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ENVIRONMENTAL CONCERNS REGARDING THE OCCURRENCE OF NEONICOTINOID INSECTICIDES IN BERRY FRUITS

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Abstract

Goji berry (Lycium barbarum) and wolfberry (Lycium chinense) became popular nowadays due to their supposed health benefits. Conventionally grown goji berries are often treated extensively with pesticides that led to high levels of pesticide residues in the imported products in Europe and in the USA. Among the insecticides, neonicotinoid type active ingredients have frequently been found in wolfberry products of Chinese origin, which occasionally exceeded the corresponding official maximum residue limit (MRL) values. In addition to imidacloprid and thiamethoxam, much older and obsolete active ingredients have also been detected, e.g. carbofuran. The negative effects of neonicotinoids on pollinators, particularly bees, led to the ban of 3 neonicotinoids (clothianidin, imidacloprid and thiamethoxam), whereas acetamiprid is still in use in Europe. Along with the strict EU regulations, legal violations occurred related to pesticide residues in the imported goji berry products, which led to an increase in the level of official control on imports (2018/941) in the EU. Paradoxically, the corresponding MRL (EC 396/2005) e.g. for acetamiprid has been increased.

Key words: *neonicotinoid insecticides, ban, goji berry, wolfberry.*

INTRODUCTION

Nowadays goji berry (*Lycium barbarum*) or wolfberry (*Lycium chinense*) became popular in developed countries due to their supposed health benefits. The fruit of these two closely related species (Figure 1) have been long used in the cuisine of various nations, and also as an ingredient in traditional medicine in Asia. It is cultivated mostly in China, but also in other East Asian countries (Japan, South Korea), and plantations have been established worldwide in the last decades.

Figure 1. The fruits of two closely related species, goji berry (*Lycium barbarum*) and wolfberry (*Lycium chinense*)

PESTICIDE RESIDUES FOUND IN GOJI BERRY OR WOLFBERRY

Conventionally grown goji berries are often treated extensively with pesticides (insecticides and fungicides) that led to the high levels of pesticide residues determined in the imported products in the United States and in Europe. From the beginning of this century, high levels of insecticides including fenvalerate, cypermethrin and acetamiprid, and fungicides including triadimenol and isoprothiolane have been detected by the US Food and Drug Administration in some imported wolfberries and wolfberry products of Chinese origin.
The European Food Safety Authority (EFSA) report on pesticide residues in food found in 2018 in the EU (EFSA, 2020) specified exceedance rates of the maximal residue limit (MRL) and quantification rates for processed food products (excluding baby foods); quantified residue levels below the MRL were found in nearly half (46.9%) of the goji berries, and residue levels were above the MRL in a quarter of the samples analyzed. Non-

compliance rate for goji berries from China were 13% in 2018. A similar EFSA report for 2019 (EFSA, 2021a) found 45.8% quantification rate for goji products, and MRL exceedance rate was 4.2%. Processed goji berry products containing multiple quantified residues were detected in 55% of samples up to 5 ingredients in 2018 (EFSA, 2020), but an individual goji berry sample originated from China contained residues of as many as 28 different pesticides, which was likely due to mixing of different batches. Some non-approved EU substances (e.g. nicotine, anthraquinone, carbofuran) have also been found in goji berries originated from third countries. In the subsequent year (EFSA, 2021a), half of the analysed goji berry samples were without quantified residues, whereas six of the 48 samples contained one residue and multiple residues were detected in 18 samples, among them five were found to be contaminated with more than five residues. According to the provisions of Regulation (EC) No 669/2009 on import controls, in 2018, certain food products from China (tea, goji berries, broccoli), were subject to an increased level of official controls for certain pesticides at the point of entry into the EU territory.

A survey conducted in the Czech Republic with imported goji berry, reported multiple residues in all samples, and in some cases a total of over 20 different pesticide active ingredients were found (Czech Public Health Institute, 2017). These results are in accordance with those found in Germany, where only the two organic products and two of the 24 conventionally grown goji berry samples met the legal requirements (Hacker et al., 2010). Among the insecticides, neonicotinoid type ingredients (mainly imidacloprid, thiamethoxam and acetamiprid) were frequently found in some wolfberries and wolfberry products of Chinese origin, which occasionally exceeded the corresponding MRL. Particularly high detection frequencies were reported in Germany, where contamination rates for acetamiprid and imidacloprid were found 92% and 65% of the samples, respectively (Hacker et al., 2010). Similar results were obtained in the Czech Republic, as mentioned already, where according to the report of a public health institute, acetamiprid and imidacloprid were found in detectable amounts in all goji berry samples analysed (Czech Public Health Institute, 2017). In addition to imidacloprid and thiamethoxam, several old and outdated insecticide active ingredients (e.g., carbofuran, myclobutanil, carbendazim) were also detected, that have long been banned in the European Union (EU) (Hacker et al., 2010).

A national summary report on pesticide residues measured in Czech Republic in 2017 (EFSA, 2019a) also confirms multiple residues in goji berry samples (21 and 24 compounds). Detections notified in the EU Rapid Alert System for Food and Feed (RASFF) were related to propargite and carbofuran in goji from China, and also 2,4-D and acetamiprid were found at lower levels.

In Germany, only 6 out of 26 samples studied met the legal provisions in force set for pesticide residues (Hacker et al., 2010). Legal violations were mainly related to acetamiprid residues as its level exceeded the MRL in 77% of the conventionally grown goji berry samples, but MRL violations were also found for chlorothalonil and propargite. In addition, 11 pesticide residues per sample were detected in an average. In total, 38 different pesticides were found, and pollution rates for the most often found 17 active ingredients ranged from 23% to 92%. There is no point in using such a large number of pesticides, thus it indicates that farmers are not sufficiently advised by experts on the use of plant protection products (PPPs), or the cultivation areas are extremely polluted by pesticides.

High detection frequencies together with the high number of pesticide residues detected per sample have led to increase the levels of official control on imports of certain feed and food of non-animal origin in the EU, including goji berry as well (EU 2018/941). In contrast, however, the corresponding MRL (EC 396/2005) e.g., for acetamiprid changed from the original 0.01 mg/kg value to 0.1 mg/kg. According to EU regulation (Commission Regulation, 2018), the same MRLs apply to goji berry or wolfberry products as to tomato. After modification of the existing MRLs for acetamiprid in various crops (EFSA Journal, 2021), the corresponding MRL determined for tomato is currently 0.5 mg/kg. The same value was in force for imidacloprid as well, but now the MRLs for tomatoes has been lowered to 0.3 mg/kg (Commission Regulation, 2021)

Organochlorine pesticides also appear to still being used in certain regions of the world in commercial wolfberry cultivation to mitigate infestation by insects. As by China's Green Food Standard, some amount of pesticide and herbicide use is permitted, the organic claims for berries might differ from the European standard. A recently published study (Zhang et al., 2022) on the levels of major hazardous pesticides and metals in *L. barbarum* from China concluded that metal exposure is more hazardous than pesticide exposure. Only dichlorvos was detected at low levels from the six pesticides (dichlorvos, omethoate, cypermethrin, fenvalerate, malathion, and deltamethrin, nonetheless dichlorvos being the most hazardous of them) measured in supermarket samples. Deltamethrin was not detected in any sample, whereas the levels of other active ingredients were considerably below their MRLs in the plantation samples. Health risk assessment found low values, however removal rates of pesticides by washing and drying as well as consumption modes will substantially affect the results that should be further investigated.

More pesticide active ingredients were monitored in a study (Xing et al., 2022), where 200 wolfberry samples were analysed, and 23 pesticides were detected in 83.5% of the samples. Acetamiprid, carbendazim and imidacloprid were detected in 79%, 50% and 43% of the samples, and pyridaben, acetamiprid and difenoconazole exceeded the MRL the most frequently, in 19.5%, 12% and 11% of the wolfberry samples, respectively. Acetamiprid, imidacloprid carbendazim, pyridaben, propargite and thiamethoxam were detected in samples from all regions. Thus, neonicotinoids (imidacloprid, acetamiprid and thiamethoxam) were major components in all three categories (most frequent, high MRL exceedance rate, wide-spread occurrence). Nonetheless, the accessed risk posed by chronic and acute dietary intake of pesticide residues in wolfberry were low.

Another recently published study (Kim et al., 2021) on pesticide residue monitoring also confirmed that there is no significant health risk for consumers via consumption of *Lycium chinense*. Pesticides were detected in 14 of 15 total samples, and the number in each sample ranged from 2 to 21 different pesticide active ingredients with an average of 10.3 kinds per sample. In total, 42 different pesticide active ingredients were detected, including 27 insecticides and 15 fungicides. The most commonly detected pesticides were pyraclostrobin and tebuconazole (64.3% of 14 samples), but other ingredients (e.g. dinotefuran, imidacloprid) were also frequently found. The highest concentration was measured for boscalid (3.14 mg/kg), and the maximum residue concentration of insecticides ranged between 0.009 and 0.369 mg/kg. Regarding the mode of action, among the detected insecticides, neonicotinoids were the most predominant type, followed by pyrethroids.

NEONICOTINOIDS

Due to the high amounts and wide-scale use of the earlier applied neonicotinoids, they became ubiquitous contaminants in surface water and in the soil (Morrissey et al., 2015; Mörtl et al., 2020; Goulson, 2013). As these are systemic pesticide active ingredients, they readily translocate from the soil into the xylem fluid, spread throughout the entire plant, and their residues appear in the fruits, flowers, the nectar, the pollen, and the guttation fluid as well. Guttation is the exudation of droplets of xylem sap that appears on the tips or edges of the leaves. Berries (e.g., strawberry), but also tomato, corn and some grasses or weeds (e.g., red poppy) often and intensively guttate (Figure 2). Neonicotionoids and other systemic pesticides, which were uptaken by the roots from the surrounding polluted soil or irrigation water (Mörtl et al., 2016, Mörtl et al., 2020), can be detected in the guttation droplets. Analysis of the guttation liquid indicate the presence of pesticides residues; however, the exact concentrations depend on numerous factors including species, pollution level of the environmental area, soil properties and meteorological conditions as well.

The presence of chlothianidin was proven in the guttation drops of corn plants up to two months upon exposure, but thiamethoxam could also be easily detected over a month (Mörtl, 2020). In contrast, according to a study

(Li et al., 2019a) conducted with the commercial formulation of dinotefuran and thiamethoxam and goji berries in northwest China, the dissipation of neonicotinoids in goji seems to be fast. Thiamethoxam dissipated in the goji plant ecosystem with half-lives about one day in goji berry, whereas DT_{50} values ranged between 2.04 and 4.25 days in soil. The final residues of thiamethoxam were <0.005-0.382 and <0.005-1.120 mg/kg in goji berry and soil, respectively.

Figure 2. Guttation drops on leave edges of strawberry, tomato and corn plants

REGULATION OF NEONICOTINOIDS

Negative effects of neonicotinoids reported on non-target insect species, particularly on bees, led to the restriction of the use of imidacloprid, thiamethoxam and clothianidin in the EU (The European Commission, 2013), and then all their applications were banned except their use in closed greenhouses (The European Commission, 2018a; 2018b; 2018c). Acetamiprid is still in use in Europe as the EU's food safety authority (EFSA) established low risk to bees, but it was also banned together with thiacloprid from 2018 for example in France (France government, 2018). Based on a conclusion by the EFSA (EFSA, 2019b), the European Commission has not renewed the authorisation of thiacloprid in 2020 due to environmental concerns related to the use of this pesticide, particularly its impact on groundwater, but also related to human health, in reproductive toxicity. Currently, only acetamiprid is approved as a neonicotinoid active substance in the EU for the use in PPPs from the five neonicotinoid insecticides that were most often used. The agricultural sector that has been hit most hard by the ban of neonicotinoids is probably sugar beet cultivation. That is why Ministerial Order provisionally authorised the use of sugar beet seeds treated with neonicotinoid PPPs in 2021 in France. Defra in UK approved an emergency authorisation to use neonicotinoid-treated seed due to the risk posed by virus yellows for the 2022 sugar beet planting season, growers will be allowed to use seed treated by a PPP containing thiamethoxam. Emergency authorizations of the use of neonicotinoids on sugar beet have been issued by EU Member States e.g., Belgium, Croatia, Denmark, Finland, France, Germany, Lithuania, Poland, Romania, Slovakia, and Spain in several cases, and have been evaluated by EFSA (EFSA 2021c). This has triggered a debate on the definition and stricter requirements for the justification of any emergency authorization in the EU (Epstein et al., 2022).

In 2021, the EU Agriculture and Fisheries Council (AGRIFISH) established a specific protection goal of 10% as the maximum permitted level of colony size reduction resulting from exposure of honeybees to pesticides (EC, 2021). This has been communicated as a step towards the protection of bees, but in reality, it is a drastic reduction of the rigor in pollinator protection and appear to contradict ongoing EU sustainability goals (the EU Green Deal, the EU Biodiversity Strategy, the Farm2Fork Strategy), and may incapacitate current effort halting and reverting the decline of pollinators (EU Pollinators Initiative).

The review of neonicotinoid insecticides by the US Environmental Protection Agency (US EPA) has been postponed to this year for the ingredients mentioned before with exception of thiacloprid but completed with dinotefuran. Its approval was voluntarily cancelled by registrant, as it is considered to be an endocrine disruptor and is, therefore, proven to be harmful to the hormonal balance of humans and animals. The banned ingredients were applied mainly as seed coating in different crops, but other application modes (e.g., spray) were also widespread in the past (Jeschke et al., 2011). Seed coating, as a preventive pest management practice for field crops (e.g., maize, sunflower, cotton), does not comply with the principles of integrated pest management (IPM) (Goulson, 2013), therefore new alternative pest control strategies were developed to substitute these banned ingredients (Furlan et al., 2021).

ALTERNATIVE METHODS

Before using any systemic insecticides in crop protection, the risks of a pest outbreak and the yield benefit should be assessed. Small damaging attacks of some soil pest such as wireworms are often compensated by the plants, and the effect of insecticides on yield and on the net income may be negligible. In some cases, increasing habitat complexity/ diversity to provide refuges and alternative hosts and food resources to predators and parasitoids may be effective. Upon high risks, alternative methods have been proposed. Among others the use of biological tools (e.g., attract-and-kill strategies) against soil pests, mating disruption based on the use of synthetic sexual pheromones, exclusion netting or natural-derived insecticides instead of synthetic chemicals were also tested. The main drawback is that these methods, in contrast to the prophylactic uses of highly toxic pesticides such as neonicotinoids, require a complex treatment. They are generally suggested to be applied in combination with or without low-risk pesticides for organic farming and IPM practices.

After the ban of neonicotinoids in the EU, increased use of certain outdated active ingredients (e.g., chlorpyrifos) has unfortunately been also observed, despite the fact, that the Green Deal aims to a 50% reduction of pesticide usage and a 25% increase in the proportion of organic farming by 2030 (Green Deal, 2022). These restrictions and withdrawals, of course, do not apply at other regions of the world, where these ingredients are still frequently used to prevent damages caused by insects, but this definitely affects the acceptability considerations in the RASFF system.

Among the methods in the pest control of goji berry, alternative to chemical crop protection using neonicotinoids or other PPPs, artificial defoliation against gall mites was also assessed (Li et al., 2019b). The procedure proposed enabled almost a complete defoliation, and *A. pallida* galls also fell off from the affected plants along with the defoliation process, and subsequent regenerated foliage escaped from mite attack. After defoliant application, the densities of mite galls decreased by more than 80% compared with those found in the pesticide treatment. Worthy of note, that the defoliation formulation consisted of a suspension concentrate containing the persistent diuron and the moderately persistent thidiazuron herbicide active ingredients. These generally are not approved in EU, only diuron is used in Bulgaria.

ORGANIC PRODUCTION IN EUROPE

Except for China and other Asian countries, as Malaysia, current goji production at a commercial scale initially began at the beginning of the century in Brazil, Canada and in the United States as well. The interest in Europe was skyrocketing after the intensive promotion of goji as a "superfruit" and the market demand increased very fast. In the European history of goji, legal violations related to the goji berry products, attempting to avoid the strict EU regulations, have unfortunately often occurred. The increasing demand for fresh goji berries in Europe and the pesticide residue problems with imported products on the European market also contributed to one initiative in Portugal to find out the best production processes for growing goji berries in Europe. In the frame of an EIP-AGRI project (Inspirational ideas: European goji berries, 2020.), cultivars of *L. barbarum* in organic production have been established, components in the fruits are monitored, and agricultural practices as well as soil and climatic conditions will be evaluated. There appears to be a growing interest in goji berry cultivation in other European countries, for example in Italy, Greece, Lithuania, Poland, and Romania. For instance, in Italy, as analysis performed at the border customs, found alarming levels of pesticide residue on the imported goji berries, mainly from China, the Italian farmers started to grow goji by their own, since 2007. In 2016, the Italian goji production reached 50 tons, making Italy the largest European producer (Knowles, 2016). In 2017, the area planted with goji was over 35 ha (15 ha in Veneto and 20 ha in Calabria), with a total of 60,000 plants, still being the largest farm in Europe (Zordan, 2016). Today, the Goji growers are mainly located in Calabria, Veneto, Puglia, Lazio and Tuscany. The Italian Capodaglio fresh goji berries are produced under the Organic Forest certification, the sustainable farming method promoting respect for natural ecosystems, genetic and biological cycles, and biodiversity (Zordan, 2016). Another Italian producer, Agricultural Company Leggero Luca, Canavese, Villareggia (Toscany) has now 3.5 ha of organic certified plantations, including a specialized nursery recognized and authorized by Piedmont regional office, and certified as Organic, that produces varieties of *L. barbarum* originally coming from the Ningxia region, China (ItalGoji, 2022). The same organic trend is apparent for the production of goji berry in Romania. The first company planted 2 ha of goji in 2011, with a variety develop by themselves, Erma, in Ciuperceni, Satu Mare. Their business grew and diversified with processed products, so the total planted area reached 11 ha in 2019 (GojiLand Romania, 2022). In Brașov area, another company started in 2014 the cultivation on 2.5 ha, with the Kronstadt variety, another variety registered in Romania and later diversify its activity both in developing their own brand and a certified organic nursery producing two of the Romanian varieties, that they later registered in the Official catalogue of cultivated plant varieties from Romania. Today, in Romania are registered 7 goji berry varieties in the *Official catalogue of cultivated plant varieties from Romania*, namely: 'Erma', 'Transilvania', 'Kirubi', ''Kronstadt', 'Bucur', 'Sara' and 'Anto', all being registered from 2017 to 2021 (ISTIS 2017, 2018, 2019, 2020, 2021). In Spain, GojiVital, S.L. started with 12 ha of goji plantation in the south of Spain (40,000 plants), since 2011, the company being GlobalGAP certified (GojiVital, 2022). Farmers from Poland have developed a special breeding technology for Goji berry, in greenhouses, the yield being increased starting the first year and fruiting period extended. The JB1 variety was produced *in vitro*, by a European company named Bio Tree Ltd (Biotree, 2022). In Hungary, goji cultivation is only at a start, triggered by improved yield performance of the variety Góliáth introduced in 2014. Beside small family orchards, goji berries are commercially cultivated and propagated in a 0.5 ha plantations at Ásotthalom (Csongrád-Csanád county).

CONCLUSIONS

The growing demand for goji berry as a healthy fruit is in contrast with its quality regarding its contamination with pesticides. Control activities of imported products, including pesticide residue analysis by RASFF, showed extensive amounts of residues and high numbers of pesticide active ingredients in goji berry. Among these contaminants, neonicotinoids, especially acetamiprid and imidacloprid, have frequently been found in shipment lots of imported goji berry from third countries. The latter active ingredient is currently banned in the EU, and other pesticides not approved in the EU have also been often detected.

Organic cultivation in the EU may satisfy the demand, and at the same time ensures the proper quality of the goji berry based products. The cultivated area with goji berry varieties is increasing yearly, Italy, Romania and Spain taking the lead of EU production, and the number of European registered varieties is constantly growing. The majority of producers are organic certified, but some pursue the ideal of having a chemical pesticide-free agriculture, and started gradually, first by adopting the

GlobalGAP certification, the set of standards for good agricultural practices. As consumer expectations regarding food quality are growing higher, it is expected that future pesticide residue analysis will show a decreasing trend of residues identified.

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