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REVIEW ARTICLE

Pesticides in honey: bibliographic and bibliometric analysis towards matrix quality for consumption

Pesticidas no mel: análise bibliográfica e bibliométrica com respeito à qualidade da matriz para consumo

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Abstract

Honey is a matrix noted for its wide consumption as a sweetener and its anti-inflammatory, antioxidant, and antimicrobial properties; however, its physicochemical quality can be compromised by the presence of toxicants such as pesticides. This review aims to gather recent information on pesticides in honey from the approach to their detection, understanding, and adverse effects on human health. A bibliographic and bibliometric analysis was carried out in academic databases limited to the last five and thirty years, respectively, comprising the keywords "*honey*", "*pesticides*" and their types of pesticides or the agrochemical compound directly. It was found that there are about 30 pesticides detected in honey, in which organochlorine, organophosphate, and neonicotinoid compounds stood out for their concentrations concerning Maximum Residue Levels (MRL). Their physicochemical alteration was not well explored beyond slight variations in brightness and manganese concentration, and its consumption may have repercussions on human reproductive health. It was also determined that there was limited development on the scientific subject seeing that it is important to explore and investigate more on the issue due to the great impact of honey as a product of high consumption at a global level.

Keywords: Agrochemicals; Bibliometrix; Insecticides; Lindane; Chlorpyrifos; Maximum residue limit; LC-MS/MS; GC-MS/MS.

Resumo

O mel é uma matriz que se destaca pelo seu amplo consumo como edulcorante e por suas propriedades anti-inflamatórias, antioxidantes e antimicrobianas; no entanto, sua qualidade físico-química pode ser comprometida pela presença de elementos tóxicos, como os pesticidas. Esta revisão tem como objetivo coletar informações recentes sobre pesticidas no mel sob a perspectiva de sua detecção, compreensão e efeitos adversos à saúde humana. Foram realizadas as análises bibliográfica e bibliométrica em bases de dados acadêmicas limitadas aos últimos cinco e trinta anos, respectivamente, com as palavras-chave "honey" (mel), "pesticides" (pesticidas) e tipos de pesticidas ou o nome do composto agroquímico diretamente. Foram detectados cerca de 30 pesticidas no mel, dos quais se destacam os compostos organoclorados, organofosforados e neonicotinoides, devido às suas concentrações em relação aos Limites Máximos de Resíduos (LMR). A alteração físico-química do mel é pouco explorada, sendo observadas pequenas variações de brilho e concentração de manganês. Note-se que o consumo de mel com pesticidas pode ter repercussões na saúde reprodutiva humana. Foi também verificado que existe a pesquisa científica sobre o assunto é limitada e que é importante explorar e investigar mais sobre o assunto, devido ao grande impacto que o mel tem como produto de alto consumo em todo o mundo.

Palavras-chave: Agrotóxico; Bibliometrix; Inseticidas; Lindano; Clorpirifos; Limite máximo de resíduos; LC-MS/MS; GC-MS/MS.

Highlights

- Honey contamination is a Food Safety issue of global concern
- Pesticides in honey can alter both bee and human health
- Insecticides are a critical group in the contamination and safety of honey consumption

1 Introduction

Honey is a natural product that has been consumed by humans since ancient times. The composition, aroma, flavor, and color of honey depend on the type of plant where pollination takes place, as well as the geographic regions, climate, and the species of honeybees involved in its production (Oroian et al., 2018). Honey has been used as a food of high nutritional value since it contains essentially carbohydrates, along with minerals, amino acids, vitamins, volatile chemical substances, phenolic acids, flavonoids, and carotenoid-like substances, among others (Kamal et al., 2019). This product has extensive use and consumption not only as a sweetener, but also for its therapeutic properties offered in alternative medicine as an anti-inflammatory, antioxidant, and antimicrobial (Meo et al., 2017). These properties make honey consumption attractive to a society that is becoming interested in a healthier and safer lifestyle, largely owing to the impact of the coronavirus pandemic and this consumption habit is reflected in market studies where it is established that the global size of the honey market for 2021 was valued at around USD 8.58 billion with a projected growth of 5.2% to 2030 (Grand View Research, 2022). Accordingly, the quality of honey plays a fundamental factor in the success of the production and marketing of this product, where it is expected that the components of this type of matrix meet optimal standards of safe consumption, thus ensuring the nutritional contribution and acceptability by the consumer (Ballesteros et al., 2019; Tsagkaris et al., 2021). Within this physicochemical understanding, a critical problem stands out, which are related to those factors that can compromise the quality and safety of the honey, *i.e.,* the pesticides. Pesticides are a group of chemical compounds used to eliminate pests (such as insects, rodents, fungi, or unwanted plants -weeds-) and/or agents that can cause damage to crops (World Health Organisation, 2020). Research on the effects of pesticide use in agriculture where the beekeeping industry is involved has determined that these compounds contribute to the decline of honeybee products and affect crop yields (Fikadu, 2020). Although the number of investigations on the effect of pesticides in the beekeeping industry has increased, the focus on the relationship between pesticides and honey and their effects has not been developed extensively, thus, it is noteworthy to gather recent information on pesticides found in honey. This objective is achieved by updating information about pesticides found in honey, as well as the physicochemical alteration that can be caused by the presence of the pesticide in the matrix, and possible dangers to human consumption, along with the trend evaluation in order to respond to the rising interest in honey.

2 Methodology

The methodology was based on the first stage of bibliographic analysis and the second stage of bibliometric analysis. However, these processes were carried out simultaneously to achieve a complete development of the review.

2.1 Bibliographic analysis

ScienceDirect, PubMed, and Google Scholar databases were consulted for literature on the topic of interest with limitations to the last five years (2017-2022). For the literatures where the time range determined for the update was not met, the articles were gathered up from 2010 to 2016 (as a list of pesticides or studies in Colombia, see results). The main search keywords were "honey" with the Boolean connector "AND" for the focus on pesticides and their types ("pesticides"; "insecticides", "fungicides", "herbicides", "rodenticides", "bactericides", "larvicide"), or the chemical compound belonging to the toxicants to be evaluated with the Boolean connector "OR" ("atrazine", "pirimicarb", "coumaphos", "permethrin", "chlorpyrifos", among others). For the specifications, "honey", the type of pesticide (or as general), or the chemical compound of interest was included together with the concepts of the field to be worked on (i. e.: "bioaccumulation", "detection", "physicochemical parameters", "physicochemical characteristics", "microbiome", among others). Thus, some of the search combinations used were:

- "*honey*" AND "*insecticides*"
- "*honey*" AND ("*chlorpyrifos*" OR "*coumaphos*")
- "*honey*" AND "*chlorpyrifos*" AND "*bioaccumulation*"
- "*honey*" AND ("*fungicides*" OR "*herbicides*") AND "*physicochemical characteristics*"
- "*honey*" AND "*pesticides*" AND ("*detection*" OR "*human health*")

2.2 Bibliometric analysis

A bibliometric analysis was carried out through the bibliographic database Scopus, where articles and reviews from the last 30 years (1991-2001) were selected. The search algorithm was proposed as focus terms "honey" and "pesticides" with some of their respective types (insecticides, fungicides, herbicides, rodenticides, bactericides, and larvicides) using the Boolean connector "AND", and as variants depending on the terms, different names of compounds belonging to the group of pesticides established based on the literature on the presence and contents of pesticides (and toxicants) in honey were proposed using the Boolean connector "OR". The search algorithm used was:

(TITLE-ABS-KEY ("honey") AND TITLE-ABS-KEY (pesticides OR insecticides OR fungicides OR herbicides OR rodenticides OR bactericides OR larvicide) AND TITLE-ABS-KEY (atrazine OR benalaxyl OR pirimicarb OR acetamiprid OR carbendazim OR "carfentrazone-ethyl" OR chlorpyrifos OR chloroanthranilic OR coumaphos OR deltamethrin OR dimethoate OR diphenylamine OR azoxystrobin OR imidacloprid OR malathion OR malaoxon OR methoxyfenozide OR propargite OR pentachlorophenol OR permethrin OR pinostrobin OR tebuthiuron OR "thiophanate-methyl" OR trifloxystrobin OR thiamethoxam OR lindane OR clothianidin OR cypermethrin OR dinotefuran OR quinalphos OR "pirimiphos-methyl" OR diazinon OR phosmet)) AND (LIMIT-TO (PUBYEAR, 2021) OR LIMIT-TO (PUBYEAR, 2020) OR LIMIT-TO (PUBYEAR, 2019) OR LIMIT-TO (PUBYEAR, 2018) OR LIMIT-TO (PUBYEAR, 2017) OR LIMIT-TO (PUBYEAR, 2016) OR LIMIT-TO (PUBYEAR, 2015) OR LIMIT-TO (PUBYEAR, 2014) OR LIMIT-TO (PUBYEAR, 2013) OR LIMIT-TO (PUBYEAR, 2012) OR LIMIT-TO (PUBYEAR, 2011) OR LIMIT-TO (PUBYEAR, 2010) OR LIMIT-TO (PUBYEAR, 2009) OR LIMIT-TO (PUBYEAR, 2008) OR LIMIT-TO (PUBYEAR, 2007) OR LIMIT-TO (PUBYEAR, 2006) OR LIMIT-TO (PUBYEAR, 2005) OR LIMIT-TO (PUBYEAR, 2004) OR LIMIT-TO (PUBYEAR, 2003) OR LIMIT-TO (PUBYEAR, 2002) OR LIMIT-TO (PUBYEAR, 2001) OR LIMIT-TO (PUBYEAR, 2000) OR LIMIT-TO (PUBYEAR, 1999) OR LIMIT-TO (PUBYEAR, 1998) OR LIMIT-TO (PUBYEAR, 1997) OR LIMIT-TO (PUBYEAR, 1995) OR LIMIT-TO (PUBYEAR, 1994) OR LIMIT-TO (PUBYEAR, 1992) OR LIMIT-TO (PUBYEAR, 1991)) AND (LIMIT-TO (DOCTYPE, ar") OR LIMIT-TO (DOCTYPE, "re")).

From the results of the algorithm, data analysis on scientific production was obtained in this same tool according to years, countries, and type of document. Bibliometric analysis was carried out using VOSviewer software (Van Eck & Waltman, 2010), with indicators of data relationship and collaboration (i.e., co-occurrence of keywords and co-authorships between authors and countries), according to methodology proposed by Baena-Pedroza et al. (2021). In addition, time-dependent clustering analyses of these selected co-occurrences were obtained. The minimum number of occurrences of a keyword was selected to be 41 (total number of key terms used in the search algorithm), where out of 5738 words 740 meet the threshold and 110 keywords were obtained for filtering according to the desired approach.

Using the same search algorithm extracted from Scopus, Bibliometrix package within R environment was used to analyze and map the bibliographic data with the version 1.442 of RStudio in Windows 10. Bibliometrix's functions was used to analyzing dataset of articles including annual scientific production and three fields plot. In addition, some functions as conceptual, intellectual, and social structure were analyzed.

3 Results

The results are focused on a compilation of recent information on the topic of interest, but as can be seen below, there will be a development of articles with dates before the time range used due to the limited research development on pesticides in this matrix.

3.1 Pesticides found in honey

The direct interaction of bees and nectar of pollinated plants is the source of the arrival of pesticides to honey since many of them are found in crops that receive agrochemical treatment with these compounds. Although the bee is the agent directly affected, many of these pesticides through their transformation or their residues can accumulate in honey and other bee products, as shown in Figure 1 (Belsky & Joshi, 2020; Vargas-Valero et al., 2020). Pesticides found in honey are considered persistent organic pollutants (POPs), which can persist in the environment through bioaccumulation in different matrices and biomagnifies in ecosystems, and have properties of chronic toxicity, one of their main compounds being lindane (Wang et al., 2019). Some of these POPs are classificated as "commonly used pesticides (CUP)", which are more water-soluble, less persistent, and less bioaccumulative than other POPs, including organophosphates such as chlorpyrifos (Adeyinka et al., 2022). However, their massive use and extensive amounts represent a critical hazard for those organisms that interact with the matrices containing the product, due to their carcinogenic, neurotoxic, adverse growth, endocrine disruption, or respiratory effects (Degrendele et al., 2022; Villalba et al., 2020).

Figure 1. Honeybee exposure to pesticides and arrival of agrochemical residues in honey. Honeybees mostly perform pollination from plants linked to agrochemical processes for pest control. The pesticide is introduced into the animal's metabolism by dermal contact and/or ingestion of the plant products. Within all the metabolic processes of the bee residues of pesticides (direct or biotransformed) to bee products such as honey, there is a concern about the components of this matrix. Modified from Crenna et al. (2020).

The identification of these pesticides in honey can be an indirect indicator of Good Agricultural Practices (GAPs) in terms of agrochemical management of crops in which honeybees take place, because it is of great importance to understand the type of pesticide to approach in the best way strategies to improve the quality of life for organisms and abiotic factors related to these practices, beyond the physicochemical knowledge of this product (Kumar et al., 2018). Another important consideration about honey as an indicator of practices related to the application of pesticides is that the study of pesticides in honeybees directly cannot be fruitful due to the rapid metabolic transformations that these products can have within them and the selectivity and representativeness of the colony at the time of proposing the sampling of the experiment. So, the analysis of their products could be useful for the confirmation or complementation of information on the analysis of pesticides (Murcia-Morales et al., 2022).

Extracting and quantifying the concentration of these chemical compounds in the different matrix in traces has been very important. Extraction methods such as solid-phase extraction (SPE) or dispersive extraction (QuEChERS) are used for some pesticides that can be quantified between 0.005 and 0.01 $ng \cdot g^{-1}$ (Mejías & Garrido, 2022) (Table 1). Pesticides detection in honey (and another matrix) can be done using spectrometric and chromatographic analysis, such as liquid chromatography-tandem mass spectrometry (LC-MS/MS) and gas chromatography-mass spectrometry (GC-MS) (Almutairi et al., 2021; Mejías & Garrido, 2022; Tette et al., 2016) (Table 1). Here it is highlighted that the detection of pesticides, due to its growing interest, is a field that is proposing new techniques and new analytical tools for faster and more sensitive detection of pesticides (Fauzi et al., 2021).

Table 1. List of common pesticides detected in bee honey. MRL: maximum residual limit; QuEChERS: quick, easy, cheap, effective, rugged, and safe method; SPE: solid-phase extraction; SLE: liquid-phase extraction; GC-MS: gas chromatography coupled to mass spectrometry; LC-MS/MS: liquid chromatography coupled to tandem mass spectrometry; TSD: specific thermionic detector (nitrogen and phosphorus detector); DAD: diode array detector.

Type of pesticide	Main group	Mechanism of action	Chemical product	Method of analysis and extraction (Mejías & Garrido, 2022)	MLR ($mg\ kg^{-1}$) (European Comission, 2022c)	References
Insecticides	Organophosphates	Acetylcholinesterase inhibition	Chlorpyrifos	QuEChERS and GC–MS	0.01	Adeyinka et al., 2022; Carral, 2016; Vargas-Valero et al., 2020
			Coumaphos	SPE and LC-MS/MS	0.1	Adeyinka et al., 2022; Vargas-Valero et al., 2020
			Dimethoate	SPE and LC-MS/MS	0.01	Adeyinka et al., 2022; Carral, 2016; Vargas-Valero et al., 2020
			Malathion	QuEChERS and GC–MS	0.05	Adeyinka et al., 2022; Carral, 2016; Vargas-Valero et al., 2020
			Malaoxon	SLE and GC-TSD*	0.01**	Adeyinka et al., 2022; European Comission, 2022a; Matus et al., 2010; Vargas-Valero et al., 2020
			Phosmet	SPE and LC-MS/MS	0.05	Adeyinka et al., 2022; Carral, 2016; Sanchez-Bayo & Goka, 2016
			Diazinon	QuEChERS and GC–MS	0.01	Adeyinka et al., 2022; Carral, 2016; Sanchez-Bayo & Goka, 2016

Type of pesticide	Main group	Mechanism of action	Chemical product	Method of analysis and extraction (Mejías & Garrido, 2022)	MLR (mg kg^{-1}) (European Comission, 2022c)	References
			Pirimiphos-methyl	QuEChERS and GC–MS	0.05	Adeyinka et al., 2022; Sanchez-Bayo & Goka, 2016
			Quinalphos	QuEChERS and GC–MS	0.05	Adeyinka et al., 2022; Sanchez-Bayo & Goka, 2016
	Carbamates	Acetylcholinesterase inhibition	Pirimicarb	QuEChERS and GC–MS	0.05	Choi et al., 2020; Carral, 2016; Silberman & Taylor, 2022
	Pyrethroids	Alteration of the function of voltage-dependent sodium channels	Fluvalinate	QuEChERS and GC–MS	0.05	Chrustek et al., 2018; Carral, 2016; Erban et al., 2019
			Permethrine	QuEChERS and GC–MS	0.01**	Carral, 2016; European Comission, 2022a; Vargas-Valero et al., 2020
			Deltamethrin	QuEChERS and GC–MS	0.05	Chrustek et al., 2018; Carral, 2016; Vargas-Valero et al., 2020
	Organochlorines	Alteration of the function of voltage-dependent sodium channels and γ-aminobutyric acid (GABA) neurotransmission	Pentachlorophenol	SPE and LC-MS/MS*	0.01**	Jayaraj et al., 2016; Kraševec et al., 2021; Vargas-Valero et al., 2020
			Lindane	QuEChERS and GC–MS	0.01	Kraševec et al., 2021; Sanchez-Bayo & Goka, 2016
	Organosulfurates	Inhibition of mitochondrial ATP synthase activity, inhibiting oxidative phosphorylation	Propargite	QuEChERS and GC–MS	0.05	Carral, 2016; Lewis et al., 2016; Vargas-Valero et al., 2020
	Anthranilic diamides	Alteration of the internal calcium reservoirs causes exhaustion and therefore muscular paralysis of the insect	Chlorantraniliprole	SPE and LC-MS/MS	0.05	Carral, 2016; Lewis et al., 2016; Vargas-Valero et al., 2020
	Neonicotinoids	Inhibition of the active site of nicotinic acetylcholine receptors (nAChR)	Acetamiprid	SPE and LC-MS/MS	0.05	Carral, 2016; Ihara & Matsuda, 2018; Vargas-Valero et al., 2020
			Imidacloprid	SPE and LC-MS/MS	0.05	Carral, 2016; Ihara & Matsuda, 2018; Vargas-Valero et al., 2020
			Dinotefuran	QuEChERS and LC-DAD*	0.01**	Carral, 2016; European Comission, 2022a; Ihara & Matsuda,

Type of pesticide	Main group	Mechanism of action	Chemical product	Method of analysis and extraction (Mejías & Garrido, 2022)	MLR (mg kg^{-1}) (European Comission, 2022c)	References
						2018; Rabea et al., 2018; Sanchez-Bayo & Goka, 2016
			Thiamethoxam	SPE and LC-MS/MS	0.05	Carral, 2016; Ihara & Matsuda, 2018; Sanchez-Bayo & Goka, 2016
			Clothianidin	SPE and LC-MS/MS	0.05	Carral, 2016; Ihara & Matsuda, 2018; Sanchez-Bayo & Goka, 2016
			Thiacloprid	SPE and LC-MS/MS	0.2	Carral, 2016; Ihara & Matsuda, 2018; Renaud et al., 2018
	Sulfoximines	Competitive inhibition of nicotinic acetylcholine receptors (nAChR)	Sulfoxaflor	QuEChERS and GC–MS*	0.05	Capela et al., 2022; Carral, 2016; Kim et al., 2017; Lewis et al., 2016
	Hydrazines	Antagonist/agonist action on the E20 hormone, essential in the molting and metamorphosis process of some insects	Methoxyfenozide	QuEChERS and GC–MS	0.05	Carral, 2016; Lewis et al., 2016; Vargas-Valero et al., 2020
Fungicides	Phenylamides	Disruption of fungal nucleic acid synthesis through inhibition of RNA polymerase I	Benalaxyl	SPE and LC-MS/MS	0.05	Choi et al., 2020; Carral, 2016; Lewis et al., 2016
	Benzimidazoles and carbamates	Inhibition of mitosis and cell division by affecting beta tubulin	Carbendazim	SPE and LC-MS/MS	1	Carrasco et al., 2017; Lewis et al., 2016; Vargas-Valero et al., 2020
			Tiophanate-methyl	SPE and LC-MS/MS	1	Carrasco et al., 2017; Lewis et al., 2016; Vargas-Valero et al., 2020
	Strobilurins	Inhibition of electron transfer in photosynthesis	Fluoxastrobin	SLE and LC-DAD*	0.05	Carral, 2016; Feng et al., 2020; Sun et al., 2017; Vargas-Valero et al., 2020
			Pyraclostrobin	QuEChERS and LC-MS/MS*	0.05	Carral, 2016; Feng et al., 2020; Lin et al., 2022; Vargas-Valero et al., 2020
			Trifloxystrobin	QuEChERS and LC-MS/MS*	0.05	Carral, 2016; Feng et al., 2020; Luo et al., 2020; Vargas-Valero et al., 2020
Herbicides	Triazines and derivatives (triazolinones)	Inhibition of electron transfer in photosynthesis	Atrazine	QuEChERS and GC–MS	0.05	Choi et al., 2020; Carral, 2016; Grasselli et al., 2018

Type of pesticide	Main group	Mechanism of action	Chemical product	Method of analysis and extraction (Mejías & Garrido, 2022)	MLR (mg kg^{-1}) (European Comission, 2022c)	References
		Cell membrane disruptor by blocking chlorophyll biosynthesis and inhibiting the enzyme polyphenol oxidase (PPO)	Carfentrazone-ethyl	QuEChERS and GC–MS	0.05	Carral, 2016; Lewis et al., 2016; Vargas-Valero et al., 2020
	Thidiazole-ureas	Inhibition of photosynthesis with alteration of photosystem II (PSII)	Tebuthiuron	LC-MS/MS*	0.01**	European Comission, 2022a; Lewis et al., 2016; Thomas et al., 2020; Vargas-Valero et al., 2020

*Citation not corresponding to central citation of extraction and analysis methods, see reference column of the pesticide in question. **The MRL of the pesticide is not specifically defined, so by general default its value is 0.01 mg kg^{-1} (European Comission, 2022a).

The detection and quantification of pesticides in honey became a critical activity as it is found in important and considerable quantities, which can even exceed the maximum residue limits (MRL) of pesticides, defined as the highest possible level of tolerance to traces of pesticides in products for human consumption or others when a correct application of Good Agricultural Practices, established by regulatory agents such as the European Union, is carried out (European Comission, 2022b; Food and Agriculture Organization of the United Nations, 2020). Hence, Table 1 summarize some of the pesticides commonly detected in honey, together with their mechanism of action, method of extraction and analysis, and MRL.

In Table 1, it can be observed that among the pesticides indicated in this table, data was not shown about bactericides, rodenticides, and larvicides, since the majority of studies on the subject are focused on the effects on the honeybee as a key insect of the Colony Collapse Problem (CCP) and as the fungus as one of the main phytopathogenic agents in crops (Cullen et al., 2019; Douglas et al., 2020). In some studies, on the detection and quantification of pesticides in raw honey, residues of pesticides such as imidacloprid, deltamethrin, and (beta)cypermethrin are described above the MRL, and for commercialized honey, chlorpyrifos, imidacloprid, and malathion can be found in concentrations higher than those indicated by the MRL (Valdovinos-Flores et al., 2017; Xiao et al., 2022). At the same time, it is common to detect a large amount of these pesticides below the MRLs (Darko et al., 2017). In a more particular case, one of the few studies conducted on this issue in Colombia in 2014 showed that only 5% of the analyzed kinds of honey exceeded the MRL according to the pesticide analyzed (the critical ones being lindane and other organochlorines, and chlorpyrifos) (López et al., 2014). The fact that they continue to be detected even below the limits continues to be a factor of concern due to the bioaccumulation capacity of many commercially used pesticides, where they are chronically exposed to concentrations that usually result in lethal and sublethal effects in humans and other organisms, with a lack of knowledge of these effects due to the lack of delimitation of the time ranges for this exposure (Gupta & Gupta, 2020). As can be seen in Table 1, there are pesticides with extremely low tolerance, so their understanding of bioaccumulation and adverse effects on humans through the consumption of this type of product should be evaluated promptly. One reason why the concentrations of pesticides found in honey are relatively low, and therefore not much investigated and explored, is because to other honeybee products (such as beeswax) these concentrations in the matrix of interest are much lower, possibly also linked to the different synthesis of these two products by this animal (Calatayud-Vernich et al., 2018; Leska et al., 2021).

3.2 Physical and chemical alteration of honey and risk to human consumption

Several studies had analyzed changes in the physicochemical parameters of honey with the presence of pesticides.

Main conclusions are related to the fact that there is no physicochemical alteration of the matrix and the risk associated with the consumption of honey exposed to the mentioned pesticides is negligible (Mulugeta, 2017). However, it is possible to find, although infrequently, honey samples that exceed the MRLs; as in the case of Kędzierska-Matysek et al. (2022), where 0.9% of analyzed honey samples exceed the MLRs in terms of their concentration of amitraz, glyphosate, acetamiprid, thiacloprid, azoxystrobin, among others. For more information on the agrochemicals found in honey and their MRLs, please refer to Table 1. In addition, it has been described that those honey with toxicant contamination (where pesticides are also evaluated) may present differences reflected in higher hidroximethylfurfural (HMF) contents or color changes; and the presence of neonicotinoids has been related to low manganese concentrations and the brightness of the analyzed honey samples (Scripcă & Amariei, 2021).

The lack of information about the possible physicochemical and microbiological alterations of this matrix may be due to the primordial need to directly detect pesticides in honey, due to the risks already associated with them, leaving in the background the research on how honey is compromised in its nutritional contribution and its innocuousness in the possible transformations that the pesticide may undergo until it reaches the moment of consumption by humans. Similarly, the literature already explains how honey can be a vehicle for intoxication with pesticides at the time of consumption and finds a close relationship with a negative impact on human reproductive health. In general, honey is conceived as a product that, even if the pesticide is present in its composition, its risk index is not considerable, and it is improbable to observe the development of these adverse effects through its consumption (Naggar et al., 2017). According to El-Nahhal (2020), there is also evidence of the effects of this pesticide-contaminated honey which may contain a significant content of a wide variety of agrochemicals in very broad range of concentrations. About 92 types of pesticides from different international sources record, in average daily adult honey consumption rates, chlorpyrifos and lindane at values of 14.83 and 22.42 ng g^{-1}, respectively, while acetamiprid and imidacloprid are set at 0.17 ng g^{-1}, respectively (according to hazard index discussed by the author); these exposure data are linked to the possibility to have a decreased semen quality (in organophosphate pesticide type) and a high risk of infertility in females (organochlorine type). It is also reported that the consumption of honey containing lindane (along with other types of pesticides) may be linked to miscarriages, reduced implantation processes, shortening of the menstrual cycle, and even prostate cancer (Mukiibi et al., 2021). Despite the impact it can have, a study on the simulation of these adverse effects in humans found that the only pesticide with carcinogenic properties, with compounds such as diazinon and permethrin being evaluated, is lindane (Mahdavi et al., 2022).

3.3 Bibliometric analysis

The bibliometric analyses in terms of scientific production establish that there begins to be a significant production of the topic around 2005 onwards, with the exponential growth of research since 2011 approximately (Figure 2A); the topics has evolved since techniques of quantitation, the recognition of the main pesticides that affect bee health and honey production, up to the inclusion of this topic with a food safety approach, as seen in Figure 2B. The plots obtained in Figure 2C are surely explained by the honeybee population crisis that occurred between 2006 and 2007 in the United States of America (USA), and with significant losses in the numbers of this same population around 2013 (United States Environmental Protection Agency, 2022); accordingly, it is the country with the highest number of research and scientific work on the subject, followed by China and other countries with a fairly low number of publications concerning the former. The analysis regarding keywords is also related to the persistent issue of research focused on to the bee health, leaving to a secondary degree of importance the understanding of exposure, residues, among others of the products of the bee concerning human consumption. It is important to remark the cooperation and the efforts seen between the international community (like Italy and the USA) in this subject directed to comprehend not only the adverse effects in the bee but also in honey and its possible repercussions in human health due the pesticides. In the case of Latin America (except Brazil), there is no production of more than 25 publications for each of the countries evaluated, and there are four documents produced in Colombia. A relevant aspect of this analysis is that of all the documents available in the database used, only 2.5% correspond to the type of review, so it makes sense to explore and work on the line of reviews on this critical issue not only globally, but also in Colombia.

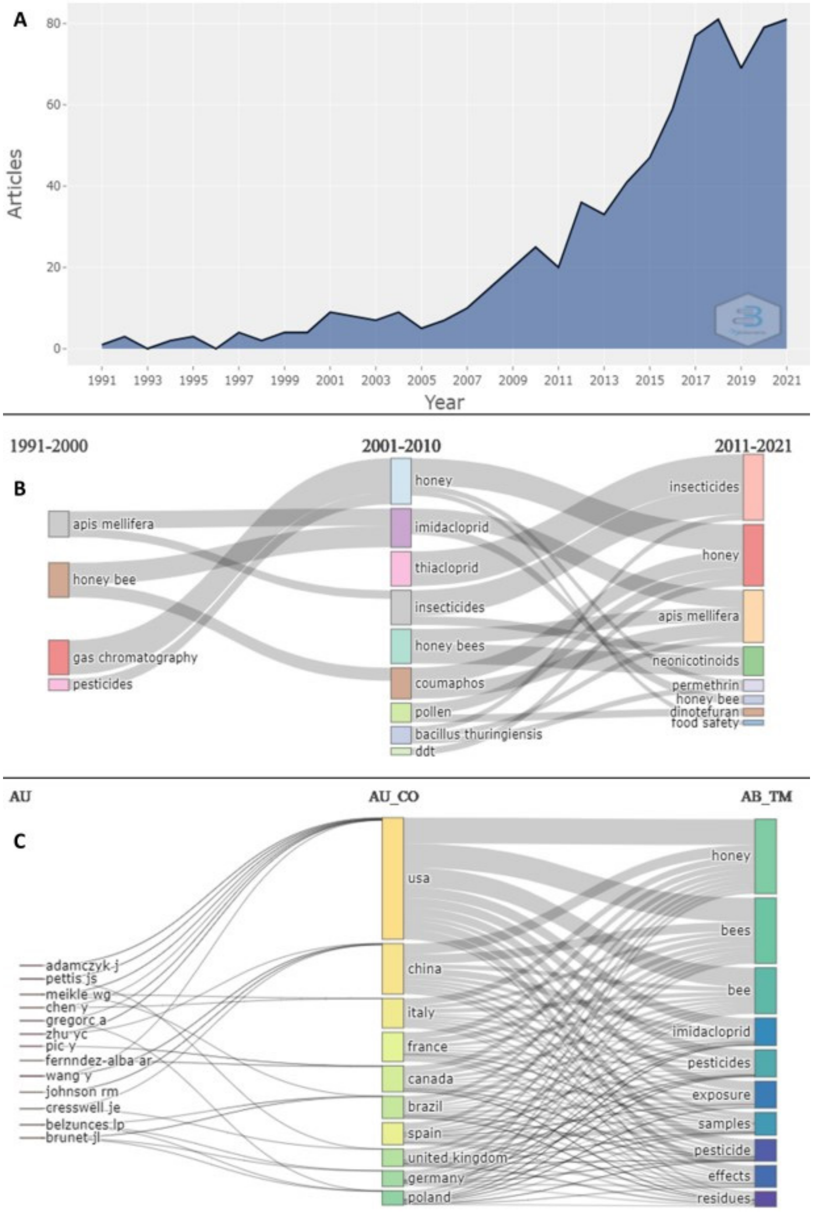

Figure 2. Analysis of honey and pesticide publications as a function of the production of scientific material. A) annual scientific production for 30 years; B) thematic evolution based on keywords author (cutting points years 2000 and 2010); and C) three fields plot (left: main authors; middle: countries and, right: keywords). Plots realized in Bibliometrix for R.

For keywords network analysis obtained through co-occurrences, the formation of the network on 25 items grouped in two clusters was evidenced. The orange cluster (left section) finds the characteristics of its grouping by the association of research on pesticide honey (where acaricides and fungicides are included) with the analysis techniques (liquid and gas chromatography, and tandem mass spectrometry), where within the chemical compounds grouped in this section (pyrethroids -fluvalinate-, and organophosphates -chlorpyrifos, coumaphos, dimethoate-) share the particularity that they are insecticides whose mechanism of action is through the inhibition of the enzyme acetylcholinesterase or one of the types of its receptors (nAChRs), generating adverse effects on the nervous system of certain organisms studied (Rey-Henao et al., 2020). The yellow cluster (right section) refers more directly to the association of the chemical compounds (and their chemical families) about their classification as insecticides; for neonicotinoids used to be grouped as a type of nitro-guanines (Navarro, 2016). Within this type of insecticide are acetamiprid, thiacloprid, imidacloprid, clothianidin, and thiamethoxam (pesticides included in the co-occurrence network). In turn, many of the pesticides commonly used in agriculture related to beekeeping and its products are usually imidazoles, thiazoles, and guanidines, whose relationship characteristic is that they are nitro compounds (Figure 3A). In the timeline it is observed that the studies of honey of those compounds that were associated by occurrence and the analysis techniques have been worked from 2013 until approximately 2016; but that the understanding of pesticides with their structures and chemical families is more recent (Figure 3B). This distribution of the timeline is closely related to the anthropological and scientific understanding of the research of toxicants as a direct risk of consumption to humans to understand these same toxicants as a risk in environmental actors, which in this case would be the processes of toxicity caused by insecticides beyond humans also in honeybees (Kirchhelle, 2018).

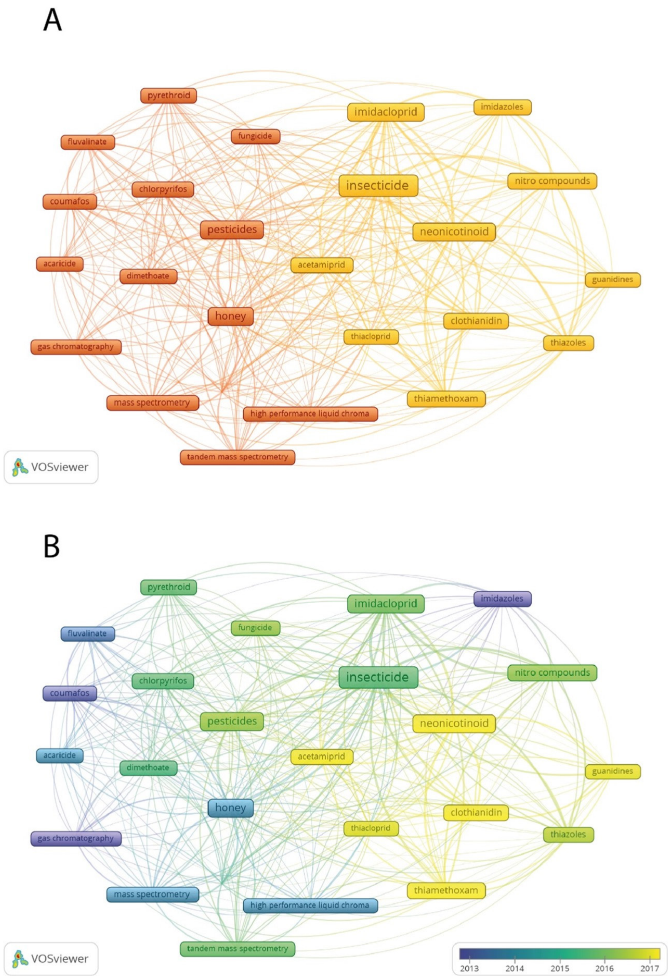

Figure 3. Co-occurrence network plots (by keywords) on honey and pesticides (types and chemical compounds). A) clustering by co-occurrence and B) clustering by timeline. Plots realized in VosViewer (10).

4 Conclusion

The compilation of information regarding scientific research on pesticides as toxicants in honey is very limited by the focus of study in this field, the main actor is the honeybee. Although it is possible to determine that honey may contain a great variety of pesticides (where insecticides, fungicides, and herbicides stand out), the literature does not delve into the physicochemical alteration in terms of minerals and brightness of honey, nor into the toxic impact that this matrix may have beyond the human reproductive system, some even stating that it is improbable that pathologies develop as a result of this product; however, this type of statement may be considered imprudent given the little development of research on these toxicants in honey. On the other hand, organochlorine, organophosphate, and neonicotinoid insecticides have been identified as the main actors in this problem, where the registration of persistent organic pollutants (POPs i.e., lindane) with levels higher than the Maximum Residue Levels (MRL) in some kinds of honey studied and commonly used pesticides (CUPs) such as chlorpyrifos (organophosphate) and imidacloprid (neonicotinoid), along with another important number of pesticides detected below the MRL (with low tolerance levels) that may represent future health problems due to their bioaccumulation properties and others. An invitation is extended to continue exploring this problem with a special focus on its physicochemical properties and its bioaccumulation effects in humans, due to its importance as a basic product of consumption both nationally and internationally, and above all to strengthen its research in Colombia.

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