

# Progress in obtaining chitosan from crustacean biowaste using eco-friendly processes: a bibliometric mapping

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## Abstract

The traditional methods for recovery of chitin from crustacean biowaste and its subsequent deacetylation for the production of chitosan uses strong acids and bases. Chemical demineralization, deproteination and deacetylation have several drawbacks for the final product and are harmful to the environment. Thus, the conversion of the raw material into chitosan by eco-friendly or sustainable processes has challenged researchers in recent years. The growing range of chitosan applications, which are determined mainly by their molecular weight, degree of deacetylation and type of processes employed, have also driven research. The landscape of literature over the last 22 years is discussed in this paper by using bibliometric analysis and mapping based on bibliographic metadata from the Web of Science database. It was possible to have an overview of the history, trend, forecast of studies and the current collaboration network in order to obtain chitosan with environmentally friendly and sustainable methods.

**Keywords:** bibliometric mapping, biopolymers, chitin production, crustacean shell, biotechnological processes.

## 1. Introduction

Chitosan is polysaccharide produced through the deacetylation of chitin, a polysaccharide found in the exoskeleton of crustaceans, through an alkalization process under high temperatures. Chitin is the polysaccharide more abundant in nature after cellulose [1], being synthesized by an enormous amount of living beings [2, 3]. However, its commercial production is mainly made from crustacean biowastes due to their high chitin contents and availability [3]. The amount of biomass discarded during the processing of shrimp and crabs to obtain their meat is very large, configuring a polluting problem if an adequate destination is not found. Producing chitosan can be a good alternative to reduce the impact caused by the accumulation of these residues in the places where they are generated.

The production of chitosan-based biomaterials has been the focus of several researches, since the use of natural polymers for diverse applications contributes to the advances of science and presents several advantages of easy applicability, biocompatibility and biodegradability [4]. Some of the main areas of chitosan application are: agriculture, water treatment, food industry, cosmetics and biopharmaceutical industry, with the biomedical area being one of the promising ones (surgical sutures, dental implants, bone reconstitution, contact lenses, encapsulation materials, among others). Such applications depend on the sources and mainly on the obtaining processes [1, 4, 5].

The chemical processes of demineralization and deproteinization used to obtain chitin, and deacetylation to obtain chitosan have several drawbacks, such as poorly controlled depolymerization, resulting in molecular weight reduction and hydrolytic deacetylation and are energy consuming. If the use of chitin/chitosan from biowaste is a sustainable approach, the chemical processes can bring environmental problems, such as the disposal of wastewater, making this process ecologically aggressive due to the high concentration of strong acids and caustic chemicals employed [6].

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4 Biotechnological processes such as fermentation or enzymatic processes, have appeared as  
5 promising alternatives to the chemical processes [6–8]. Microbial fermentation and enzymatic  
6 extraction for recovery of chitin from crustacean waste was reviewed by Gortari and Hours [6].  
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8 In this review, critical issues encountered in biotechnological processes for recovering chitin  
9 from crustacean shells, along with the creation of the most advanced discoveries during the last  
10 decades are shown.  
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18 Bibliometric analysis presents increasing development in terms of software and uses  
19 and may help enormously in the developmeent and prospection of new research topics.  
20 Bibliometric studies have provided support in the areas of sustainable biopolymers [9],  
21 construction industry [10], PVC polymer [11], lipase immobilization [12], food packaging  
22 applications [13], with medical and dental application [5], among others.  
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30 The development of new materials based on chitin and chitosan to be used in different  
31 technological applications is a very attractive field of research, which has been contributed to a  
32 large number of scientific publications. The biotechnological extraction of chitin and  
33 production of chitosan from crustacean shell residues has been increasingly researched on a  
34 global scale. However, chitin bioextraction and chitosan production are not currently exploited  
35 to its full potential at the commercial level, in order to be considered as a green, clean,  
36 ecological and economical process, as an alternative to chemical processes. In this sense, the  
37 evolution of knowledge about the processes for obtaining, characterizing and evaluating the  
38 properties of chitosan by eco-friendly processes is the main objective of this bibliometric study.  
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## 53 **2. Materials and Methods**

54 This research attempted to assess the scientific literature on the possibilities of eco-  
55 friendly chitosan production using systematic bibliometric analysis from major crustacean  
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4 biowastes, i.e., from shrimp and crab shells. Bibliographic metadata was collected from the  
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6 Web of Science (WoS), one of the main international scientific databases.  
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9 Scientific databases, which are indexing systems for journals, reports, books, theses,  
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11 annals of events, among others, can be used in a more systematic way when using mathematical  
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13 and statistical tools in order to facilitate searches for bibliographic references, to serve as a  
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15 theoretical platform for the most diverse purposes of future research [14]. In addition to being  
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17 an instrument that facilitates the retrieval and use of scientific knowledge in research,  
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19 bibliometric analyzes contribute to the establishment of indicators of the potential impact of a  
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21 given journal, authors, research groups, countries in an area of knowledge and which are the  
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23 new trends.  
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27 During the search for bibliographical references, some key words were carefully  
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29 selected that encompassed and delimited our object of study. The choice for the WoS database  
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31 was made because it is one of the main indexing platforms for scientific citations and the best  
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33 data quality; furthermore, the merging of WoS and Scopus collections is still a very problematic  
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35 task [15] . Thus, the search was restricted to scientific articles published from 2001 onwards,  
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37 focusing on articles and reviews in English and in the following bibliographic metadata: title,  
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39 abstract and authors' keywords and keyword plus. The keywords were selected along with the  
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41 Boolean operators OR (for union of concepts) and AND (for intersection). The final expression  
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43 used in the WoS website were: (chitosan) AND (shrimp\* OR crab\*) AND (\*acetyl\* OR  
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45 enzym\* OR \*bio\* process\* OR eco-friendly). The flowchart methodology for searching  
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47 references in the Web of Sciences scientific database is presented in (Figure 1).  
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52 From the records obtained by the WOS bibliographic search, all metadata were exported  
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54 to bibtex files (\*.bibtex), including bibliographical references. These records were then  
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56 imported into the R language environment using the bibliometrix package [16] gathered and  
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58 converted in a dataframe and later exported to Excel format (\*.xlsx) using the openxlsx package  
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[17]. This Excel file was later loaded by the Biblioshiny application using the *biblioshiny()* function. Biblioshiny is a web-based environment through which all graphics presented in this article were generated.

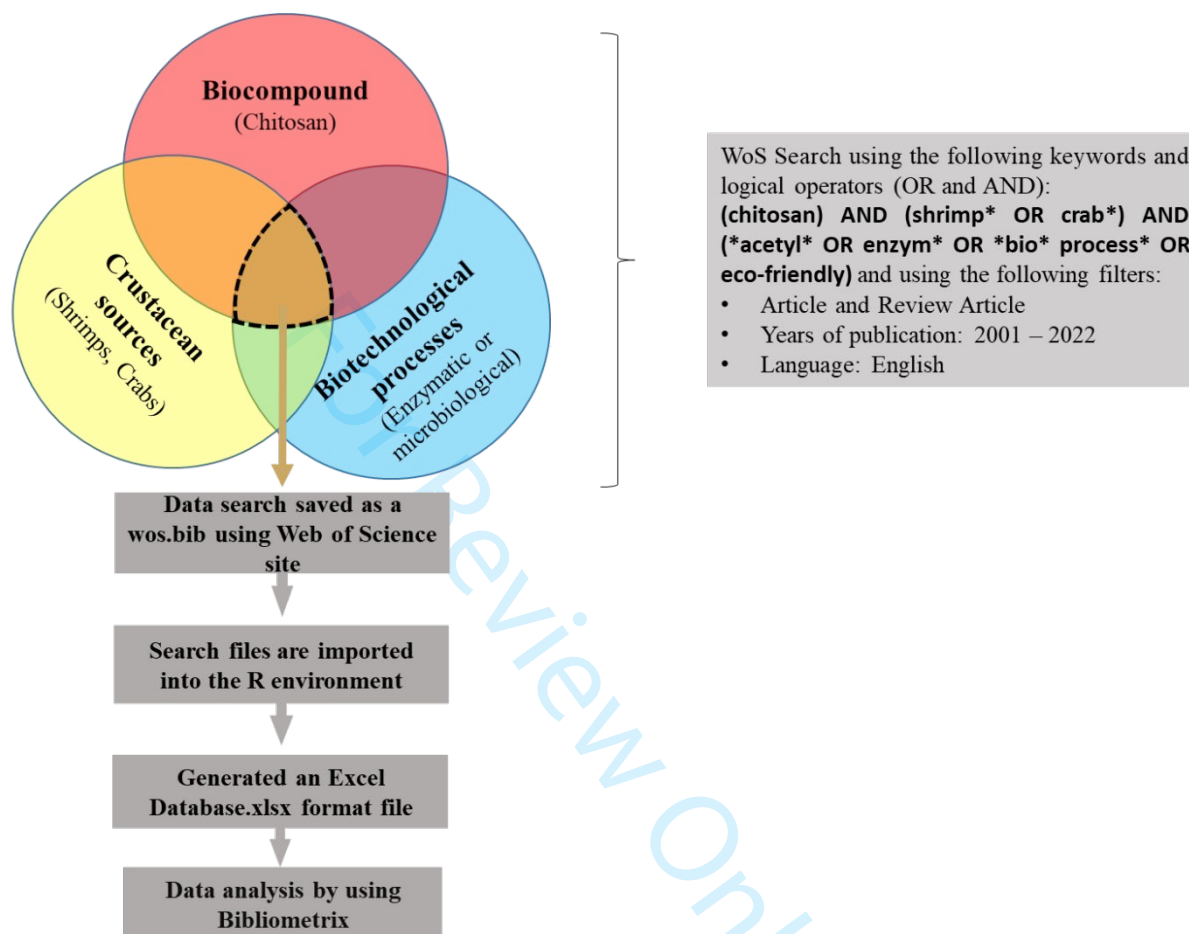


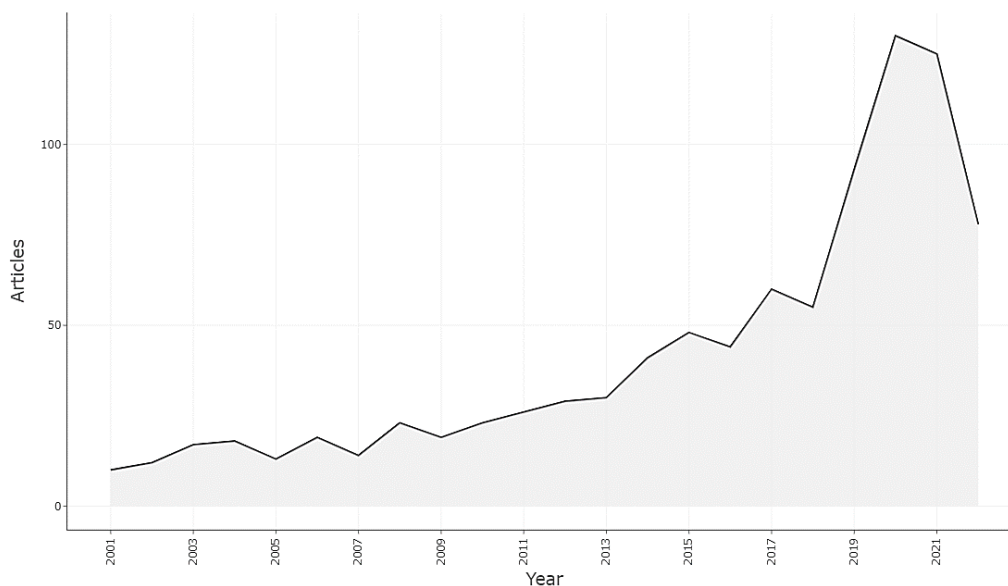
Figure 1. Flowchart methodology for searching the references in the WoS database.

### 3. Results and Discussions

#### 3.1 Mapping and bibliometric analysis

From the search strategy applied to the WoS database, 939 records were found, with 848 articles and 91 review articles being found and the evolution number of publications per year is shown in Figure 2. The scientific production, from the beginning of the study period until the year 2015 is low, with a publication rate below 50 articles per year. The significant

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4 increase in the number of articles published since 2016 shows that the scientific community's  
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6 interest in obtaining chitosan in a fully sustainable manner has gained a lot of interest.  
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29 Figure 2. Evolution of the annual scientific production.  
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33 Figure 3 lists the top 10 journals in terms of total number of publications collected in  
34 this study. The main journals are the *International Journal of Biological Macromolecules*,  
35 which has the highest number of publications with 65 publications, followed by *Carbohydrate*  
36 *Polymers* with 57 publications, and then *Polymers* with 22 publications. The *International*  
37 *Journal of Biological Macromolecules* is a well-established international journal on chemical  
38 and biological aspects of all natural macromolecules. *Carbohydrate Polymers* is a leading  
39 journal covering the study and exploration of polysaccharides with current or potential  
40 application in areas such as bioenergy, bioplastics, biomaterials, biorefining, chemistry, drug  
41 delivery, food, health. *Polymers* is a journal dedicated to publishing innovative and significant  
42 advances in Physics, Chemistry and Polymer Technology.  
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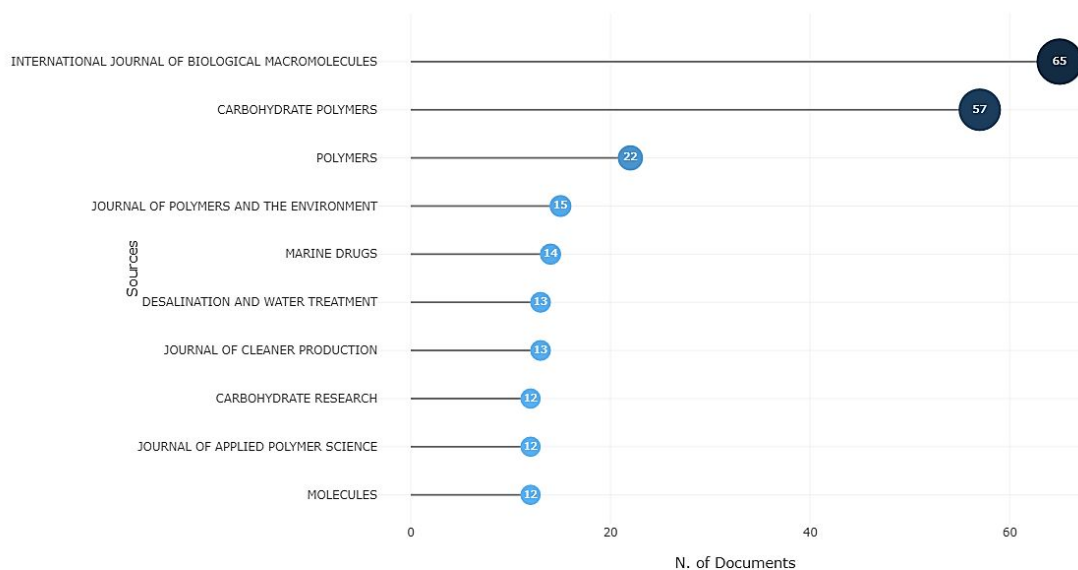


Figura 3. Most relevant sources of publication.

### 3.2 Keywords

In the 939 publications analyzed, 2,506 authors' keywords were found. Of this total, the 10 keywords of the most frequent authors are the following (number of occurrences in parentheses): chitosan (405), chitin (265), adsorption (49), deacetylation (40), chitinase (31), shrimp (30), characterization (26), shrimp shell (24), antioxidant (23) and crab shell (22), as depicted in Figure-4a. A higher frequency of the keywords chitosan and, obviously, chitin (the obtainment of chitosan preliminarily involves obtaining chitin), which are the biopolymers of interest. As for chitosan production, we can highlight the word deacetylation, purification and optimization. It is worth highlighting the presence of a large number of words related to the properties of chitosans or their applications (antimicrobial activity, molecular-weight, alpha-chitin).

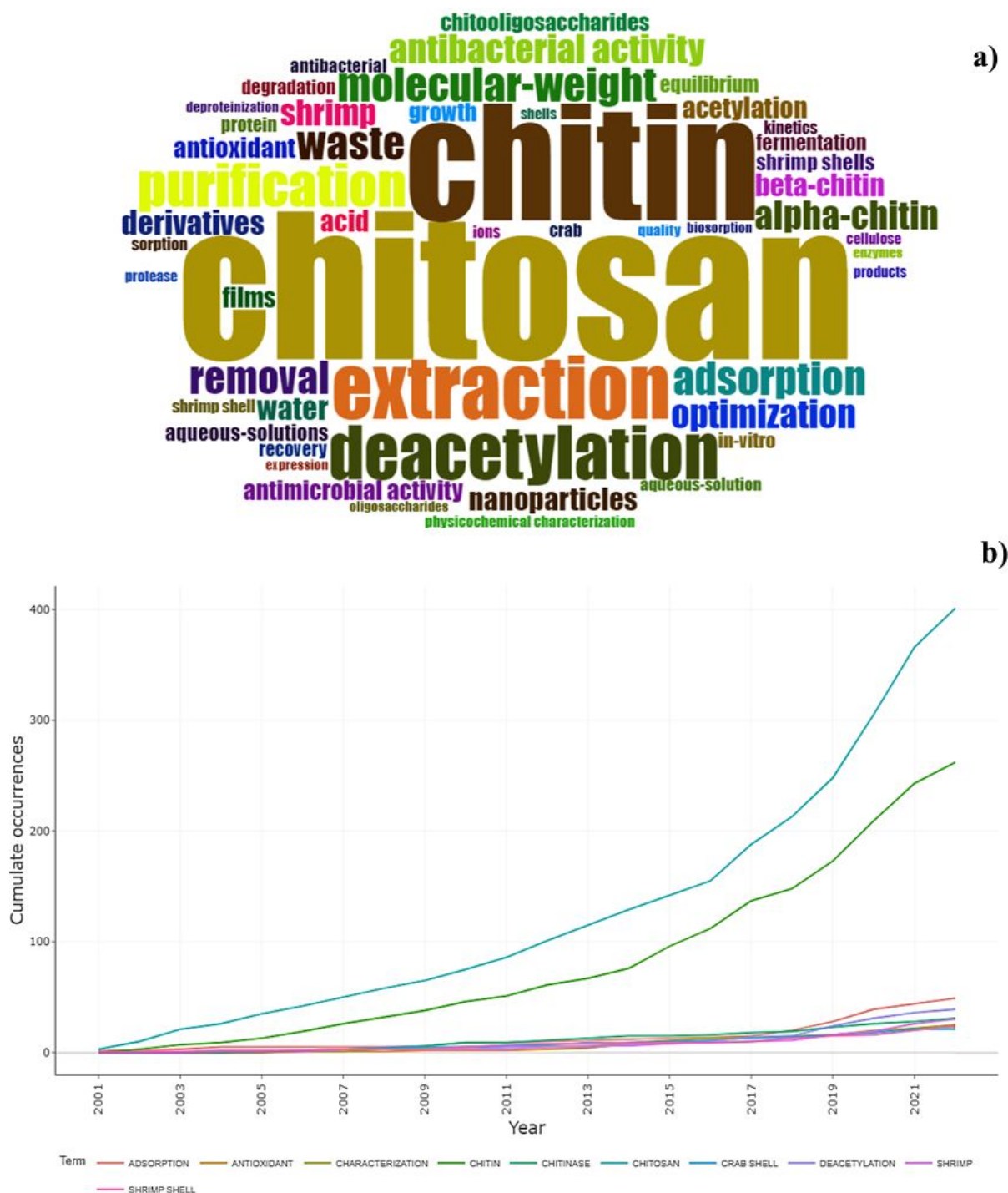


Figure 4. (a) Word cloud containing the keywords of the top 50 authors; (b) The most frequent keywords in the publications.

Figure-4b presents the word cloud map using author keywords. The word cloud represents the 50 most found keywords in the documents found in the search carried out in the

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4 WoS database and evaluated by the bibliometrix package. A word cloud highlights the most  
5 meaningful and important words in a word group. It could be a tool The word cloud represents  
6 the 50 most found keywords in the documents found in the search carried out in the Web of  
7 Science database and evaluated by the bibliometrix package. A word cloud highlights the most  
8 meaningful and important words in a word group. It may be an important tool to identify the  
9 frequency of keywords used by the authors in the study of processes for obtaining chitin and  
10 chitosan. The larger the word size, the greater the frequency used for the keyword.  
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### 23 3.3 Factorial analysis

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25 Figure-5a brings the map of the conceptual structure of the keywords, which is a map  
26 of the keywords using the multiple correspondence analysis (MCA) method. The MCA method  
27 of multiple correspondence analysis analyzes from groups of keywords that resemble each  
28 other, using strategies to unscramble the associations of keywords that can occur in small  
29 groups, from this, the formation of variable structures in two or three dimensions, in which the  
30 distance of the groups is related to the similarity between the keywords.  
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39 The importance of keywords is evidenced by their proximity to the central point of the  
40 cluster. Thus, according to Figure 5a, the occurrence of three clusters is evident, represented in  
41 the colors green, red and blue, considering the themes addressed in each cluster. The red cluster  
42 addresses topics related mainly to characterization, chemical properties, degradation, etc. The  
43 blue cluster encompasses themes related to technological application, since each and every  
44 study takes into account possible applications of the obtained product. The green cluster, in  
45 turn, encompasses words related to the topic of using bioprocesses on crustacean biowaste. The  
46 keywords that seem to receive the most attention currently are those close to the center point,  
47 i.e. the keywords addressed in the red cluster. Some keywords are far from each other, as well  
48 as the clusters, demonstrating that the research topic addressed is broad.  
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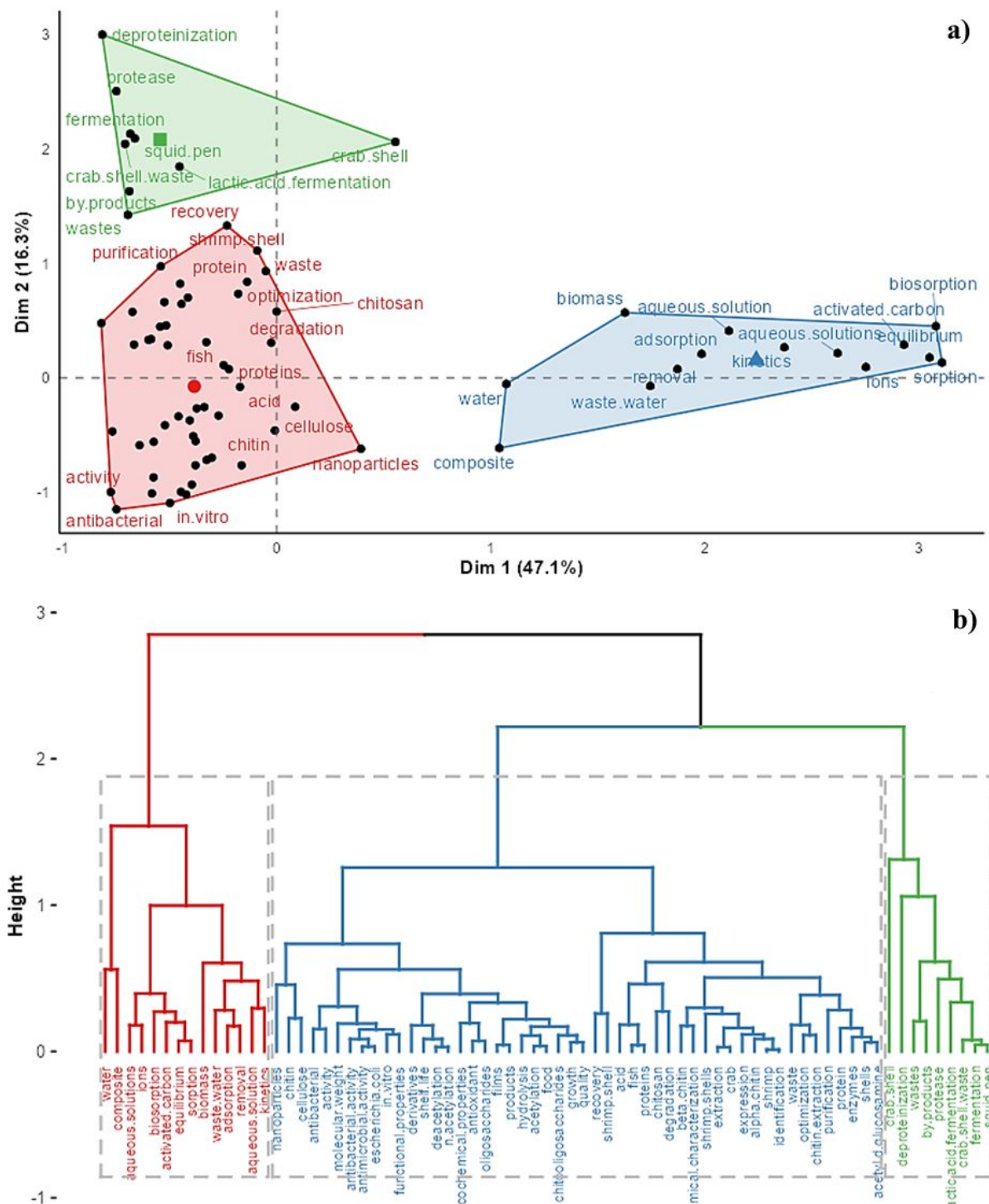


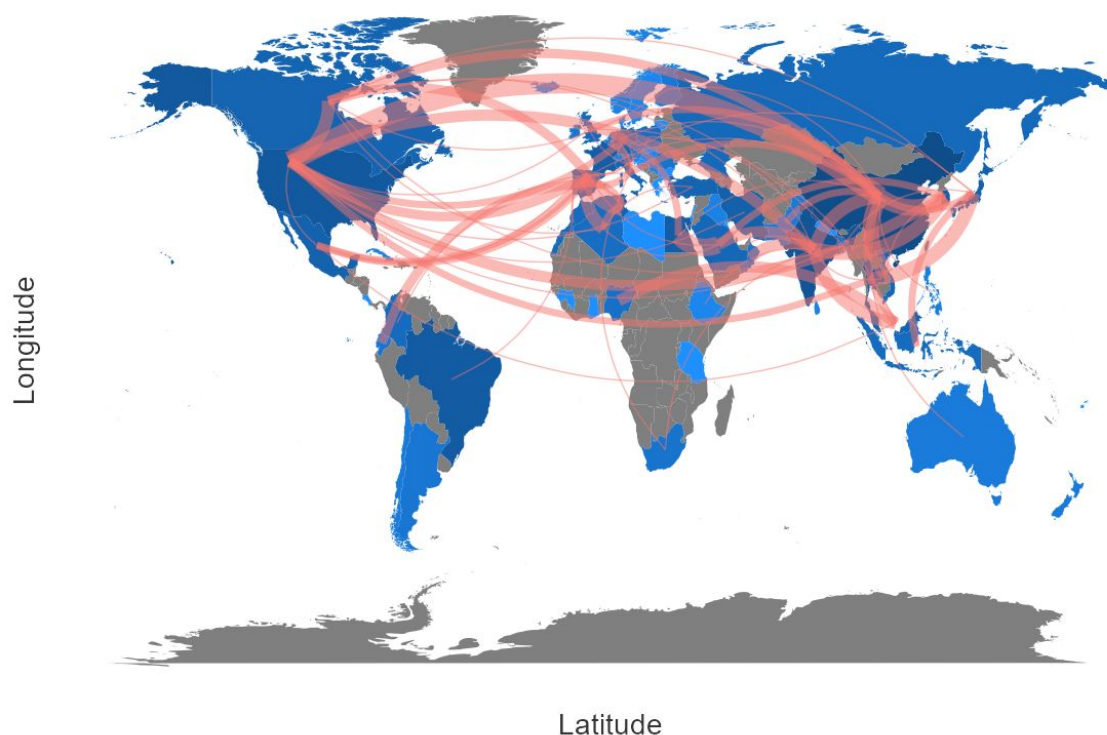
Figure 5. (a) Map of the conceptual structure of keywords; (b) Topic dendrogram concerning sustainable polymers.

The hierarchical analysis dendrogram, represented in Figure-4b, demonstrates the connection of keywords, next to each other in a hierarchical way. In the represented dendrogram, the keywords are separated into three large clusters of topics. Topics with the same height represent strong similarity to each other. Observing some topics closer to the zero axis,

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4 they are where a stronger connection of the keywords occurs. As an example, we can observe  
5 the keywords extraction and crab, shrimp and identification, deacetylation and acetylation and  
6 fish and proteins. It is interesting to note the connection of these keywords with the theme  
7 related to the production of chitosan from crustaceans in a chemical way, by deacetylation and  
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### 15 16 3.4. Social structure

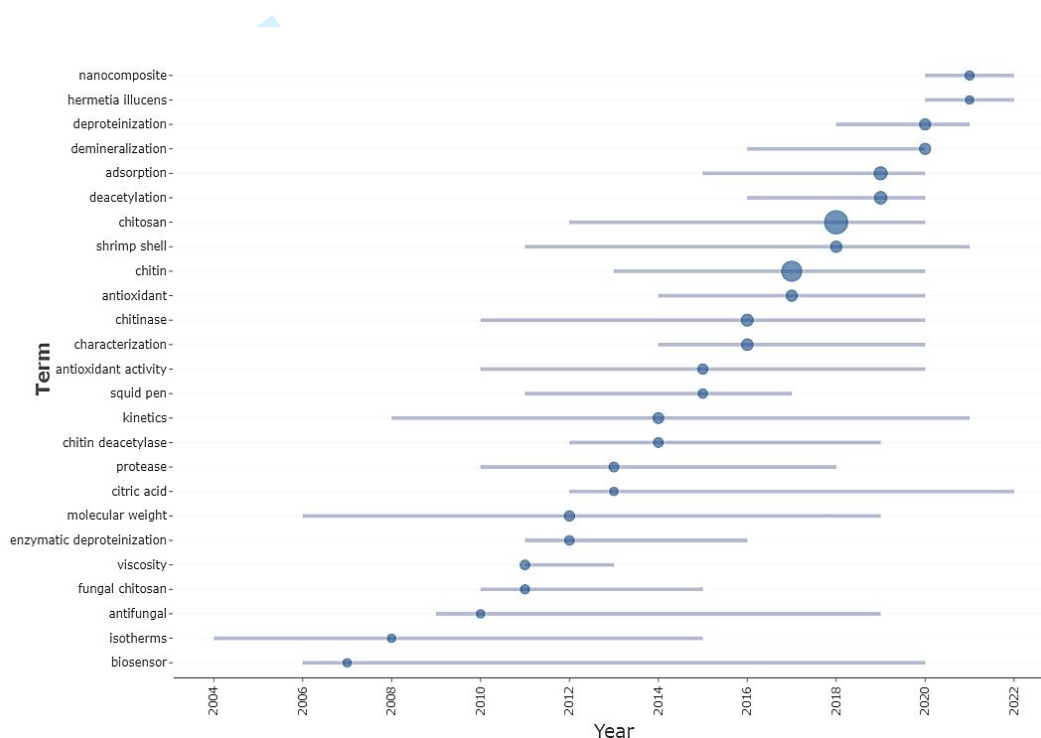
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18 Figure 6 displays a bibliographic network considering collaboration links between  
19 countries. The thicker these traits, the greater the collaboration between these countries. USA,  
20 China, Europe and Brazil appear as the countries with a lot of work, but Brazil shows little  
21 collaboration. China, USA and Europe appear better in terms of collaboration network, forming  
22 three clusters.  
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55 Figure 6. Author collaboration map by country.

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57 Figure 7 presents the trend in this field of research and the thematic evolution over the  
58 analyzed period. Some terms were kept in evidence for a long period, such as isotherms and  
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4 biosensor, and have lost their importance, while molecular molecular weight remains. Other  
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6 terms kept alternating their evidence for long periods, without considering much or little  
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8 importance in the research, such as protease, enzymatic deproteinization, antifungal and  
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10 viscosity. According to the size of the points, the terms most frequently found are chitin and  
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12 chitosan, as the basis of research in the years between 2016 and 2020. Currently, the terms most  
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14 frequently found are nanocompounds, deproteinization and demineralization, indicating that  
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16 these processes are still undergoing development towards eco-friendly processes.  
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44 Figura 7. Trend topics.  
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48 In Figure 8, Three-files plot (Sankey Diagram) shows which author's country are  
49 publishing more on a given topic and to which journal such topic is related more. We can see  
50 that deacetylation and molecular weight, processes responsible for, respectively, obtaining  
51 chitosan and its characterization, are being widely published by most of countries. It is also  
52 noteworthy that after the topics of the macromolecules (chitosan and chitin), the names of  
53 processes (extraction, purification and deacetylation) come in second place, followed by the  
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characterization of the molecular weight and then the applications (adsorption and removal) of substances .

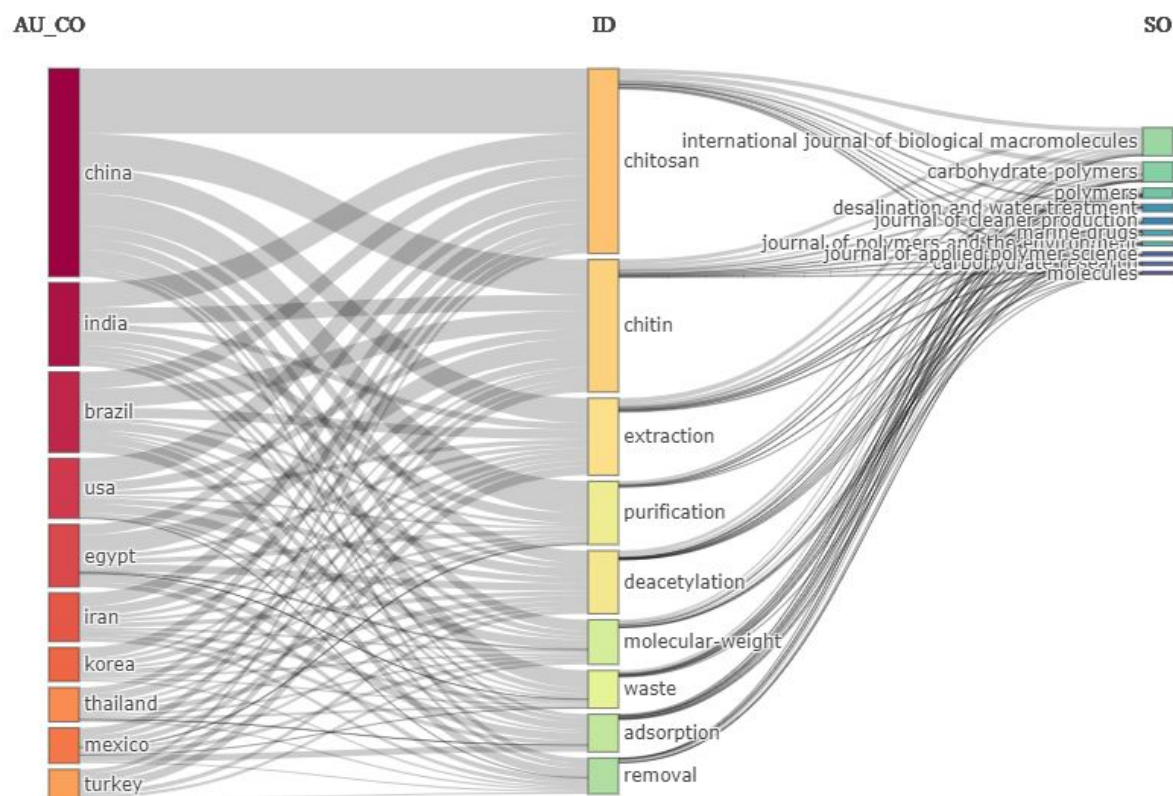


Figura 8. Three-fields plot for the relation among author's country (AU\_CO), Keyword Plus (ID) and source of publication (SO).

#### 4. Conclusions

With this bibliometric analysis it was possible to have an overview of the history, trend and forecast of studies for obtaining chitosan in an environmentally friendly and sustainable way. It was also evident that major developments have been to obtain chitin and not chitosan sustainably. Research has actually mitigated the use of techniques that are harmful to the environment. The survey also revealed that some regions need to collaborate more, such as Brazil, which, although it has publications in this area, needs to exchange experiences with other research centers in the world.

## 5. Author's Contribution

**Dielson da Conceição Emerique:** Conceptualization, Data curation, Formal analysis, Writing original draft, Formal Analysis

**Lorena Batista de Moura:** Writing original draft

**Evaldo martins da Silva:** Conceptualization, Supervision

## 6. Acknowledgments

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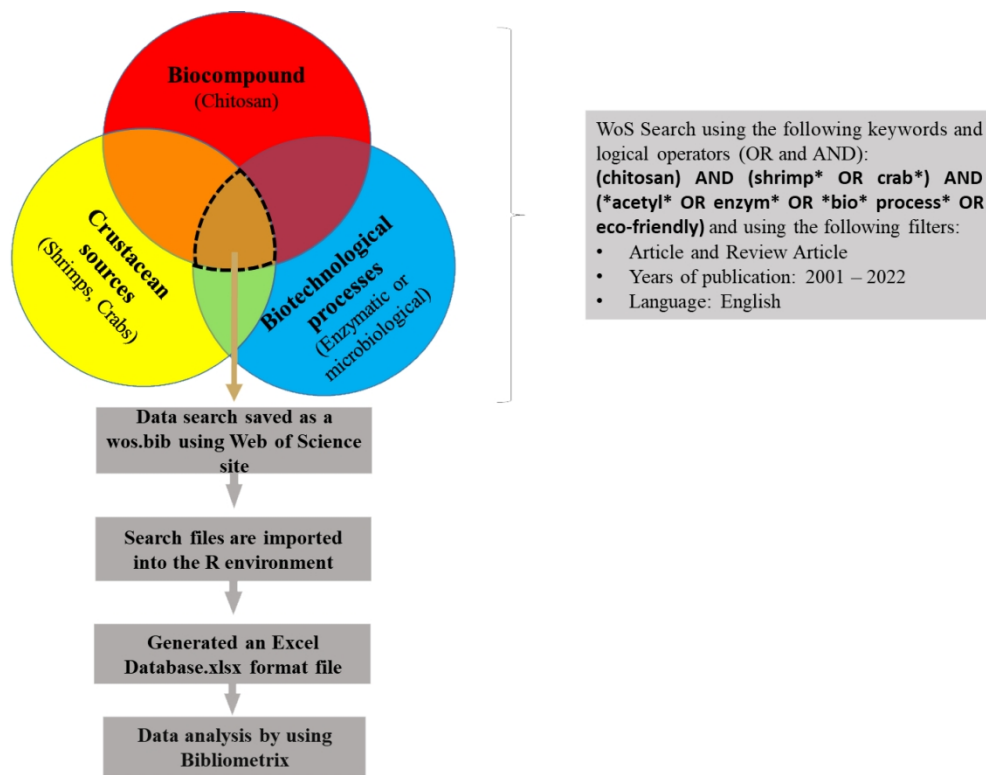
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Figure 1. Flowchart methodology for searching the references in the WoS database

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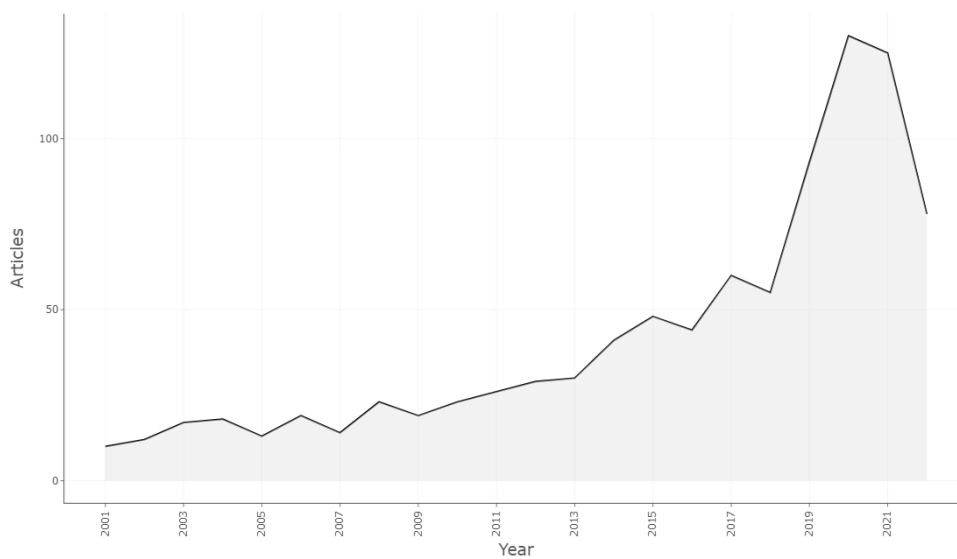


Figure 2. Evolution of the annual scientific production.

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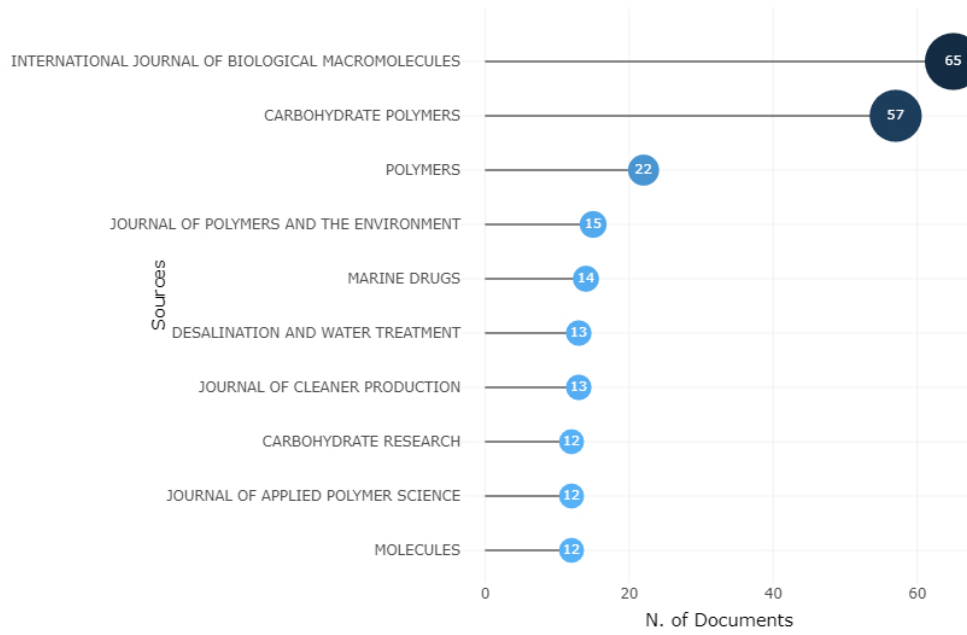


Figure 3. Most relevant sources of publication

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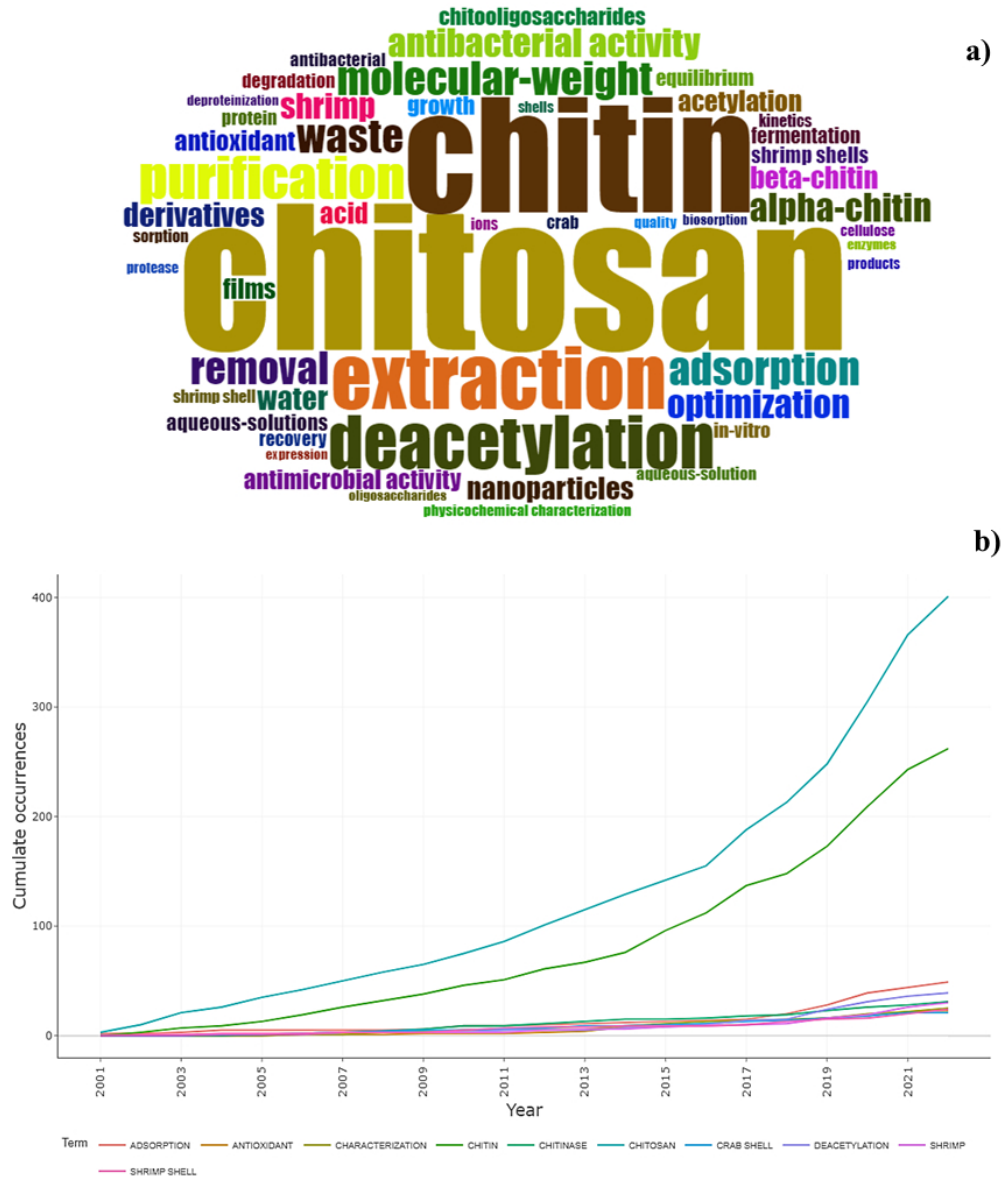


Figure 4. (a) Word cloud containing the keywords of the top 50 authors; (b) The most frequent keywords in the publications.

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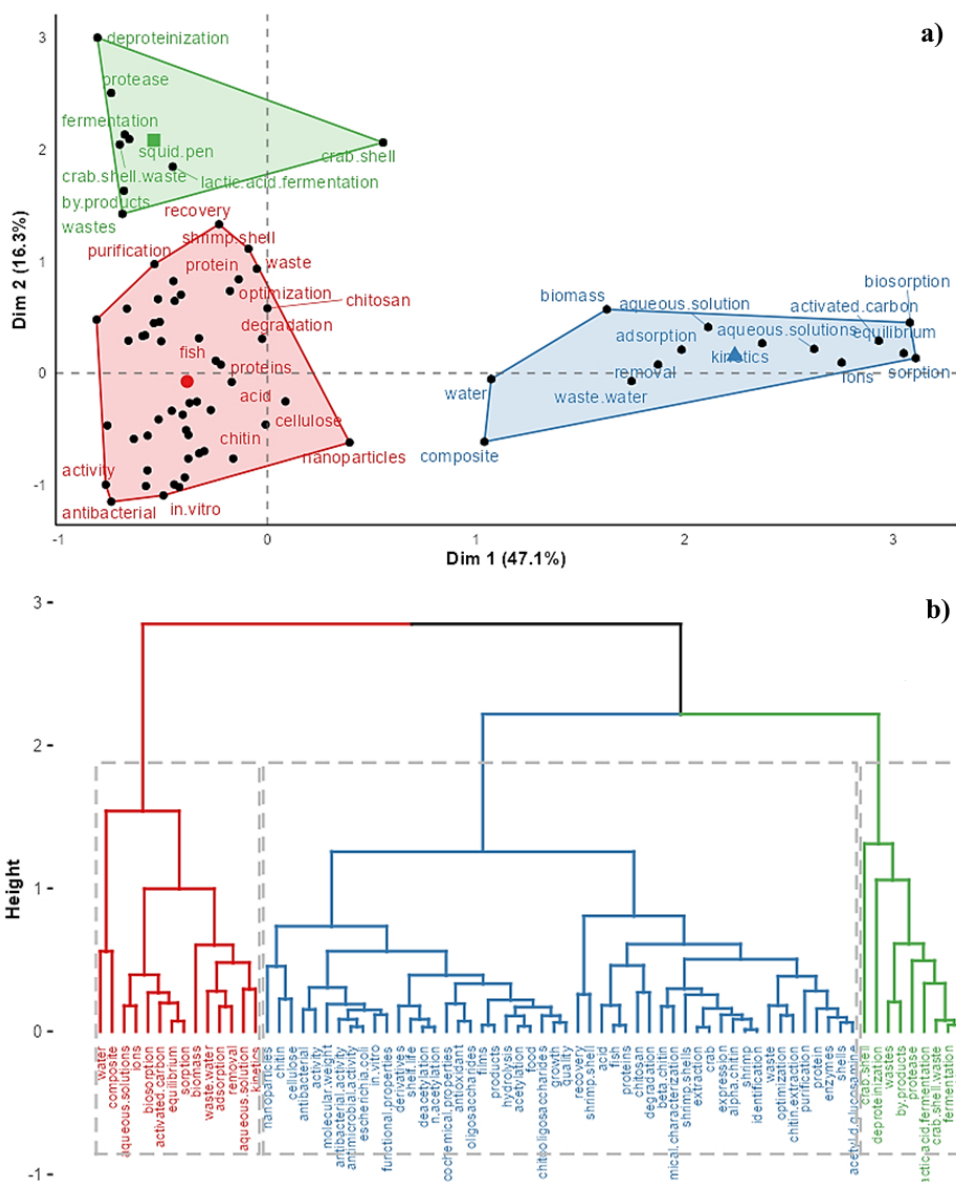


Figure 5. (a) Map of the conceptual structure of keywords; (b) Topic dendrogram concerning sustainable polymers.

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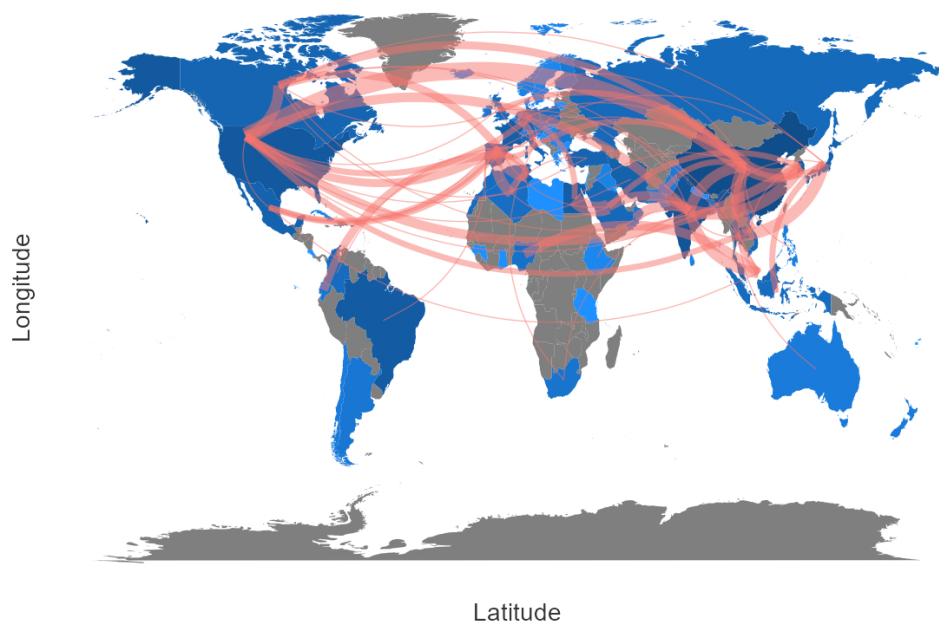


Figure 6. Author collaboration map by country.

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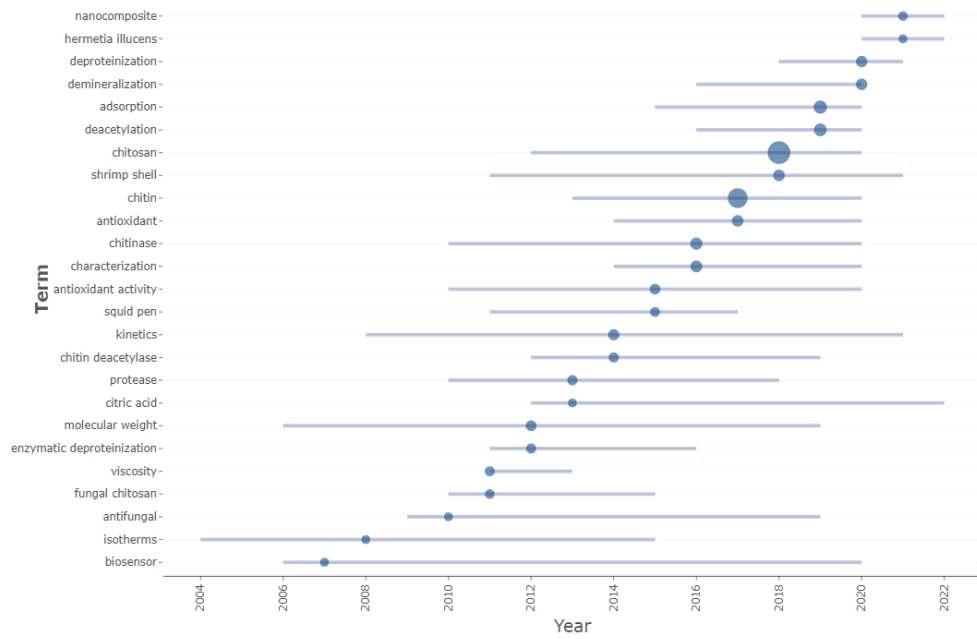


Figure 7. Trend topics

286x190mm (96 x 96 DPI)

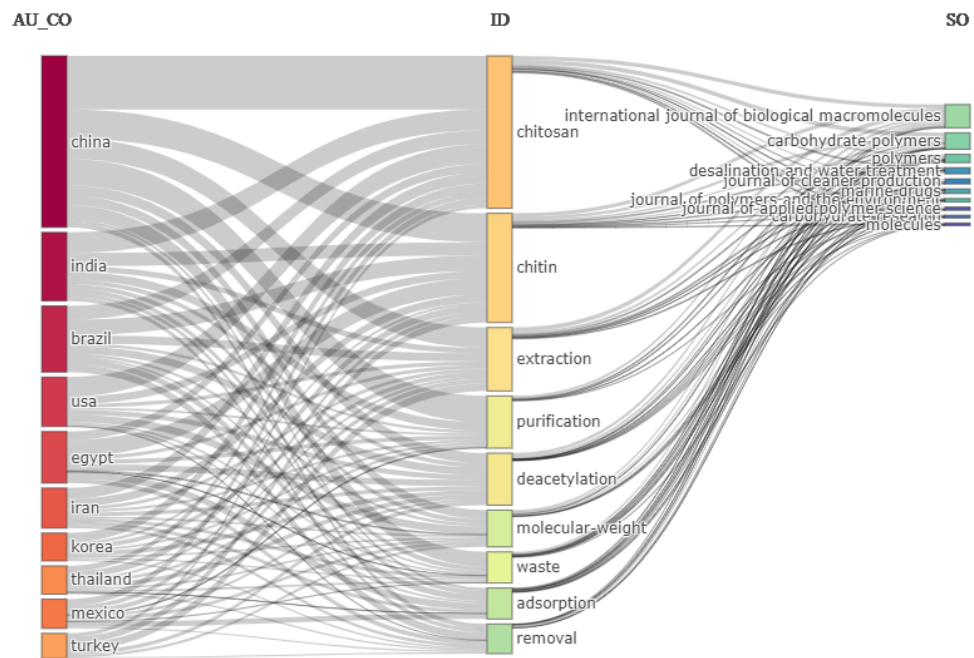


Figura 8. Three-fields plot for the relation among author's country (AU\_CO), Keyword Plus (ID) and source of publication (SO).

532x369mm (38 x 38 DPI)