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#### **Abstract**

This work aims to provide a comprehensive study of the available research information on pesticide residues in honey through literature analysis. The research advancements within this research field from 1948 to 2019 are addressed using the Web of Science database. The results from the 685 articles analyzed indicate that this research field is in the focus of interest nowadays (Price index: 47.5 %). The yearly production increased steadily from 2001 on, and authors, journals, and institutions followed Lotka's law. On the other hand, Pico, Y (Spain) (2.5 %), Journal of Chromatography A (5.8 %), the USA (15.0 %) and Agricultural Research Service (USA) (4.0 %) were the most productive author, journal, country and institution, respectively. The research hotspots of this field, according to keyword analysis, are related to the chromatographic techniques for the determination of pesticides such as imidacloprid, neonicotinoids, or coumaphos in honey and derivate products such as propolis and wax.

**Keywords:** Bibliometric review; Honey; Honey-bees; Pesticide; Residues

### **1. Introduction**

Pesticides are a category of toxic chemicals that are formulated to kill or repel pests or to interrupt their reproduction (Gilden et al., 2010; Sabarwal et al., 2018), including insecticides, acaricides, algicides, herbicides and fungicides, and are some of the most toxic, environmentally stable and mobile substances in the environment (Regueiro et al., 2015). Insecticides and acaricides kill insects and mites by disrupting their neuronal activity, molting process or other specific metabolism mechanisms of these arthropods. Algicides and herbicides kill plants and algae by disrupting photosynthetic capacities or the synthesis of essential organic compounds. Fungicides kill fungi by inhibiting, for example, the formation of cell membranes in such organisms (Sanchez-Bayo and Goka, 2016). The destructive rate of pesticides on body organs and tissues depends on the dosage, mode of contact, biological changes and the associated metabolites supply (Jorsaraei et al., 2014).

Honey can be defined as a natural, complex, and sweet food produced by honey bees (*Apis mellifera* L.) which can be used directly by humans with no further processing (Osman et al., 2020). Honey bees collect the nectar of flowers or from secretions of living parts of plants as well as from excretions of plant-sucking insects on the living part of plants. These collected products are transformed and combined by honey bees and then they are deposited for further dehydration and storage in honeycombs for final ripeness and maturing (Kahraman et al., 2010; Shafiee et al., 2013). Honey is a natural product composed by an aqueous supersaturated sugar solution, it is an excellent source of energy, and possess antibacterial, prebiotic, antioxidant and antimutagenic properties. This substance contains 80 % of carbohydrates, mainly glucose (35 %), fructose (40 %), and sucrose (5 %), 20 % of water, and more than other 180 minority components including amino acids, proteins, organic acids, lipids, waxes, vitamins, minerals, aroma compounds, flavonoids, pigments, pollen grains and enzymes (Geană et al., 2020; Kahraman et al., 2010). The composition of honey depends on several factors including the type of flowers and climatic conditions in which the plants grow and mature close to beehives where honey bees forage (Osman et al., 2020).

As in the case of other natural products, European regulations (European-Commission, 2006) have been designed to strictly control the possible presence of chemical residues in honey (Al-Waili et al., 2012; Jamal et al., 2020). Therefore, hygienic standards and measurements to guarantee residue-free substances should be maintained during the whole honey production and processing chain (Jamal et al., 2020). It is known that honey can contain compounds classified as harmful to human health such as pesticides (Crenna et al., 2020; Raimets et al., 2020), which also influence the honey bee colonies. In this sense, to date, residues of up to 173 different compounds, such as HCH, DDT, carbaryl, or coumaphos (Blasco et al., 2003; Cervera-Chiner et al., 2020; Del Carlo et al., 2010), have been found in beehives (Sanchez-Bayo and Goka, 2014, 2016).

Pesticides can reach honey through various routes including the exposure of honey bees to agricultural pesticides contained in water, pollen nectar, dust-spray droplets and guttation drops, and pesticides used by beekeepers for the control of parasites in the beehives (Sanchez-Bayo and Goka, 2016). Pesticide risk to honey bees can be reduced by spraying the crops in the evening since honey bees forage takes place between sunrise and about an hour before sunset, all within a 2–4 km radius from their beehives. Nevertheless, they can travel as far as 7 km or more when their local sources are scarce (Beekman and Ratnieks, 2000; Sanchez-Bayo and Goka, 2016). Due to the body of honey bees is covered with hairs, they can capture atmospheric pesticide residues such as dust-spray droplets, which can be taken back to the beehives (Martinello et al., 2020). Pesticide residues in pollen, nectar and guttation drops are also taken by the forager

bees to their colonies and remain in the beebread and honey for quite some time. Honey bees drink from puddles, irrigation ditches, ponds and streams, and if these waters are contaminated with pesticide residues, the forager bees can ingest these compounds as well (Sanchez-Bayo and Goka, 2016). The use of chemicals by beekeeper to control parasites infesting *A. mellifera* such as virus or mites is a common practice in beekeeping; therefore, those pesticides can also appear as contaminants in honey (Jamal et al., 2020).

It is widely known that the massive loss of honey bee colonies is an ongoing worldwide issue caused, among other reasons, by the extensive use of pesticides (Calatayud-Vernich et al., 2016). Honey bee products can also be used as indicators for environmental contamination because honey bees gather raw material from both wild plants and crops throughout the foraging season. Thus they serve as a means for effective environmental sampling (Raimets et al., 2020). In this sense, it has been reported that the most frequently detected pesticides in death honey bees are imidacloprid, chlorpyrifos, thiachloprid, chlothianidin, and thiametoxam. For its part, the most common pathogens responsible for infection are Deformed Wing Virus, Acute Bee Paralysis Virus, Chronic Bee Paralysis Virus, *Nosema ceranae* and in *Varroa* mite (Martinello et al., 2020).

On the other hand, bibliometric studies are based on the research methodology employed in the science of information and library, and it includes a series of qualitative, quantitative and visual procedures to generalize the patterns and dynamics of publications (Yaoyang and Boeing, 2013). Specifically, the bibliometric technique is a useful method to access the development, growth trend and research trends of a specific research area (Ma et al., 2018). Bibliometric studies have attracted growing interest over the last years; in fact, several bibliometric analyses have been recently

performed in many different research fields (Andreo-Martínez et al., 2020a; Andreo-Martínez et al., 2020b; Ma et al., 2018; Olisah et al., 2019; Sinha, 2012; Zheng et al., 2015; Zhi and Ji, 2012). Therefore, due to the lack of bibliometric reviews on pesticide residues in honey, this work aims to carry out a descriptive bibliometric analysis on pesticide residues in honey from inception to 2019, in order to provide updated information on how this field has evolved over the time and publication framework and to try to identify the different research topics in the field.

#### **2. Method**

The scientific output information of the present bibliographic study was obtained from the Science Citation Index Expanded (SCI-E), Web of Science© (WOS). The temporary search period chosen was from inception to 2019 and the search in WOS database was performed in February 26th, 2020. The Boolean strings selected were: theme: ("pesticide residue\*") *AND* theme: (honey\*) in Web of Science Core Collection database.

Microsoft Excel (v. 2019) was used to analyze publication characteristics, and the frequency analysis was performed using BibExcel (Persson et al., 2009). Network diagrams for cooperation analysis were generated using Pajek64 5.08 (Batagelj and Mrvar, 2004) and VOSviewer software (van Eck and Waltman, 2013). VOSviewer software was also used to perform the co-occurrence of all keywords. Journal Citation Reports (JCR) Science Edition 2018 was used to obtain the journal impact factor (IF) (Clarivate\_Analytics, 2018).

The bibliometric parameters used to analyze the articles related to pesticide residues in honey during the last 71 years were those reported elsewhere (Andreo-Martínez et al.,

2020a; Andreo-Martínez et al., 2020b): type and language of documents; publication characteristics (total publications, authors number, average authors number per article, number of references cited, number of references per article, number of pages per article, and average number of pages per article); performance of authors, journal publication patterns; WOS categories; countries and institutions publication patterns; collaborations of authors, countries and institutions analysis; Lotka's law, Price's index; h-index of authors, journals, countries and institutions using WOS records; and research trends obtained by the co-occurrence analysis of all keyword.

#### **3. Results and discussion**

# **3.1. Publications patterns and characteristics**

After excluding possible duplicates, 795 publications related to pesticide residues in honey, published between 1948 and 2019, were identified in the WOS database. Among the 795 publications, 685 (86,2 %) were articles, followed by reviews (52; 6,5 %), proceeding articles (20; 2,5 %), proceeding papers (10; 1,3 %), editorial material (9; 1,1 %), notes  $(6; 0.8 \%)$ , meeting abstracts  $(5; 0.6 \%)$ , book chapters articles  $(3; 0.4 \%)$ , corrections  $(2; 0.3 \%)$ , book chapters reviews  $(2; 0.3 \%)$  and reprints  $(1; 0.1 \%)$ . There were about 6 articles per review and about 34 articles per proceedings paper. As can be observed, the majority of the publications were articles published in scientific journals; therefore, only 685 articles were selected for further analysis.

Among the 685 articles, 636 (93.1 %) were published in English, followed by German (17; 2,5 %), Chinese (7; 1,0 %), French (6; 0,9 %), Italian and Japanese (5; 0,7 %), Polish and Hungarian  $(3; 0.4 \%)$ , Spanish  $(2; 0.3 \%)$  and Lithuanian  $(1; 0.1 \%)$ .

The works of Eckert (1948) and Metcalf (1948) were found to be the earliest articles indexed in WOS related to pesticide residues in honeybees. This author first related the effects that certain pesticides such as DDT or DFDT can pose on honeybees, already highlighting in the 1950s the damage that the extensive use of pesticides could cause to honeybees.

As can be observed in Fig. 1A, the number of publications related to the presence of pesticide residues in honey increased from 2 in 1948 to 101 and 77 in 2018 and 2019, respectively, and the number of articles increased from 2 in 1948 to 85 and 68 in 2018 and 2019, respectively. From 2001, the number of publications grew quickly, as reported elsewhere for several research fields (Andreo-Martínez et al., 2020b).

The cumulative number of articles per year from 1948 to 2019 is shown in Fig. 1B. The data were fitted to a four-order polynomial model with a high determination coefficient  $(R^{2} = 0.994)$ , with no apparent influence of the slight decrease in the number of published articles observed in 2019. Using this model, it can be predicted an increase in the number of articles related to pesticide residues in honey research in coming years; specifically in 2022 and 2025, the articles in the field could account more than two and two and half times (1420 and 1774, respectively) those published in 2019 (685). This type of analysis has been proved to be a reasonably effective tool for the prediction of the number of future publications in a given research field (Andreo-Martínez et al., 2020a).

**Fig. 1**

The characteristics of the articles related to pesticide residues in honey during the last 71 years are shown in Table 1. As discussed before, there was a remarkable growth in the number of articles per year from around 2001. The mean number of authors per article increased by 590% from 1.0 in 1948 to 5.9 in 2019. The mean number of references cited per article increased by 566 % (from 8.5 in 1948 to 48.1 in 2019) and the average article length was 7.3 pages.

#### **Table 1**

On the other hand, it can be highlighted that although the first work in the field was published 71 years ago, it is still in the focus of interest as shown by the Price's index value of this research area, which was found to be 47.5 %. It is known that when this index is close to 50.0 %, theat current literature (less than 5 years old) is more abundant and the topic remains relevant and it is actively being researched (Andreo-Martínez et al., 2020a; Andreo-Martínez et al., 2020b; Price, 1965).

#### **3.2. The feature of authors**

In order to perform this analysis, the name of the authors was conscientiously standardized by hand (i.e. Huidobro, JF by Huidobro-Canales, JF; Melgar, MJR by Melgar, MJ or del Nozal, MJ by Nozal, MJ). The 685 articles of the present bibliometric analysis were signed by 2328 different authors. Among those 2328 authors, 1838 (79.0 %) appeared in one article, followed by 313 (13.4 %) in two, 89 (3.8 %) in three, 46  $(2.0\%)$  in four, 18 (0.8 %) in five, and 9 (0.4 %) appeared in six articles.

The highest number of authors in one article was nine (1; 0.1 %), followed by thirteen (3; 0.4 %), twelve or eleven (4; 0.6 %) and ten (5; 0.7 %). Thirty-two articles (4.7 %) displayed a single author. The most common number of authors per article was four (128; 18.7 %), followed by five (127; 18.5 %), three (101; 14.7 %) and two (88; 12.8 %). Seventy-seven (11.2 %), 60 (8.8 %), 34 (5.0 %) and 21 (3.1 %) articles were authored by six, seven, eight and nine authors, respectively.

### **Fig. 2**

Fig. 2A shows the model used to calculate the Lotka's Law for authors and the number of articles published in the field  $(X^{3.207} \text{·Y} = 2638.6)$ , providing a good correlation coefficient ( $R^2$  = 0.9749) in the range of one to nine authors (only 0.2 % of the 2328 authors published more than nine articles). It can be observed that this research field follows the Lotka's Law for authors as the exponent found in the power model was higher than 1.2 and lower than 3.5 (Andreo-Martínez et al., 2020a; Andreo-Martínez et al., 2020b).

# **Table 2**

The top 10 authors in the field, according to their h-index, are shown in Table 2. As a note, 23 articles lacked the RP field; therefore, the rank of RP authors considered was based on 662 articles. Taking into account the time that an author was cited as reported by the WOS core collection times cited count, Poco, Y. had the highest h-index (14),

followed by Bernal, JL (10) and Nozal, MJ (9). Those three authors also offered the highest number of articles ( $2.5\%$ ),  $16$  ( $2.3\%$ ), and  $15$  ( $2.2\%$ ), respectively. As can be observed, the mentioned authors are from the same institution (University of Valencia, Spain), highlighting the leading role of this institution in the research field of pesticide residues in honey. Among the 10 most productive authors, the author who appeared more often as RP was Bernal, J (8), followed by Pico, Y (7), and Jimenez, JJ and Tadeo, JL (4); and Bernal, JL, Thrasyvoulou, A, and Bernal, J appeared 2 times as FP.

### **Fig. 3**

A total of 1530 collaborations were found in the analysis of author collaborations. Fig. 3 shows collaboration networks for frequencies equal to and higher than 3 times. The most common collaboration number was 1 (1371; 89.6%), followed by 2 (118; 7.7 %) and 3 (23; 1.5 %). The greatest number of author collaboration was 13 (0.1%): Bernal, JL and Nozal, MJ; followed by 9 (0.1 %): Nozal, MJ and Bernal, J, and Bernal, JL and Bernal, J. As discussed before, those authors were also among the most productive and had the highest h-index.

### **3.3. The feature of journals and Web of Science categories**

The analysis of journals showed that a total of 187 different journals published at least one article in the research field of pesticide residues in honey. Table 3 shows the top 10 productive journals ordered by the total number of articles published in the field from  to 2019. The top 10 journals published 33.6 % of the 685 articles and seven of them are specialized in the research field. Journal of Chromatography A ranked first with 40 (5.8 %) articles published, followed by Plos One (32; 4.7 %) and Journal of Economic Entomology (28; 4.1 %). The highest h-index value (28) was also shown by the Journal of Chromatography A, followed by Plos One (20); however, the Journal of Economic Entomology (12) ranked in fifth place. Science of the Total Environment Journal had the highest impact factor (5.589), followed by Talanta journal (4.916), and Journal of Chromatography A (3.858). Regarding the position of the top 10 journals by quartiles, 6 (60.0 %) were Q1, 3 (30.0 %) Q2, and 1 (10.0 %) Q3.

# **Table 3**

The annual publication pattern for the top 6 most productive journals is shown in Fig. 4A. The increase in the number of publications was progressive from 1948 to 2004, showing a sharp increase from 2005 to 2017 when the maximum was reached (15).

# **Fig. 4**

The analysis of WOS categories was performed on 684 articles as one article lacked the WOS category field. The 684 articles were published in 52 different categories of the WOS. The top five WOS categories from 1948 to 2019 are shown in Fig. 5. Chemistry, Analytical contributed with 160 articles, followed by Entomology (143), Food Science & Technology (135), Environmental Sciences (131), and Biochemical Research Methods (78). Entomology was the leading category until 1994; however, it was surpassed by Food science and technology in 1995. Environmental Sciences was the leading category for the last two years followed by Chemistry, Analytical, and Food Science and Technology. One hundred (53.5 %) journals published only one article in the field, which denotes the transversal nature of the field. Twenty-nine (15.5 %), 15  $(8.0 \%)$ , 2 (1.1 %), 5 (2.7 %), 7 (3.7 %) journals published two, three, four, five and six articles, respectively. In this sense, Fig. 2B displays the power model to calculate the Lotka's Law for the journals and the number of articles published in this research field, showing a correlation coefficient of 0.7687 in the range of one to six journals (15.5 % of the 187 journals published more than six articles). Although this correlation coefficient was not very good, the journals and the number of articles published in this research field followed the Lotka's Law, with the exponent found in the power model of 1.833. Therefore, it can be said that the number of journals that will publish articles in the pesticide residues in honey research field will increase as the number of articles also rises (Andreo-Martínez et al., 2020a; Andreo-Martínez et al., 2020b).

# **Fig. 5**

#### **3.4. The feature of countries and institutions**

Among the 685 articles found in the present bibliometric analysis, 622 were chosen for the analysis of institutions and countries since 63 articles lacked the author affiliations. Besides, Fed Rep Ger was changed by hand by Germany.

# **Table 4**

Seventy different countries from five continents published at least one article on pesticide residues in the honey research field, with Australia being the continent with the fewest articles published  $(6; 1.0 \%)$ , followed by Africa  $(21; 3.4 \%)$ . Twenty-six (37.1 %) countries published more than 5 articles, and 4 (5.7 %) countries more than 50. The highest number of countries in one article was 9 (Bonmatin et al., 2015), followed by 5 in two articles, 4 in one, three in 10, two in 80 and one in 528 articles. As can be observed, the majority of the articles were single-country publications. The 20 most productive countries, ordered by TP, are shown in Table 4. Top 20

countries contributed to more than 99 % of the total publications of the topic. The USA ranked first with 103 (15.0 %) articles, followed by Spain (84; 12.2 %), China (66; 9.6 %), Italy (55; 8.0%) and France (49; 7.1 %). The USA also had the highest h-index value (28), followed by Spain (27), Italy (23), France (21) and UK (17). It is known that the USA and China are the leaders in publishing on several research fields (Andreo-Martínez et al., 2020a; Andreo-Martínez et al., 2020b). However, although it has been reported that China was the most productive country in the bibliometric study on organochlorine pesticides research in biological and environmental matrices from 1992 to 2018 (Olisah et al., 2019), followed by the USA, China does not have a dominant role in this field and, in this sense, it will be difficult to surpass the USA shortly.

The annual publication patterns of the top 5 productive countries are shown in Fig. 4B. The USA was the only country that contributed until 1983 and the growth in the number of articles published discussed in Fig. 1A can be also observed in Fig. 4B.

At an international level, the most collaborative countries are USA (25; 26.5 %), followed by France (21; 22.3 %), Italy (20; 21.2 %), and Spain (16; 17.0 %) which are, except China, the ones with the highest number of publications. The top 6 productive countries were also the ones with the highest number or articles as both the first and corresponding author. Poland and Iran did not publish articles in collaboration with other countries.

A total of 111 collaborations was found in the co-occurrence analysis of country collaboration and the country collaboration networks for frequencies equal to and higher than 3 is shown in Fig. 6A. The most common collaboration number was 1 (80; 72.1 %), followed by 2 (17; 15.3 %) and 3 (7; 6.3 %). Spain-Italy collaborations ranked first (8), followed by USA-Canada (7), and USA-China and USA-France (5). The country with a higher number of international collaborations is France (18), followed by the USA (17) and Italy (17), Germany (15), and Canada and UK (12). Therefore, it can be stated that the research on pesticide residues in honey was centralized in Europe, followed by America.

# **Fig. 6**

The 622 articles selected in this analysis represented 742 different institutions of which (44.7 %) were single-institution articles. 547 (73.7 %) had only one article related to the research field, 102 (13.7 %) two articles, and 36 (4.8 %) three articles. The highest number of institutions in one article was 12 (1), followed by 8 (1) and 7 (2). Fig. 2C shows the Lotka's Law for the institutions and the number of articles published in the pesticide residues in honey research field, showing a good correlation coefficient (0.9732) in the range of one to eight journals (only 1.3 % of the 742 institutions published more than eight articles). Therefore, it can be said that the number of institutions implicated in this research field will increase as the number of articles increases.

Table 5 shows the 20 most productive institutions performing research on pesticide residues in honey from 1948 to 2019 where Agricultural Research Service (ARS) was the leader with 25 articles published, followed University of Valencia (18), University of Valladolid (14) and University of Bologna (13). Of note, ARS also was the most productive institution in the bibliometric study on global biopesticide during 1996–2008 (Sinha, 2012), and in a different research field such as biofuel until 2012 (Yaoyang and Boeing, 2013), highlighting the multidisciplinary nature of ARS and the important role of this institution and the USA in the field.

# **Table 5**

The University of Valencia had the highest h-index value (15), followed by the University of Bologna (12), and ARS and Instituto Nacional de Investigación y Tecnología Agraria y Alimentaria (10). The University of Bologna, Institut national de la recherche agronomique (INRA), Centre national de la recherche scientifique (CNRS), Purdue University, University of Milan and Ministerio de Agricultura published all their articles together with other institutions. The USA was the country with the highest number (6) of institutions in the top 20 productive institutions, followed by Spain (5), France, Italy and China (2), and Canada, Poland, and Greece (1). As discussed before, this fact can corroborate the dominant role of Europa in the research field, followed by America, which can be explained by the advanced food security policies of the European Union and the USA.

A total of 854 collaborations was found in the co-occurrence analysis of institution collaboration and the institution collaboration networks for frequencies equal and higher than 3 is shown in Fig. 6B. The most common collaboration number was 1 (785; 91.9 %), followed by 2 (51; 6.0 %) and 3 (11; 1.3 %). ARS-University of Tennessee collaborations ranked first (6), followed by University of Valencia-University of Bologna, and INIA-University of Almería (5). ARS also collaborated four times with North Carolina State University and Penn State University. The institution that has collaborated with most institutions is ARS (33), followed by the CNRS (30), INRA (26), and University of Bologna (17).

# **3.5. Keyword analysis**

The analysis of keywords is usually carried out considering author keywords, keyword Plus and title words. These keywords are distributed in several periods (generally 5 year-intervals) to try to reduce annual fluctuations and to ensure a rational period of time (Andreo-Martínez et al., 2020a; Andreo-Martínez et al., 2020b). The distributions of words in different periods can provide information for finding research focus. In addition, this analysis can minimize some of the uncompleted meaning of single words in the title, the small sample size for author keywords, and the indirect relationship between Keywords Plus and the research emphases (Fu and Ho, 2016).

On the other hand, a co-occurrence analysis of all keywords (author keyword and Keyword Plus), using VOSviewer software (van Eck and Waltman, 2013), can also be carried out to find research focus. In this sense, a recent study reported that similar results can be obtained using either the analysis of author keywords, keyword Plus and title words or the co-occurrence analysis of all keywords (Andreo-Martínez et al., 2020b). Therefore, the keyword analysis of the present bibliometric study has been performed using the co-occurrence analysis of all keywords.

Although VOSviewer software conduct an automatic keyword standardization process, some keywords had to be standardized by hand (i.e. honey bee and honeybee by honeybee, neonicotinoids insecticides by neonicotinoids or gas chromatography by gaschromatography). 2151 different keywords were obtained in this analysis. 1470 keywords (68.3 %) appeared one time, followed by 257 (11.9 %) two times and 130  $(6.0 \%)$  three times. 184  $(8.5 \%)$  keywords appeared more than five times, 52  $(2.4 \%)$ more than twenty times, 34 (1.6 %) keywords appeared thirty or more times and only 19 (0.9 %) appeared more than 50 times.

# **Table 6**

Table 6 shows the keyword co-occurrence restricted to a minimum number of 30 together with total link strength and grouped into three different clusters. The total link strength value gives information on the times that a keyword has been linked with others, highlighting the importance of a keyword in the research field. Fig. 7 shows keywords trend over the period analyzed (1948–2019), where the three different clusters can be observed. The highest cluster (red) contains 18 keywords which refer mainly to chromatographic techniques for the determination of pesticide residues in honey or vegetables. The green cluster (13 keywords) include some pesticide residues such as imidacloprid or neonicotinoids in bees. Finally, the blue cluster (3 keywords) refers to the presence of coumaphos pesticide in beewax. Therefore, the main research lines identified in the field are related to the development of chromatographic techniques for

the determination of pesticides such as imidacloprid, neonicotinoids or coumaphos in honey and derivate products such as propolis and wax.

**Fig. 7**

### **3.5.1. Main pesticide residues in honey**

As discussed in the introduction section, up to 173 different pesticides have been identified in beehives, among which neonicotinoids, imidacloprid and coumaphos are the most frequently reported.

Neonicotinoids are a group of insecticides that include nitenpyram, dinotefuran, thiamethoxam, imidacloprid, clothianidin, acetamiprid, and thiacloprid (Xiao et al., 2011). Neonicotinoids act as agonists on insect neural nicotinic acetylcholine-regulated receptors (nAcChR) (Seifert and Stollberg, 2005), causing over-stimulation to nerve cells and resulting in paralysis and death (Wang et al., 2020). This group of pesticides is less toxic to mammals due to the highly selective affinity to nAcChRs of insects over vertebrate (Xiao et al., 2011). In this sense, although imidacloprid (N-(1-((6 chloropyridin-3-yl)methyl)imidazolidin-2-ylidene)nitramide) offers lower toxicity to mammals than to invertebrates (Hashimoto et al., 2020), it can induce oxidative stress on male mice, being hepatotoxic and leading to impairment in the male reproductive system of rats (Khalil et al., 2017). It also may cause oxidative stress and inflammation on the central nervous system and livers in non-target organisms in rats (Xiao et al., 2011).

Neonicotinoids are extensively used as a seed treatment on crops where *A. mellifera* most usually forage, including oilseed rape and ornamental garden plants. They are also applied as foliar sprays on fruits such as pears or apples. Besides, it has been reported that neonicotinoid residues are detected in many water resources, such as surface water and guttation water, and persist in the soil, which can be deemed as an additional source of pesticide exposure for honey bees (Wang et al., 2020). Cyano-substituted neonicotinoid insecticide acetamiprid has low intrinsic toxicity to *A. mellifera*. In contrast, nitro-substituted neonicotinoids such as imidacloprid, thiamethoxam, and clothianidin, is significantly more toxic to honey bees in comparison with the cyanosubstituted neonicotinoids (Wang et al., 2020). In this sense, the use of imidacloprid, clothianidin, thiamethoxam, and fipronil was restricted in 2008 due to the negative effects on honey bee health (Martinello et al., 2020). However, although the potential threat of neonicotinoids for honey bees has been reported, it is still under debate; therefore, more studies are needed to investigate the impacts of neonicotinoids on *A. mellifera* (Buchori et al., 2020).

Coumaphos (O-3-chloro-4-methyl-2-oxo-2H-chromen-7-yl O,O diethylphosphorothioate) is an organothiophosphate pesticide and a stable lipophilic compound. It is active by contact, ingestion, and vapor action, and causes phosphorylation of the acetylcholinesterase enzyme of tissues, allowing the accumulation of acetylcholine at cholinergic neuroeffector junctions (muscarinic effects), and at skeletal muscle myoneural junctions and autonomic ganglia (Del Carlo et al., 2010).

Coumaphos is one of the most commons acaricides used by the beekeepers to combat the parasitic mites *Varroa jacobsoni* and *Ascophera apis* in beehives (Del Carlo et al., 2010; Kochansky et al., 2001). Coumaphos exposure causes negative effects on honey

bees such as impairs olfactory learning and memory, affects locomotion and grooming behavior, reduces trophallaxis, drone sperm viability, and also shows negative effects on queen development, body weight and sperm volume during the rearing and early life of the queen (Chaimanee et al., 2016). Mismanagement of coumaphos in beehives can lead to the appearance of this pesticide in honey with the consequent risk to human health.

Finally, as pesticides often co-exist as mixtures of complex compounds in realistic environments, which may induce extra effects on honey bees and contaminate honey compared with the effect of a single pesticide (Wang et al., 2020), further research is needed in this regard.

# **4. Conclusions**

The present bibliometric review discussed the global trends of pesticide residues in honey research between 1948 and 2019 based on retrieved articles from WoS. The results of the 685 articles analyzed indicated a steady increase in publications, authors, journals, WoS categories, and the number of involved countries and institutions. Specifically, this research field is in the focus of interest nowadays, and linear regression together with Lokta distribution models suggests that a high number of research outputs will be achieved in the near future. Pico, Y and Journal of Chromatography A were the most productive author and journal, respectively. Among the countries, the USA was leading the path, followed by Spain and China, and the most productive institution was ARS (USA), which has a multidisciplinary nature and an important role in the pesticide research field, followed by the University of Valencia (Spain). The co-occurrence analysis of keywords revealed that the chromatographic techniques for the determination of pesticides such as imidacloprid, neonicotinoids or coumaphos in honey and derivate products such as propolis and wax are hot points in this research field.

On the other hand, the possible limitations of the results shown can be related to different aspects such as the database used, the selected Boolean strings, the manual standardization of authors' names, institutions and keywords, the selection of "articles" as the only type of publication to perform the analysis, and the bibliometric parameters used to analyze the publications selected. However, despite these limitations, this bibliometric study provides a global overview of the pesticide residues in honey from to 2019. Finally, the findings in this study might be compared and expanded in the future.

# **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this manuscript.

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#### **Figure captions**

**Fig. 1**. A: the number of publications and articles on pesticide residues in honey research by year from 1948 to 2019. B: the cumulative number of articles published per year from 1948 to 2019.

**Fig. 2.** A) Lotka's Law to calculate the correlation of the number of authors with the articles published; B) Lotka's Law to calculate the correlation of the journals number with the articles published; C) Lotka's Law to calculate the correlation of the institutions number with the articles published.

**Fig. 3.** Author collaboration network for frequencies  $\geq 3$ .

**Fig. 4**. A) Annual publication patter of the six most productive journals; B) Annual publication patter of the five most productive countries.

**Fig. 5**. Top 5 productive Web of Science categories from 1948 to 2019.

**Fig. 6.** A: Countries collaboration networks for frequencies ≥3. B: Institutions collaboration networks for frequencies  $\geq 3$ .

**Fig. 7.** All keywords trend.

### **Table captions**

**Table1.** Characteristics by article year from 1948 to 2019.

**Table 2**. 10 most productive authors ordered by h-index from 1948 to 2019.

**Table 3**. Top 10 productive journals, total number of articles (percentage), h-index, IF

(2018), journal categories, and journal position by categories according to JCR from

to 2019.

**Table 4.** 20 most productive countries from 1948 to 2019.

**Table 5.** 20 most productive institutions from 1948 to 2019.

**Table 6.** Keyword co-occurrence, total link strength and cluster restricted to a minimum

number of 30.

















TP: total publications; 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.



TP: total publications; FP: publication with first author; RP: publication with corresponding author; R: Rank.



TP (%): total publications and percentage of total publications; IF: Impact factor; JCR:

Journal Citation Report.



TP: total publications; SP: single country publication; CP: internationally collaborative publication; FP: publication with first author; RP: publication with corresponding author; R (%): Rank and share in publication; CP%: the percentage of internationally collaborative publications in total publications for each country.



TP: total publications; SP: single country publication; CP: internationally collaborative publication; FP: publication with first author; RP: publication with corresponding author; R (%): Rank and share in publication; CP%: the percentage of internationally collaborative publications in total publications for each institution.

