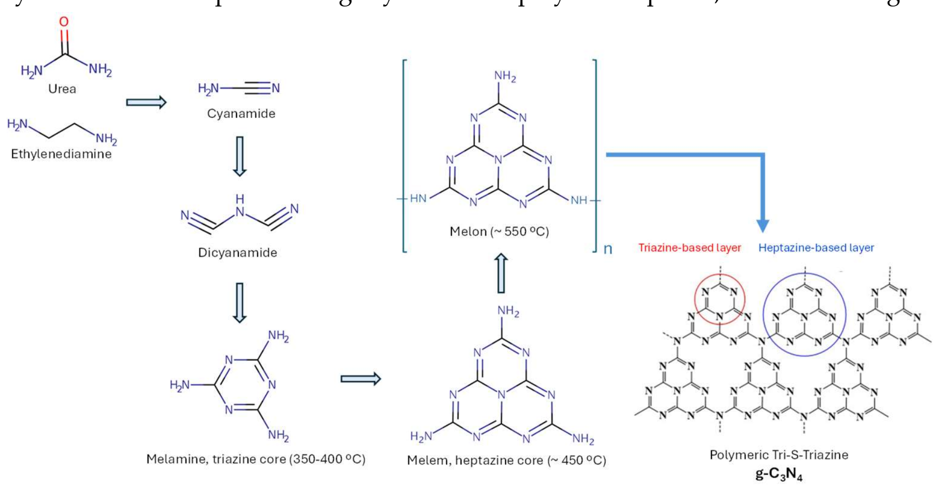

Figure 2. Reaction pathway from precursors to graphitic carbon nitride (g-C_3N_4).

The methods used to prepare and characterize bg-C_3N_4 are well established, although common bulk material defects such as little specific surface area and fewer active sites hinder more advanced improvement of bg-C_3N_4. It is for this reason that mesoporous materials (MM) have been a hot topic in the last few decades. MM are known for the pore channels (< 50 nm in size) inside them, which give them superior properties to the bulk materials for various applications. In general, mesoporosity has several outstanding characteristics, including ordered pore structure, high specific surface area, more active sites, low density and high adsorptive capacity [21–23]. The transformation of g-C_3N_4 into mesoporous material is valuable for its catalytic activity, the increase of active sites, the improvement of photon operation rate, and the promotion of further research and extensive application of g-C_3N_4 materials. Therefore, a lot of effort has been devoted to the preparation and study of MM. These include several major methods used to produce mesoporous g-C_3N_4 (mg-C_3N_4), including hard- and soft-template methods, template-free method, sol-gel method, supramolecular preorganization method, exfoliation method and others [3,20,24,25]. These methods yield products with diverse pore regularity and other properties using different strategies and precursors.

1.2. Photocatalytic Properties and Applications

mg-C_3N_4 is an effective heterogeneous catalyst for many processes due to its distinctive electronic structure and the improvements of its mesoporous structure. In addition, the catalytic performance of mg-C_3N_4 can be further enhanced or its range of applications extended by in situ or post-modification. For example, when mg-C_3N_4 is used as a photocatalyst, appropriate changes can greatly improve its photocatalytic activity. To date, reducing the Eg to gain light absorption in the VIS region or improving the detachment of photogenerated electrons (e^-) and holes (h^+) have been the most common modification routes to improve the performance of g-C_3N_4 as a photocatalyst to reduce their recombination, which can improve the quantum efficiency of photocatalysis. The changes can cause light absorption to undergo a red shift, which broadens the absorption capacity and results in improved utilization of sunlight. Currently, several methods of modification for mg-C_3N_4 exist. Non-metal doping, noble metal loading, metal oxide loading, dye photosensitization, and polyoxometalate immobilization are the most representative and typical modification methods [16,18,26–30]. Generally, elemental doping is assumed as an effective strategy for performance enhancement through modification of its electronic structure and surface properties of g-C_3N_4 for effective photocatalyst. To improve the separation of photogenerated e^- and to inhibit the recombination of e^-/h^+ pairs, the heterojunction structure is always used. Combining simultaneously doping and heterojunction engineering to modify g-C_3N_4 with a great potential for efficient visible light photocatalysis is expected to be very useful. The enhancement of heterojunctions based on doped g-C_3N_4 has been recently the focus of many studies [31].

Although TiO_2 is an excellent catalyst because of its stability, non-toxicity, wide band gap (Eg = 3.2 eV) and charge recombination, it has low catalytic efficiency as photocatalyst because of the large band gap, which limits the use of a wide spectrum of solar light (λ < 388 nm), leading to much lower quantum efficiencies when using solar spectra [32,33]. As compared to TiO_2, g-C_3N_4 has a suitable medium-wide Eg for efficient absorption of visible light. In addition, flexibility in improving the photocatalytic properties by doping with metal or non-metal ions to generate active sites with generous melon moieties, design of optimized heterojunctions, and morphological modification to increase the surface area are another important point to improve its photocatalytic performance. By changing the band gap, morphology, and separation of photogenerated e^- and h^+, efficiency can be improved, making g-C_3N_4 a very promising material for use as a photocatalyst for environmental remediation and other applications [1,34,35].

Heterogeneous photocatalysis (HP), one of the best studied Advanced Oxidation Processes (AOPs), involves four main processes: i) light harvesting, ii) charge excitation, iii) charge separation and transfer, and iv) surface electrocatalytic reactions [28,36]. Briefly, HP is based on the semiconductor (SC) irradiation particles (in this case g-C_3N_4), commonly suspended in aqueous solutions (slurry), with a wavelength energy ≥ Eg. In the course of photocatalysis, g-C_3N_4 is wholly excited by the absorption of photons with energies (hv) greater than its Eg. This drives an e^- into the CB, leaving an h^+ in the VB and encouraging the migration of the e^- and h^+ to the particle surface (Figure 3). The energy is dissipated as heat and the e^- and h^+ can recombine on the particle surface in a short time. The photoexcitation process of g-C_3N_4 can be summarized as follows (Eq. 1):

$$g - C_3N_4 + hv = g - C_3N_4 - h^+ + \ g - C_3N_4 - e^- \quad (1)$$

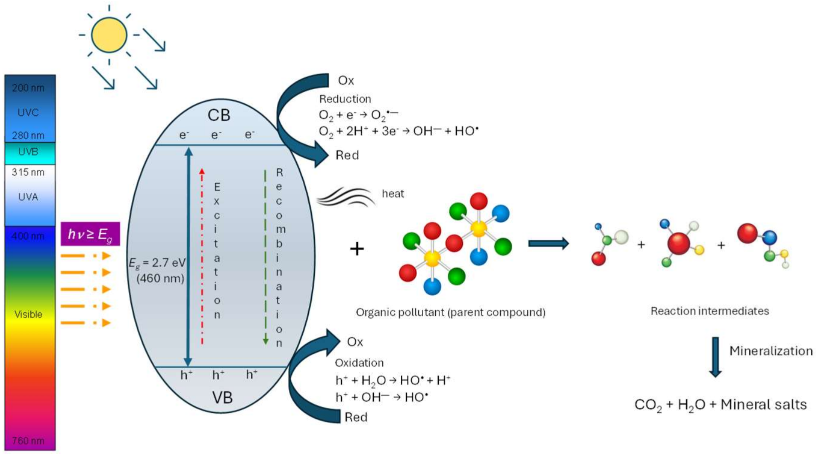

Figure 3. Scheme for photodegradation of organic pollutants by heterogeneous photocatalysis with g-C_3N_4.

Relying on the fact that the lifetime of the photogenerated charges is longer than the recombination rate of them, there is still an evident amount of e^- and h^+ available. They can be trapped by defect sites in g-C_3N_4 or transferred to its surface where energy is lower than its CB, which helps separate and transfer carriers for successful surface redox reactions. On the other hand, the undesired recombination of most of the e^-/h^+ pairs cause tremendous energy loss through the release of heat or the emission of light. In general, for the reduction of surface-adsorbed molecules to generate radicals and for the direct oxidation of organic contaminants, the surface e^- and h^+ serve to donate and accept electrons, respectively [30].

During surface reactions, the redox aptitude of h^+ and e^- essentially depends on the VB (-1.30 eV) and CB (1.40 eV) potentials of g-C_3N_4 [24]. Hence, photogenerated e^- can readily react with adsorbed O_2 to produce non-selective radicals ($O_2^{\bullet-}$) owing to the less negative standard redox potential of $O_2^{\bullet-}/O_2$ (- 0.13 eV vs NHE) [37]. However, due to the redox potential of $HO^\bullet/OH^-$ (+1.99 eV vs NHE) is much higher than the VB potential of g-C_3N_4, the remaining h^+ is not able to be scavenged by H_2O and OH^- to form $HO^\bullet$ in a kinetic manner [38]. Similarly, a small amount of $HO^\bullet$ is generated by the reduction of adsorbed O_2 via multi-electron reaction processes [16]. During photocatalytic process, $O_2^{\bullet-}$ and h^+ have a critical role for g-C_3N_4 in the degradation of pesticides to CO_2, H_2O and other small molecule products [39]. In addition, the production of reactive oxygen species (ROS) from adsorbed O_2 by the capture of e^- also helps to limit the recombination of e^-/h^+ pairs.

Note that surface reactions can only happen if the reduction/oxidation potentials are more positive or negative than the CB and VB values, respectively. Compared to TiO_2 and other semiconductor materials, g-C_3N_4 has the most negative CB level (-1.3 V vs. NHE at pH = 7) and a medium band gap (2.7 eV) [40], which facilitates its large application in VIS-light photocatalysis. Thus, the Eg of g-C_3N_4 can be further narrowed by simple doping, defects, and other possible sensitization actions to achieve a higher utilization of VIS-light. Considering its strong reduction ability, activity in VIS-light, abundance, easy fabrication, stacked 2D layered structure, non-toxicity and high stability, its direct use in the field of sustainable chemistry as a multi-functional heterogeneous metal-free photocatalyst is possible. However, g-C_3N_4 mostly shows low photocatalytic efficiency because some serious disadvantages of the material itself, such as high e^-/h^+ recombination rate, low surface area (~10 $m^2 \cdot g^{-1}$), insufficient visible absorption below 460 nm, high degree of monomer condensation, moderate oxidation capacity, and small active sites for interfacial photoreactions, grain boundary effects, slow surface reaction kinetics, and low charge mobility interfering with electron delocalization [37]. Although these prominent challenges severely limit the improvement of photocatalytic performance, they also provide more chances for future synthesis of more efficient g-C_3N_4-based photocatalysts.

1.3. Pesticides and Pharmaceuticals as Emerging Pollutants

Pesticides and pharmaceuticals are widely used throughout the world, with recognized benefits to human health and well-being. However, despite their necessity, these classes of chemicals are also associated with serious risks to human and environmental health and are common micropollutants of aquatic environments. In addition, knowledge of the risks of their transformation products in the aquatic environment is very important [41].

Pesticides (*substances intended to repel, destroy or control pests and diseases and/or prevent undesirable plant growth*) have the potential to pollute surface- and groundwater, among other environmental compartments, in addition to other emerging pollutants (EPs). They can reach surface waters through soil runoff and cause groundwater pollution by leaching through the soil profile [42]. The occurrence of pesticide residues in surface- and groundwater is increasing in OECD countries, with a significant number of samples exceeding the legal limits. A European Environment Agency (EEA) report shows that 16% of EU surface waters are of unknown chemical status, 38% are of good chemical status, while 46% are not of good chemical status [43]. Another EEA technical report focusing on 39 European countries assessed the incidence of pesticides and their main transformation products in surface- and groundwater, showing exceeding rates ranging from 5-15% for herbicides and 3-8% for insecticides in surface water, while in groundwater the percentages were 7% and < 1%, respectively with lower frequencies for fungicides in both surface- and groundwater [44]. In addition, for recalcitrant pesticides, the polluted water (from agricultural, industrial and urban sources) treated by standard wastewater treatment plants (WWTPs) is in some cases inadequate to achieve regulatory purity requirements. This issue is particularly important where low levels of rainfall do not offer adequate water supplies to meet the needs of the agricultural sector, which requires increased reuse of effluent from WWTPs [45]. Furthermore, particularly in developing countries where monitoring data is often lacking and chemical analyses are often not performed due to lack of facilities or financial constraints, further research efforts are needed to better understand the presence and impacts of pesticide use in water bodies [46,47].

On the other hand, pharmaceuticals (*compounds used in the treatment or prevention of human and animal diseases to restore, correct, or modify organic function*), are often found in high concentrations in the aquatic environment all over the world [48]. This is probably due to their continuous release from WWTPs, which is significantly faster than their removal rates. Pharmaceuticals, which are not readily biodegradable and may persist and remain toxic, have attracted considerable attention for their frequent detection in natural and wastewater bodies as well as drinking water [49]. Consequently, pharmaceutical residues present current and potential risks to human and environmental health.

In this context, AOPs have achieved high interest in the last years, and their applications to remove pesticide and pharmaceutical residues from water have recently increased, especially solar heterogeneous photocatalysis [50–56]. The main benefit of these technologies is that they eliminate or at least decrease pesticide residues by mineralizing rather than transferring, as happens in conventional treatments [57–59].

1.4. Bibliometric Study

Bibliometric reviews are now very common and consist of the analysis of scientific publications using statistical methods to provide an overview and general structure of the target research area [60]. In science, bibliometric studies serve to reveal the bibliometric structure that comprises the system of organization among the components of a field of knowledge and that helps to reveal its intellectual dimension, based on groupings of relevant topics in that field of research [61]. In other words, bibliometric analyses provide a qualitative and quantitative approach to a particular field of research. Thus, it uses bibliometric data about the research field, such as total number of publications, authors, citations, institutions and countries, to build a complete picture of the research area [62]. In addition, bibliometric approaches are increasingly central to literature reviews in many fields of knowledge, as they help to analyze and visualize the status, structure, hotspots and future research trends in each discipline. Indeed, the large number of bibliometric reviews published in different scientific disciplines is striking, as a search for the keyword "*bibliometric analysis*" in the title field yields almost 15,500 publications using the Web of Science (WoS®) database.

Therefore, the aim of this work was to provide accurate and up-to-date bibliometric information on the development and future prospects of the use of g-C_3N_4 as a heterogeneous photocatalyst in applications for the remediation of water contaminated with pesticide and pharmaceutical residues reported in the literature to date, since, to the best of the authors' knowledge, there is no bibliometric review of its use and could help researchers, especially those new to the field, to identify possible future lines of research by summarizing the main aspects already covered and identifying critical points of interest.

2. Results and Discussion

2.1. Publication Patterns and Characteristics

The WoSCC database identified a total of 629 publications related to the g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water research field. The 629 publications were published between 2015 and 2024, of which 106 (18.85%) were published as open access articles. According to the WoSCC nomenclature, the 629 publications were: 576 (91,57%) Article, 46 (7.31%) Review, 2 (0,31%) Review; Early Access, 2 (0.31%) Article; Early Access, 1 (0,15%) Meeting Abstract, 1 (0.15%) Article; Proceedings Paper, 1 (0.15%) Article; Retracted Publication. In addition, 627 (99.68%) were published in English, the language of science [50], and 2 in Chinese.

Table 1, created with the BibExcel tool, shows the main characteristics of the 629 publications found. As can be observed, the annual number of authors increased significantly from 19 in 2015 to 1,093 in 2023. The largest number of publications occurred in the last four years (2020 to 2023), accounting for 81.2% of the total publications, with 2023 being the year with the highest scientific production, accounting for 26.6% of the total. The average number of authors per publication was 6.3, and the average number of pages per publication (13.8) increased from 12.7 in 2015 to 13.5 in 2023.

Table 1. Publication characteristics by year from 2015 to 2024.

PY	TP	TP AAI (%)	AU	AU AAI (%)	AU/TP	NR	NR AAI (%)	NR/TP	PG	PG AAI (%)	PG/TP
2015	3	0.0	19	0.0	6.3	260	0.0	86.7	38	0.0	12.7
2016	4	33.3	23	21.1	5.8	185	-28.8	46.3	42	10.5	10.5
2017	7	75.0	38	65.2	5.4	352	90.3	50.3	76	81.0	10.9
2018	21	200.0	144	278.9	6.9	1486	322.2	70.8	279	267.1	13.3
2019	31	47.6	205	42.4	6.6	2062	38.8	66.5	343	22.9	11.1
2020	82	164.5	505	146.3	6.2	5011	143.0	61.1	1014	195.6	12.4
2021	119	45.1	735	45.5	6.2	8919	78.0	74.9	1605	58.3	13.5
2022	143	20.2	965	31.3	6.7	10032	12.5	70.2	1896	18.1	13.3
2023	167	16.8	1093	13.3	6.5	11291	12.5	67.6	2258	19.1	13.5
2024	52	-68.9	317	-71.0	6.1	4499	-60.2	86.5	1419	-37.2	27.3
Average		53.4		57.3	6.3		60.8	68.1		63.6	13.8

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	62	404	4409	897
Total	9	4	7	0

TP: total publications; AAI: average annual increase; AU: authors number; AU/TP: average of authors per article; NR: cited reference count; NR/TP: average of reference per article; PG: page count; PG/TP: average of pages per article.

Price's Law was used to analyze the productivity of this research field [63]. A linear fit of the data obtained, according to the equation y = 68.733x – 138598, and a further fit to an exponential curve, according to the equation $y = 0e^{0.574x}$ were carried out to assess whether the growth of the scientific production of g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water research field follows Price's law of exponential growth. As shown in Figure S1A, mathematical fitting to a linear curve gave a correlation coefficient of $R^2 = 0.8313$. Furthermore, an exponential fit of the measured values gave $R^2 = 0.9696$. From these data it can be concluded that the database analyzed is better adapted to an exponential adjustment and that the postulates of Price's Law are fulfilled.

Price's Index measures the percentage of documents cited in bibliographies in recent years. An index value close to 50% indicates that the current literature is abundant, and the topic remains relevant, suggesting that it has not yet reached its final stage of exploration. For this research field, Price's Index was found to be 70.99%. This value is significantly higher compared to other research fields, such as emerging contaminants in coastal waters (43.4 %) [64] or the QuEChERS sample preparation procedure (50.3%) [65], because it is a relatively new research. In addition, this high value signifies that this research area is currently in vogue, as it is significantly above the 50% threshold.

A four-order polynomial model with a high coefficient of determination ($R^2 = 0.9997$) was established to describe the relationships between the annual cumulative number of publications and the year of publication from 2015 to 2023 in relation to g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water research field (Figure S1B). This model predicted an increase in the number of future publications for 2027, with an estimated number of 1018 publications, almost double the papers published in 2023 (582).

2.2. Author Performance

The 629 publications included in this bibliometric review were authored by 3,269 different authors. Lotka's Law, a principle used in bibliometrics to categorize authors based on their productivity, is expressed as $X^n \cdot Y = C$. Here, Y represents the number of authors who have published X articles, while C is determined by the specific field of study. The value of n typically falls between 1.2 and 3.5 [66,67]. Figure S2A illustrates Lotka's Law ($X^{3.141} \cdot Y = 2{,}535.1$), which demonstrates the relationship between the number of authors and the number of publications. The line in the Figure S2A aligns perfectly with the theoretical prediction, yielding an R^2 value of 1 across the entire range studied (from one to teen publications). This indicates that Lotka's Law holds true, suggesting that an increasing number of scientists will likely engage in this area of research.

Authors can be categorized based on their publication count: those with more than 10 works are considered "great producers," those with 5 to 9 works are termed "moderate producers," authors who have published between 2 and 4 works are labeled "aspirants," and those with only 1 published work are referred to as "passers-by" or "occasional authors" [68]. In the context of this research field, out of the total authors, 3 (0.1%) were classified as great producers, 36 (1.1%) as moderate producers, 424 (13.0%) as aspirants, and 2806 (85.8%) as occasional authors.

The collaboration index, calculated by dividing the total number of authors of articles that have multiple authors by the total number of articles with multiple authors [69] was found to be 6.43. This fact suggesting that the research team consists of 6–7 authors, data that are consistent with the above-mentioned average number of authors per publication (6.3). Collaboration among authors reflects effective teamwork, which is vital due to the diverse nature of modern research and its associated costs.

Table S1 shows the list of the 20 most prolific and most cited authors on g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water research field, ranked by h-index. It can be noted that there are not many authors who stand out for their high contribution to this field of research and that most of them are of Chinese nationality. In this classification, the authors P. Singh, G. Zeng and L. Li stand out with 10 publications (1.6%). The most cited authors are GM. Zeng (1251), P. Singh (861) and P. Raizada, who have the highest h-indexes of 9, 10 and 8 respectively. The two most important authors in this area are P. Singh (Shoolini University, India), whose research group is working to generate new knowledge about photocatalytic materials and processes in order to develop new decontamination treatments with greater efficiency and applicability, and GM. Zeng (Hunan University, China), whose work focuses on environmental systems engineering, urban waste recycling and clean production techniques and methods.

2.3. Journals Performance and WoS Categories

The 629 publications were published in 117 different journals with a publication range between 1 and 113 publications. The power model applied to calculate Lotk*a*'s Law for journals, as well as the number of publications in the g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water research field, indicated a correlation coefficient very close to 1 (R^2 = 0.9972), with a range of one to six journals (see Figure S1B). Therefore, we can expect a wider variety of journals in this research field in future publications.

Table S2 shows the 15 most productive journals ranked by h-index. Chemical Engineering Journal ranked first with 113 publications (h-index of 55 and total citations of 8588). Journal of Hazardous Materials and Chemosphere ranked second and third with 44 (h-index of 33 and total citations of 2919) and 69 publications respectively (h-index of 31 and total citations of 2456).

Further, Bradford's Law [70,71] states that scientific journals can be ranked based on their productivity in a specific research area. In this sense, the Bradford's core of g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water research field consisted of 3 journals (Chemical Engineering Journal, Chemosphere, and Journal of Hazardous Materials) (Figure S3), all with fourty-four or more publications, accumulating 226 (35.9%) of the 629 publications.

Additionally, the 629 documents were published across 50 different subject categories in the WoS, suggesting that the research field is inherently multidisciplinary. Figure S4 illustrates the six most productive WoS categories in this research field from 2015 to 2023. It is evident that the category "Environmental Sciences" has stood out since 2019, followed by "Engineering, Environmental" and "Engineering, Chemical". This facts suggest that there will be a guarantee that additional journals will be included in the research area.

2.4. Countries and Institutions Performance

The 629 documents were published across 58 different countries worlwide with a publication range between 1 and 421 publications. The twenty most productive countries in terms of h-index are listed in Table S2. China lead the ranking alone with 421 (66.9%) publications and a h-index of 76 in the research field, followed by India with 96 (15.2%) publications and a h-index of 36, and USA with 31 (4.9%) publications and a h-index of 23. The environmental monitoring policies that China has implemented in recent years may be one of the reason for its top ranking, similar to what occurs in other areas of environmental research [64]. Of note, a review on the use of g-C_3N_4 to remove tetracycline from wastewater also reported that the majority of studies are centralized in China [72] due to, mainly, that China rank first in the production and consumption of tetracycline worldwide.

Figure S5 illustrates the country collaboration map, highlighting the top collaborating countries. China also has the highest number of collaborations, totaling 150 partnerships with researchers worldwide. The principal collaborating countries include the USA (n=21), followed by Saudi Arabia (n=20) and India (n=17).

Table S3 shows the top 15 most productive institutions ranked by the h-index. The most productive institution was the Hunan University (China) with 33 publications (5.2%, h-index = 24), followed from afar by the Shoolini University (India) with 17 publications (2.7%, h-index = 15) and

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Chinese Academy of Sciences with 19 publications (3.0%, h-index = 14). In conclusion, all but 3 of the 15 most productive institutions (Shoolini University, India; King Saud University, Saudi Arabia and SRM Institute of Science & Technology, India) are also located in China, the country to which most of the most productive authors belong.

Figure S1C illustrates Lotka's Law ($X^{2.392} \cdot Y = 632.59$), which describes the relationship between the number of institutions and the number of publications. The correlation coefficient is extremely high ($R^2 = 0.996$) within the range of one to eight publications. This indicates that Lotka's Law holds true, suggesting that an increasing number of institutions are likely to become involved in this field of research.

2.5. Keywords Analysis

2.5.1. Trending Keywords

To perform a trending keyword analysis, keywords from authors between the years 2015 and 2024 were selected (Figure 4). The numbers indicate the frequency of each keyword, represented by the blue circles within the specified time range. A larger circle radius signifies a higher frequency of the keyword. The blue line illustrates the years during which these keywords were trending, marked by the first (Q1) and third (Q3) quartiles.

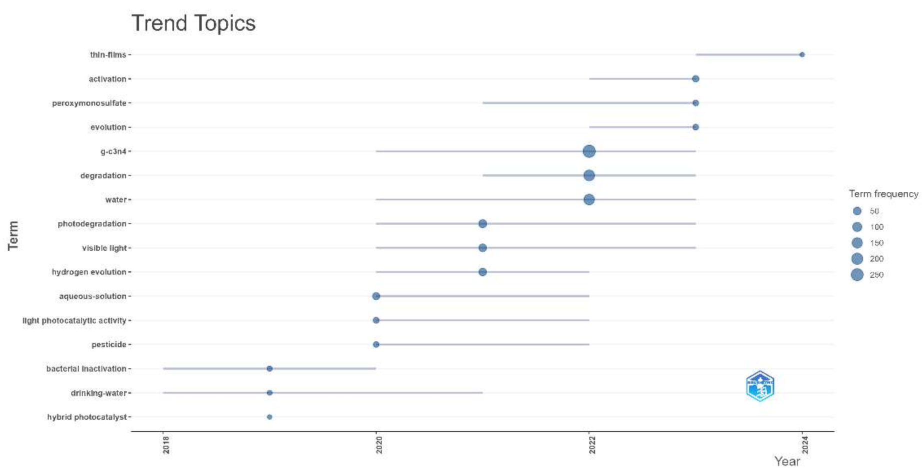

Figure 4. Trending keywords analysis from 2015 to 2024.

The most frequent keywords from 2020 to 2023 are "g-C_3N_4", "degradation" and "water" with frequencies of 269, 177 and 165 respectively. This may be because the use of g-C_3N_4 as a catalyst has shown great potential in photocatalytic water purification under visible light irradiation for the degradation of pollutants, mainly pesticides and antibiotics, as well as in bacterial inactivation processes [39,73–78]. Other keywords appear less frequently (fewer than 60 times) and some have not been mentioned since 2021, such as "drinking water" and "bacterial inactivation." The term "thin films" is the newest addition, with a low frequency of 5, but it has been observed in the years 2023 and 2024. This may be because with new synthesis developments, the use of a thin film of ground g-C_3N_4 compared to the dispersed catalyst allows for higher photocatalytic activities and reaction rate constants, increased the overall time stability of the thin film and its reusability getting a vast improvement in the photodegradation process efficiency [79,80]. This trend topic analysis highlights the continued use of g-C_3N_4 in water remediation processes alongside new methodologies.

2.5.2. Impact and Centrality of Critical Documents

Figure 5 illustrates the Impact Centrality Strategic Diagram (ICSD), which represents the impact and centrality of critical documents in the research area. The total number of documents grouped by the coupling process is represented by the sizes of colored circles, each displaying distinct content and highlighting the key topic words within the circle. The position of each cluster reflects its impact and centrality. Quadrant I (QI) represents clusters that have both high impact and high centrality, known as motor clusters. Quadrant II (QII) includes clusters with high impact but low centrality, referred to as fading clusters; Quadrant III (QIII) indicates clusters that exhibit both low impact and low centrality, known as emerging clusters, and Quadrant IV (QIV) features clusters with low impact but high centrality, called promising clusters [81].

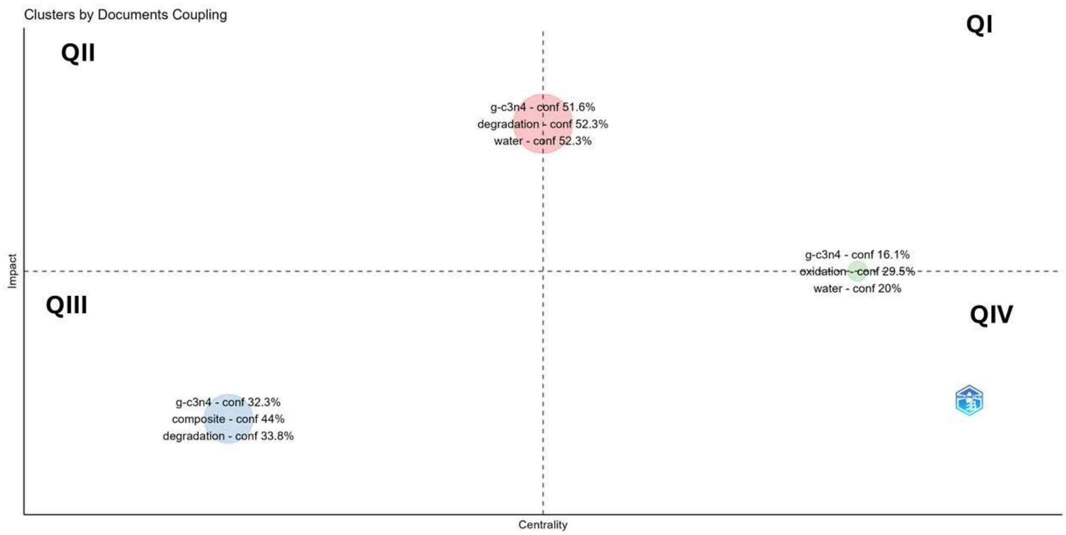

Figure 5. Impact-Centrality strategic diagram by documents.

Cluster 1 (represented in orange) lies between Quadrants I and II. It comprises 125 documents, with a centrality value of 0.37 and an impact score of 2.37. The main terms identified in this cluster include documents with "graphitic carbon nitride as g-C_3N_4", "water," and "degradation", which show their high impact and high centrality". The configuration values are of great importance, with values of 51.6%, 52.3% and 52.3% respectively. This also confirms what was discussed in the previous section (2.5.1 Trending keywords) and justifies the main use of the catalyst for the degradation of pollutants in water. Cluster 2 (represented in blue) consists of 100 documents, and is situated in QIII with a centrality value of 0.34 and an impact value of 1.74. Two words found in Cluster 1 ("g-C_3N_4" and "degradation") are also present in this cluster; however, the word "composite" is new and has a configuration value of 44%. It is known that the photocatalytic activity of g-C_3N_4 can be optimized by changing synthesis technology. It is clear that composites are an effective method for improving the physico-chemical properties of g-C_3N_4 catalysts [82,83]. The addition of other materials (doped g-C_3N_4) allows significant improvements [84]. These include an increased adsorption capacity, a reduction in the bandgap to shift light absorption towards the visible, an enhanced stability of the structure and improved recyclability [85,86]. This results in a significant increase in photocatalytic performance and utilization which is key to its effectiveness in water splitting and pollutant degradation. Some authors report that g-C_3N_4 composites with magnetic materials can be immobilised on the surface of g-C_3N_4, creating magnetically separable photocatalysts. These multifunctional visible-light-driven photocatalysts can be easily separated from the treated solution using an external magnet [74,87]. On the basis of the above, we can justify the presence of the term "composite" in Quadrant III, known as emerging word clusters.

Finally, the third cluster (represented in green) is situated between QIV and QI, comprising 48 documents where, in addition to the words "g-C_3N_4" and "degradation" the word "oxidation" is also present with a configuration value of 29.5%. This is most likely because g-C_3N_4 is an effective heterogeneous catalyst for advanced oxidation processes (AOPs), that involve the generation of highly oxidising radicals, such as hydroxyl radicals ($HO^{\bullet}$), capable of reacting with recalcitrant

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organic compounds and eliminating them [88] through oxidation/reduction processes that occur simultaneously at the catalyst surface.

2.5.3. Thematic Evolution Analysis

Figure 6 shows the thematic evolution analysis based on co-word network analysis and clustering and how the themes are related, evolve and change over time. This figure illustrates the evolution and relationships among research topics based on keywords, categorized into three distinct analysis periods. To do this, the Walktrap algorithm was used with the keywords plus (keywords of each publication generated by an algoritm os WoS), grouped into three periods of analysis and a top 1000 words with a minimum frequency of five in the respective papers pertaining to the clusters. The temporal segmentation is based on the authors' subjective judgment regarding technological development.

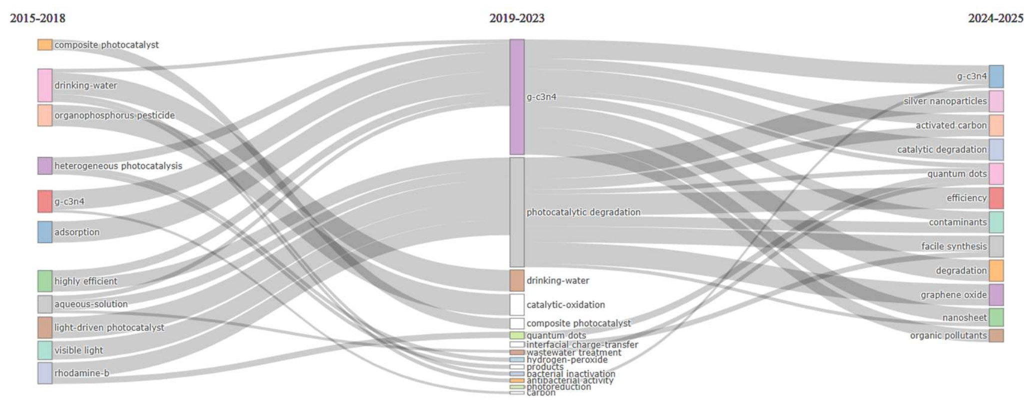

Figure 6. Thematic evolution analysis of the research field.

As general rule, the terms g-C_3N_4, (photo)catalytic degradation, contaminants and water are highly related to the other research topics such as bacterial disinfection [89]. According to this figure, we can see that the research topic "pesticide" in the first-time interval is strongly related to "catalytic oxidation" and "composite photocatalyst" in the second time interval [90], but not to any of the third time interval. The reason is that the algorithm used does not show the occurrence of the term "pesticide" within the keyword plus of the different documents analyzed. This is because they are grouped under photocatalytic degradation, which in turn is related to "contaminants" and "organic pollutants". In the second period, new research topics such as "wastewater treatment" or "quantum dots" appear in a short proportion, which grow in the third period and are grouped as "quantum dots" [91–93]. In the third period, the term "contaminants" appears more frequently, which in turn includes different types such as pesticides, antibiotics, dyes and others, due to the existence of a greater number of publications that generally study the use of g-C_3N_4 for the photocatalytic degradation of contaminants [39,73,77,90,94–98]. Finally, it's worth noting that in recent times, new terms such as "silver nanoparticles" or "nanosheets" have emerged. This is due to technological developments that allow the use of new forms of g-C_3N_4 [99–101].

4. Materials and Methods

This study analyzes research patterns, trends and advances in the field of g-C_3N_4 as photocatalyst for the remediation of pesticide and pharmaceutical polluted water.

The Science Citation Index Expanded (SCI-E) of the WoSCC database was used as the data source for this analysis. The data were obtained through a systematic search for documents matching the "topic" field (title, abstract and keywords). The search interval was from when records exist until October 29th, 2024 (the date of the search). The terms used were ("carbon nitride" OR "g-C_3N_4" OR "graphitic carbon nitride") AND (photocataly*) AND ("pesticid*" OR "pharmaceutic*") AND (water).

The full record and cited references in plain text format are included in the exported metadata. The exported metadata used includes document type, journal, language, publication year, science categories, title, total citations, cited references, corresponding author, DOI, abstract, keyword plus, and keyword. The quality of the imported data was excellent, with no loss of information in all the categories imported including document type, journal, language, publication year, science categories, title, total citations, cited references, corresponding author, DOI, abstract, keyword plus and keyword's author.

Publication patterns and characteristics, prediction of publications, Price's law, Price's index, Lotka's law, h-index of authors, journals, countries and institutions, and analysis of WoS categories were calculated using the software's Bibexcel [102] and Microsoft Excel 365. Keywords analyses were performed using the Biblioshiny package of the Bibliometrix software [103] with the Rstudio IDE desktop 2024.09.01 tool and R 4.4.2.

The Journal Citation Reports (JCR) Science Edition 2023 was used to obtain the journal impact factor (IF) and the indexing categories of each journal [104]. Extensive standardization was performed to minimize the risk of bias during analyses. For example, author keywords and Keywords plus (i.e., catalysts by catalyst, photocatalysts by photocatalyst, composites by composite, pathways by pathway, graphite carbon nitride y graphitic carbon nitride by g-C_3N_4, heterostructures by heterostructure, mechanisms by mechanism, frameworks by framework, nanocomposites by nanocomposite, nanosheets by nanosheet, pesticides by pesticide, pharmaceuticals by pharmaceutical, response-surface by response surface, semiconductors by semiconductor, titanium-dioxide by TiO_2, visible-light by visible light, waste-water by wastewater, or antibiotics by antibiotic) were conscientiously standardized.

This study has inherent limitations typical of bibliometric analyses, including the database utilized, the chosen Boolean strings, the manual standardization performed, and the bibliometric parameters applied to evaluate the selected publications [105].

5. Conclusions

This is the first bibliometric literature review study on graphitic carbon nitride (g-C_3N_4) as photocatalyst for the remediation of pesticide and pharmaceutical polluted water. This research field follows Price's law of exponential growth. Authors, journals, and institutions have adhered to Lotka's law, and Price's index (70.99%) indicates that the research field is very far from obsolete. The most productive authors and institutions are in China which also is the most productive country. Chemical Engineering Journal is the leading journal of the research field, with a focus on Environmental Science WoS category. Finally, hot research areas focus on degradation, water, composite and oxidation.

Supplementary Materials: The following supporting information can be downloaded at the website of this paper posted on Preprints.org, Table S1: The 20 most productive authors sorted by h-index, Table S2: The 20 most productive journals sorted by h-index; Table S3: The 20 most productive journals sorted by h-index; Figure S1: A) Growth of g-C_3N_4 scientific production from 2015 to 2024 with both linear and exponential adjustment of the data to verify Price's law of exponential growth; B) Relationship between the cumulative number of publications and year published, since 2015; Figure S2: A) Growth of g-C_3N_4 scientific production from 2015 to 2024 with both linear and exponential adjustment of the data to verify Price's law of exponential growth; B) Relationship between the cumulative number of publications and year published, since 2015; Figure S3: Bradford's law: Journal rank (on a logarithmic scale) versus cumulative articles; Figure S4: Bradford's law: Journal rank (on a logarithmic scale) versus cumulative articles; Figure S5: Bradford's law: Journal rank (on a logarithmic scale) versus cumulative articles.

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original draft; Writing – review & editing and Simón Navarro: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Writing – original draft; Writing – review & editing.

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