European plant protection product use: an assessment of data availability and implications for risk assessment on non-target organisms (#70098)

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European plant protection product use: an assessment of data availability and implications for risk assessment on non-target organisms.

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Besides the benefits of plant protection products (PPPs) for agricultural production, there is an increasing acknowledgement of the associated potential environmental risks. Here, we examine the feasibility of summarizing the extent of PPP usage at the country level, using Ireland as a case study, as well as at the European level. We used the area over which PPPs are applied as an initial step to assess the risks of non-target organisms to several tive ingredients (Als). In Irish agricultural systems, which are primarily grass-based, herbicides fluroxypyr and glyphosate are the most widely applied Als followed by the fungicides chlorothalonil and prothioconazole that are closely associated with arable crops. Although all EU countries are subjected to Regulation (EC) No 1185/2009, which sets the obligation of PPP usage data reporting at the national level, we only found usable data that met our criteria for Estonia, Germany, Finland, and Spain (4 of 30 countries reviewed). Overall, the most widely applied fungicide and herbicide were prothioconazole (22%, 9% and 5% of non-organic cultivated areas of Germany, Estonia and Ireland) and glyphosate (12%, 10% and 5% of non-organic cultivated areas of Estonia, Spain and Ireland), respectively. Several recommendations are proposed to tackle current data gaps and deficiencies in accessibility and usability in order to better inform environmental risk assessment and promote evidence-based policymaking.

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- 3 risk assessment on non-target organisms
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Abstract

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Besides the benefits of plant protection products (PPPs) for agricultural production, there is an increasing acknowledgement of the associated potential environmental risks. Here, we examine the feasibility of summarizing the extent of PPP usage at the country level, using Ireland as a case study, as well as at the European level. We used the area over which PPPs are applied as an initial step to assess the risks of nontarget organisms to several active ingredients (AIs). In Irish agricultural systems, which are primarily grass-based, herbicides fluroxypyr and glyphosate are the most widely applied AIs followed by the fungicides chlorothalonil and prothioconazole that are closely associated with arable crops. Although all EU countries are subjected to Regulation (EC) No 1185/2009, which sets the obligation of PPP usage data reporting at the national level, we only found usable data that met our criteria for Estonia, Germany, Finland, and Spain (4 of 30 countries reviewed) creall, the most widely applied fungicide and herbicide were prothioconazole (22%, 9% and 5% of non-organic cultivated areas of Germany, Estonia and Ireland) and glyphosate (12%, 10% and 5% of non-organic cultivated areas of Estonia, Spain and Ireland), respectively. Several recommendations are proposed to tackle current data gaps and deficiencies in accessibility and usability in order to better inform environmental risk assessment and promote evidence-based policymaking.

Keywords: pesticides; European policy; exposure; active ingredients



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Introduction

The risk of crop loss in modern agricultural production has been minimised over the past century through the use of Plant Protection Products (PPPs) (Kim et al., 2017). PPPs are synthetic or natural chemical products intended for preventing, destroying or controlling any pest causing harm to, or otherwise interfering with, the production, processing, storage, transport or marketing of plant-based food and agricultural commodities. Active Ingredients (AIs) contained in PPPs can include, among others, herbicides, fungicides, insecticides, acaricides, nematicides, molluscicides, or plant growth regulators, individually or in combination (FAO, 2006). Global PPP use (in terms of kg applied per hectare) rose steadily during the second half of the 20th century until the beginning of 21th century (Sharma et al., 2019; Zhang, 2018), and in 2009, worldwide PPP use was estimated to be ~3 billion tonnes annually (Pimentel, 2009). Globally, the quantity (tonnes of AI) of policides used is twice that of fungicides and almost four times that of insecticides (Zhang, 2018) tors influencing usage trends of PPPs include pest-control effectiveness and regulatory status (Barzman et al., 2015). New technologies are also important as modern, ightharpoonup description and to receive large quantities of herbicides, while small quantities of systemic AIs can be effective when applied as seed treatments (Baur et al., 2017). AIs such as glyphosate, 2,4-Dichlorophenoxyacetic acid (2,4-D) and paraquat are commonly used in a wide range of crop types whereas other AIs are limited to a particular crop, such as propanil use in rice production (Maggi et al., 2019). In 2014, amide-based compounds, including phenoxy hormone products and bipiridils led global herbicide usage by weight, whereas the inorganic compounds dithiocarbamates and triazoles account for the greatest use of fungicides, and organophosphates, pyrethroids and carbamates dominated insecticide usage (Zhang, 2018). While the use of PPPs has contributed to agricultural production and food security (Cooper and Dobson, 2007), research has revealed potentially harmful effects of the reliance on these substances. Drawbacks include issues such as human health impacts (Tanner et al., 2011; Alavanja and Bonner, 2012; Anderson and Meade, 2014), development of pest resistance, resurgence and secondary pest outbreaks



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(Evenson and Gollin, 2003; Oerke, 2006) and environmental contamination. The quantity of PPPs present in the environment is related to the amount applied, and also to the persistence of their associated AIs together with their metabolites. Residues of PPPs are widespread in soil where crops are grown, and 39% of residues found in the EU were considered persistent or very persistent (Silva et al., 2019), with the most common residues reported being glyphosate, AMPA (a metabolite of glyphosate) and DDE (dichlorodiphenyldichloroethylene), a derivative of DDT, which has not been licensed for use in the EU since 1983 (European Commission, 2003). The presence of PPPs and their residues in air, soil, water and food can harm non-target organisms, both in the area where they are applied and also the wider landscape (Zioga et al., 2020). Non-target organisms may come into contact with PPPs through direct application (Boutin et al., 2014), inadvertent contamination including drift (Morrissey et al., 2015), through their diet (de Snoo and Luttik, 2004), or they may be affected by a reduction in food availability (Eng et al., 2017; Hallmann et al., 2014). The PPP chemical family is an important determinant of impact on non-target organisms, for example, responses of bird species to insecticides vary from compromised migratory ability in the case of some neonicotinoids (Eng et al., 2017), to reproductive failure in the case of the organochlorine DDT (Ware, 1975). In addition, coformulants, also known as adjuvants, that are combined with AIs in PPPs include surfactants and solvents among other ingredients and are reliefly considered in risk assessment (Mesnage & Antoniou, 2018). Nevertheless, they can have synergistic or antagonistic effects on toxicity of AIs (Takacs et al 2017) or be toxic in themselves (Mesnage et al. 2019). Where they occur, such impacts are taxon-specific and may affect species that are different to the target organism. Some bees, for example, are significantly vulnerable to some fungicides and insecticides (Arena and Sgolastra, 2014; Bernauer et al., 2015; Main et al., 2018) and also to the synergistic impacts of both (Sgolastra et al., 2017). Related taxa can show different degrees of impact, as demonstrated in a meta-analysis of pesticide response in bees (Apiformes) that found non-Apis bees to be generally more sensitive to pesticides than Apis mellifera (Arena and Sgolastra, 2014). Because of the species-specific responses to different AIs, broad investigations of the impacts of PPPs on organisms can yield complex results, as illustrated by the review of Puglisi et al., (2012) on the response of microbial organisms to PPPs which found that herbicides,



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fungicides and insecticides stimulated terrestrial microbial biomass in some studies and suppressed it in others.

Concerns regarding the potential for unintended consequences of PPP use led to the establishment of international PPP usage databases. At a global level, the FAO pesticide use database (FAO 2019) presents the quantity in of the major pesticide groups (e.g. fungicides, insecticides, herbicides) used in or sold to the agricultural sector, and their constituent chemical families (e.g. carbamates, organophosphates). Additionally, the FAO pesticide indicators database (FAO, 2018), which in turn relies on the FAO land use database (FAO, 2018b), reports PPP usage in kg/ha of cropland for most countries in the world from 1990 to 2016, without specifying major pesticide groups or AIs. Likewise, the EUROSTAT database (EUROSTAT, 2019) also collates international pesticide usage data on PPP sales (kg of PPP major groups) in an accessible format, for EU Member States and another ten European countries (under Regulation (EC) No 1185/2009). Although these databases can be used to assess broad trends in global and European PPP usage (e.g. Zhang, 2018), they do not link pesticide use to specific crop types or indicate the area or geographic extent of pesticide application, both of which are important indicators of risk to nontarget organisms. The EU's Sustainable Use of Pesticides Directive (2009/128/CE; hereinafter SUD), established a framework for reducing risks to and impacts upon human health and the environment arising from pesticide use. Regulation (EC) No 1185/2009 was enacted to ensure that detailed, up-to-date and consistent information from Member States would be available for risk assessment and to monitor progress towards the goals of the SUD. Under this regulation, Member States are required to report the area treated with PPPs (ha) and the quantity applied (kg) for major crop types in their jurisdictions. The collection of statistics on PPP usage is an integral feature of the SUD, as it provides information required to assess the risks posed by PPP usage. However, no central, publicly-accessible data repository has been established for this information at an EU scale, and difficulties in comparing usage data across different nations have been reported (https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A52017DC0109). This is



important because failure to provide accessible, comparable data across EU Member States would inhibit risk assessment both at a Member State and EU-wide scale.

In this study, we aim to assess whether data on PPP usage collected under the SUD can be readily used to i) estimate total PPP usage in terms of national area of application, as a first step to determine the likelihood of PPP exposure for non-target organisms, ii) compare trends in PPP usage among European countries and iii printify the most widely used active ingredients across Europe. We used Ireland as a case study for the estimation of national PPP usage because agriculture is an important land use, accounting for 65% of the national land area (https://www.cso.ie/en/releasesandpublications/ep/p-syi/psyi2018/agri/cl/), and because a nationwide estimate for PPP use in Ireland has not previously been published. By answering these questions, we aim to demonstrate the availability, accessibility and usability of national PPP usage data in Europe. Overall, this study provides insight into the effectiveness of current legislative tools designed to evaluate the success of some SUD objectives.

Materials and methods

Irish data on usage of Plant Protection Products

The Irish Government Department of Agriculture, Food and the Marine's (DAFM) Pesticide usage reports (http://www.pcs.agriculture.gov.ie/sud/pesticidestatistics/) were used as the source of data relating to PPP usage in crops in the Republic of Ireland. These reports present Irish national statistics on PPP usage collected and collated by DAFM in line with EU Regulation (EC) No 1185/2009 in a four-year cycle for each crop type. Estimates of PPP usage provided by DAFM are based on a survey of a sample of farms within each crop type (Table 1). Fruit crops were divided into top fruit, including apples and other fruit grown on trees, and soft fruit such as strawberries, raspberries and blackcurrants. The vegetable and arable crop types were limited to those intended for human consumption, while vegetables and grain grown as animal fodder were included in the grassland and fodder report. Agricultural data held by DAFM were used to select farms growing the relevant crop type for each survey. Although the sample size varied among crop



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types, in each case, the sample was selected to be representative of the range of farms within each crop typein Ireland.

Farmers were asked to provide details of the PPPs applied, the date of application, the area where PPPs were applied and the specific crop grown in each field included in the survey in the 12 months prior to harvest. These data were then used to calculate the area of application (i.e. basic area) of AIs and of PPP groups (i.e. fungicides, herbicides, insecticides, plant growth regulators and other) at a national scale. Thus, if several applications of an AI or a PPP group occurred within a single land parcel, that land parcel was only included once in our analysis, and den licates were discarded. Specifically, the analysed data correspond to tables included in the DAFM pesticide usage reports that document the sample area, national cultivated area, and basic areas of application per AI and PPP group (i.e. fungicides, herbicides, insecticides, plant growth regulators and molluscicides; see, Tables C, 4 and 10-13 in 2014 Top Fruits Survey Report, Tables C, 4 and 10-18 in Soft Fruits Survey Report, Tables D, 4 and 10-28 in 2015 Vegetable Survey Report, Tables C, 3, and 9-19 in 2016 Arable Survey Report, and Tables C, 4, 10-20 in 2017 Grassland and fodder Survey Report). In these reports, AIs applied via seed treatments are explicitly reported. The tabulated data was provided upon request by DAFM in csv format. Full details of the methodology used to collect and process the raw data are presented in the pesticide usage reports (http://www.pcs.agriculture.gov.ie/sud/pesticidestatistics/).

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Sources of national PPP usage data for European countries were sought in September 2019 from the websites of national institutions with competencies in the implementation of the following European legislation: Residues - Regulation (EC) 396/2005, Sustainable Use Directive 2009/128/EU and Pesticide Regulation (EC) No 1107/2009. This information was made publicly available by the European Commission

(https://ec.europa.eu/food/sites/food/files/plant/docs/pesticides_legis_national-authorities_en.pdf) for Norway, Iceland and the 28 EU Member States. We selected the most up-to-date national databases that presented basic area of AIs applied to specified crop types in tabulated format. If





163 Ireland, the analysed tabulated data were provided by DAFM in csv format, and these data coincide with 164 tables included in the Irish PPP usage reports from which data are not directly exportable. Standardisation 165 levels among databases were assessed in terms of units used to report PPP usage and database structure (i.e. 166 levels of classification for crop types and PPP groups). 167 We assessed whether the accessible databases were comparable in terms of their time-span, reporting units and the categorization scheme used to define PPP groups and crop types. We then compared the basic area 168 of AIs in countries with comparable available data (Estonia, Finland, Germany and Spain) with those of 169 Ireland. We then organised AIs into standardised major PPP groups to facilitate comparison of broader 170 171 categories among countries. According to our standardised PPP groups, i) fungicides include fungicidesbactericides and fungicides mint growth regulator groups, ii) herbicides include herbicides-moss control 172 groups, iii) insecticides include insecticides-acaricides and iv) the group labelled "other" includes 173 174 molluscicides and pheromones. In addition, we extracted national cultivated areas (all the land dedicated to 175 agriculture in a country) and the national area of organic crops from the EUROSTAT databases of national 176 utilised agricultural area (code TAG00025) and organic crops (code org cropar) for the years where the 177 most recent PPP usage data was available for each country (i.e. 2015, 2018, 2017, 2017 and 2013 for 178 Estonia, Finland, Germany, Ireland and Spain, respect vely). These two databases were used to derive national non-organic cultivated areas. To compare the use of major PPP groups, we utilised maximum (total 179 180 non-organic cultivated area) and minimum (m is num basic area of the most widely used AI) values for each country. Presenting the area of AI application as a range was appropriate since, in many cases, multiple 181 182 different AIs were applied to the same parcel of land in a reporting period. Thus, adding together all of the 183 AI basic areas reported resulted in a clear overestimate of the area where PPPs are applied in each country, which in some cases exceeded the total national cultivated area. 184

such data were not presented in tabulated format, they were not considered to be accessible. In the case of



Risks of Plant Protection Products use on non-target organisms

In order to identify associated non-target organisms risks directly derived from the use PPPs, we consulted the Pesticide Properties DataBase developed and managed by the Agriculture and Environment Research Unit (AERU) at the University of Hertfordshire (https://sitem.herts.ac.uk/aeru/ppdb/en/index.htm). We collated ecotoxicological information on the risk level (low, medium, high) due to direct contact and/oral exposure of mammals, birds, fish, crustaceans, bees (*Apis*, *Bombus* and *Osmia* spp), and earthworms to the top most widely used AIs among the analysed countries. These risk levels are based on estimates of mean lethal dose (LD₅₀), mean lethal concentration (LC₅₀), and half maximal effective concentration (EC₅₀), depending on the taxonomic group.

Results and discussion

Irish usage of Plant Protection Products

The area of land under agricultural management in Ireland is dominated by grassland and fodder systems (94 %; Table 2), and grasslands alone account for 99.4% of this crop type. Arable crops are grown on 6 % of agricultural land, and vegetable and fruit cultivation each cover less than 0.1 %. Based on DAFM national statistics, PPPs are applied in more than 90% of the national area of arable, vegetable and fruit crops compared to under 10% of the national area occupied by grassland and fodder (Table 2). Although grassland and fodder systems have the lowest PPP use in proportion to area, the basic area of PPP use in grasslands and fodder exceeds that of other crop types, and this reflects the prominence of grasslands in Irish agriculture.

Herbicicism re the most widely used PPPs in Ireland with an estimated national basic area more than twice the basic area of fungicides (Fig. 1A). Fungicide is the second most widely used PPP group followed by insecticide and plant growth regulators. These represent 44%, 32% and 24% of the area where herbicides are applied, respectively. Molluscicide application, at only ~12500 ha nationally, is even less widespread. Although most PPPs are applied as spray, seed treatments represent a similar area to that treated with



fungicides (~280,000 ha, Fig. 1A). These trends are not in accordance with previous work by Zhao et al. (2013), who found that fungicides are the most utilized PPP group in Ireland when looking at units of mass per area (kg of AI per km²) and this probably reflects differences in the units used because the impact of repeated applications are reflected in measurement of AI application by mass, but not in basic area.

The area over which different PPP groups are utilized is strongly related to the crop type (Fig. 1B). For instance, arable crops have the largest proportional areas where fungicides (92.6 %), insecticides (95.5 %), molluscicides (74.3%), and plant growth regulators (99.6 %) are used. Likewise, PPPs are more widely applied as seed treatments in arable systems (93.9 %) although these can also be found in grassland and fodder systems (6.1 %). The greatest basic area of herbicide application occurs in grassland and fodder systems followed by arable crops, which represents 58.4 % and 41 % of total national area of herbicide application, respectively. Although PPPs are applied to 97 % of the cultivated area designated to soft and top fruits, the percentage area of PPP application in these systems remain marginal () compared to rest of crop types, given the small area they represent in the total Irish agricultural land (Table 2).

Because the usage intensity of PPPs varies depending on crop type, land-use changes over time are likely to have an impact on the presence of PPPs and their residues in the environment, and on exposure of non-target organisms. In Ireland, arable land is largely concentrated in the south-east of Ireland and a smaller area in the north-west (data not shown), and we may expect greater use of PPPs in these areas than in grass dominated landscapes. However, some parts of Ireland are characterised by frequent land use change between arable and grassland systems (Zimmermann and Stout, 2016), so the area in which PPPs and their residues occur may exceed the area of application captured in a 12-month SUD reporting period. In addition, a transition from a grassland-dominated system to a more diversified system that includes more arable and horticultural crops in the future (for example to adapt to climate change or diversifying markets) could lead to an increase in the extent of PPP use, if current agricultural practices persist.



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Regarding the area (ha) where specific AIs have been applied, the prevalence of some AIs over others can be observed for the main PPP groups (Fig. 2). For instance, the two most widely used AIs, the fungicides chlorothalonil, a non-systemic and broad-spectrum PPP (banned in EU since 2019), and prothioconazole, a systemic PPP, are each applied over more than 200,000 ha nationally. These two AIs were most frequently used in arable crops, especially in barley, wheat, oats and winter oilseed rape cultivation; however, they were also utilised in vegetable, and grassland and fodder cropping systems. The most widely used herbicides in Ireland were fluroxypyr, glyphosate and 4-chloro-2-methylphenoxy acetic acid (MCPA) with national basic areas ranging from approximately 129,000 ha to and 271,000 ha. These AIs are commonly applied in arable (barley, wheat and oat cultivations) and grassland and fodder systems, and glyphosate and MCPA are also used in vegetable and fruit crops. The most widely used herbicidal AIs are all systemic PPPs. Fluropyxyr and MCPA are synthetic auxins, while glyphosate is an enzymatic inhibitor of 5enolpyruvylshikimate-3-phosphate (EPSP) synthase. A single AI, lambda-cyhalothrin, dominates insecticide use and was applied in ~150,000 ha, including arable (barley, wheat, oats, oilseed rape and potatoes), grassland and fodder and vegetable (e.g. carrots, parsnips, cabbages, spinach) crops. Chlormequat, the dominant plant growth regulator, was used across a slightly larger area nationally (~170,000 ha), mainly in arable crops (barley, wheat and oats).

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Available data on European usage of Plant Protection Products

While PPP sales data show a high level of harmonisation and a common repository for all EU countries (EUROSTAT, 2019), we have found that available PPP usage data in terms of area of application is sparse, difficult to access and the different reporting formats makes usability challenging. Only four EU Member States (Estonia, Finland, Germany and Spain) reported PPP usage data that were available, accessible and in a format that enabled a direct assessment and comparison. For the remaining countries, PPP usage data were either not publically available (19 of 30 total), or in a non-accessible and non-usable format due to the





reporting units (three countries) and format (e.g. text reports in native languages where data tables were embedded in PDF files; four countries including Ireland). Reporting frequency varies among countries, as is reflected in the different database publication dates (Table 3). Although the mandatory frequency set by the SUD is five years, some countries report the data annually (Germany and Estonia), while others have not reported public data since 2013 (Spain) or report different crop types every year (Ireland), which makes a comparison of PPP usage across countries in the same year impossible.

The structure of these databases also differed in terms of the PPP groups and the reported crop types that

accompany usage data. Different disaggregation levels are reported for different countries with Ireland and Estonia having the highest number of disaggregation levels for PPP groups and crop types, respectively. Furthermore, the type of data reported are not consistent among databases. For instance, only Ireland and Spain include molluscicides as a major PPP group, while phero pipe are only included in German data. The four PPP groups that are present in all national databases are fungicides, herbicides, insecticides and plant growth regulators, assuring comparability among countries for these major PPP groups. However, for some countries, some of these PPP groups include pesticides with more than one function, such as insecticides-acaricides, fungicides-bactericides, herbicide-moss killers, and fungicides-plant growth regulators. The categorisation of crop-types is more variable than that of PPP groups, and we could not find any crop type that is common for all countries. Among all national databases, there were 11 crop types found in more than one country for all disaggregation levels: potatoes (2 countries), vegetables (3), winter (4) and spring wheat (3), cabbages (2), carrots (3), onions (3), peas (2), and spring (2) and winter barley (3). The two databases with the highest crop-type disaggregation levels, Ireland and Estonia, are also the

With regard to the units used for the area of applied PPP, only the Spanish and Irish databases include values of both basic and treated/sprayed areas, which together with the amount of AI applied per unit of area (e.g. in kg/ha) can give us an estimate of PPP use intensity. However, working with intensity data in

most similar in their categorisation of crop types.





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terms of the weight applied per unit area can be problematic, as the quantity of the AI applied is closely related to its toxicity, so the threat to non-target organisms may not be proportional to the weight of a substance that is applied. Furthermore, the quantity in a single application may be misleading. For example, trends of PPP usage over time from Northern Ireland have shown that while the weight of PPP per application to arable crops has decreased, the number of applications to the same piece of land have increased, resulting in a similar total quantity of PPP applied over time (Jess et al., 2018). Since both measures, area and intensity of application, carry complementary information, further specifications must be set on how exactly data should be reported using these specified units in order to enable data comparison across EU countries. Generally, no distinction is made between the application method (seed treatment vs spray) used to apply each AI. Only Ireland provided explicit information on the area where AIs where applied via seed treatments whereas the Finnish database excluded those areas that were exclusively exposed to seed- or seedling-treatment. Uncertainty estimates were only incorporated in the German database, where confidence intervals of basic areas were included (Table 3). Although we successfully compared pesticide usage data from a small number of countries (section 3.3), the observed reporting differences among EU member states made comparisons of PPP usage data at an EU scale difficult, even for countries with publicly available usage data, confirming the findings of the European Commission (2017). This inhibits the possibility of using this information to assess the potential hazards and risks of PPP use for a variety of purposes, including the assessment of potential harmful effects on non-target organisms and ecosystem functions and services. According to Schulz et al. (2021), the unavailability of open-access pesticide use data occurs globally, which hampers the application of advanced risk assessment approaches developed by the scientific community (e.g. Sponsler et al. 2019) to evaluate one of the crucial drivers of global biodiversity decline.



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Comparison of Plant Protection Products usage among EU Member States

For countries where data were available, agricultural systems differ in terms of area and crop types. For instance, a higher proportion of the national area is dedicated to agriculture in Ireland (65%), Germany (47%) and Spain (46%), compared to Estonia (22%) and Finland (7%). In addition, more than half of the total cultivated area was dedicated to arable crops for all countries except Ireland, where the agricultural 310 311 system is clearly grassland-dominated (90% of total cultivated area; Table 4). 312 Almost all of the most widely used AIs fungicides and herbicides in each country (Table 5), with the exception of plant growth regulators chlormequat and trinexepac in Ireland and Germany, and the 313 314 insecticide dimethoate in Spain. A number of AIs occur among the top five for several countries. For example, the systemic fungicide prothioconazole is notable for its application to 22% of the non-organic 316 cultivated area (CA) in Germany, a figure equivalent to 9.5% of the total land area of Germany, and is also extensively used in Estonia (9% CA) and Ireland (5% CA). Another widely used systemic fungicide is 318 tebuconazole, which is reported within the top five AIs of Estonia (14% CA) and Germany (21% CA). The most widespread herbicidal AI reported is glyphosate, which is among the most widely applied AIs in 320 Ireland (> 5% CA), Estonia (> 10% CA) and Spain (> 12% CA). This prevalence of glyphosate, prothioconazole and tebuconazole among the countries assessed is in accordance with results from Silva et 322 al. (2019), who showed that residues of these compounds are commonly found in European soils. Importantly, most of the Silva et al. (2019) study's soil samples contained residues of multiple AIs, a fact that is particularly relevant for non-target organisms, which may often be exposed to more than one compound at a time potentially leading to combined negative impacts of multiple AIs, a phenomenon 326 commonly known as the "cocktail effect" (Relyea, 2009; Rivera-Becerril et al., 2017; Soil Association and Pesticide Action Network UK, 2019). Unfortunately, this trend cannot be investigated in PPP-use databases currently available. 328 Although the majority of the most widely used AIs for all countries are currently approved for use, Irish 330 results showed that both chlorothalonil and clothianidin were widely applied during the last reporting period



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even though neither of these compounds are currently approved for use in the EU (Fig. 2). The decision to cease using these two substances at EU level came into effect after data were collected for the most recent usage reports in Ireland. It will be several years before the effects of EU regulation on AI use can be discerned in Irish national PPP usage data due to the legally-required five year reporting frequencies, and this impedes the short-term evaluation of the effectiveness of EU pesticide regulation.

The countries with the largest non-organic cultivated areas (CA), and therefore the largest potential area of PPP usage, was Spain and Germany. Spanish CA was 26 times the CA of Estonia, 11 times that of Finland, 5 times that of Ireland and 1.4 times that of Germany. However, the country with the greatest minimum application areas of fungicides and plant growth regulators was Germany, where prothioconazole is applied to 3,383 thousand ha and trinexapac was applied to 3,033 thousand ha (Table 5). The minimum potential area of herbicide application was similar in Spain and Germany, but insecticide use was slightly greater in Spain, where dimethoate is applied to 826,432 ha, than in Germany, where thiacloprid is applied to 81,432 ha, ha. With the exception of plant growth regulators, the minimum area of application for all groups of PPP as a proportion of the total CA was smallest in Ireland (Fig. 3; right panels). Based on minimum potential areas, both herbicide (64% CA) and insecticide (7% CA) were most widely applied in Finland, and fungicides are applied to a larger proportion of CA in Germany (22%) than in any other country. As a general trend for all the countries assessed, the minimum potential area of insecticide application was smaller than that for herbicides or fungicides (from 1.6 to 8.8 times smaller) and represents less than 10 % of CA. The minimum potential application of plant growth regulators as a proportion of total cultivated area was highly variable, accounting for the smallest proportional area of all PPP groups in Spain (0.1%), and the second greatest proportional area in Germany (19.5%). Overall, differences among countries are probably related to distinct farming systems, crop preferences, agronomic culture and traditions, and/or climatic conditions making areas more or less susceptible to pest damage.





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Where basic areas were reported for AI use, as was the case for the Member States included in our analysis (except for Finland), a comparison of AI use in different countries could be made. However, this approach does not allow for an estimate of the total area within a country treated with broader PPP groups, as different Als can be applied to the same land parcel either individually or in combination. For instance, in the Irish dataset for arable fields, a maximum and average number of 29 and 12 different AIs were used per field, respectively. It is therefore very difficult to compare overall PPP usage across different countries because they are reported at AI level with no information on the degree of overlap in the area of application. The approach we chose to overcome this limitation consisted in using the basic area of the most widely applied Als per PPP group as an e water attended at the minimum value of total basic area per PPP group. However, the risk of underestimation may be high, particularly for those crops where a combination of AIs is commonly applied. In fact, a comparison of results based on broader PPP group- and AI-disaggregated basic areas (Fig. 3 and Fig. 1, respectively), reveals that the basic areas for PPP groups are from 1.05 (for plant growth regulators) to 2.71 (for herbicides) times higher than the estimate of minimum area affected based on AI usage data. The data used in our study (i.e. basic area of PPP use) are broadly consistent with available PPP sales data available (EUROSTAT, 2019) for the years assessed. Annual PPP sales (in kg) were the highest in Germany and Spain, both of which are the countries with the largest basic area treated with PPPs. Interestingly, sales of fungicides in Spain were much higher than that of Germany, even though our analysis showed the estimated basic area of fungicide use in Spain was lower. This may indicate that fungicides are applied more often to the same parcel of land in Spain than in Germany (i.e. there is a higher intensity of use). It may also indicate that a greater variety of substances are being used (and the pre not captured in our minimum estimate which was based on the most widely used substance), or that the substances being used are applied in greater quantities.



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Implications for non-target organisms



Our results suggest that how non-target organisms come in contact with the most widely used AIs can result in varying risk levels among the countries assessed (Table 6). For instance, fish and aquatic invertebrates may be under high risk over at least 3.4% of the area of Ireland, especially during the hours following application, of the broad-spectrum fungicide chlorothatonil since its partial degradation is relatively fast in water sediments and bodies (see Table S1 in supplementary material). Likewise, the application of the organophosphate insecticide dimethoate poses high risk over birds and several bee species (Apis, Bombus and Osmia spp) across 1.6% of the land surface of Spain at least. Copper oxychloride, epoxiconazole and tebuconazole are highly persistent in soil and water (Table S1) and this, regardless of the application intensity, may be linked to long-lasting moderate risks for fish, aquatic invertebrates and different bee species in 3%, 17.39% and 2.5% of the country areas of Estonia, Germany and Spain, respectively. In fact, tebuconazole has been recently added to the Surface Water Watch List (under the Directive 2013/39/EU; https://eur-lex.europa.eu/legalcontent/EN/TXT/?uri=uriserv:OJ.L .2020.257.01.0032.01.ENG&toc=OJ:L:2020:257:TOC), which identifies substances that must be monitored with high priority across EU Member States due to its potential risk to or via the aquatic environment. However, the potential harmful effects on non-target organisms are likely to be wider than discussed above when considering drift, cocktail effects and diffuse contamination, in addition to point-source contamination of the target areas.



Although we chose to focus on basic area as the initial data needed to assess the extent of potential contact between non-target organisms and PPPs, a second step would be to include the intensity of PPP use in our analysis, as this could increase the likelihood of PPP contact for non-target organisms. Once the area of exposure and intensity of use are known, these can be combined with AI-specific factors such as persistence and water solubility to develop risk-assessments as proposed by Sponsler et al. (2019). Another relevant issue is the suitability of including other sectors apart from agriculture, such as forestry or amenity use, in the PPP usage data reported by EU Member States. For example, forestry represents a significant land-use in some European countries in terms of PPP use. This is the case in Ireland, where forestry covers over 770,000 ha, an area greater than that of arable land, and where both herbicide and insecticide application occurs. However, data on these other sectors are not currently summarized or collected with the agricultural data, which makes the estimation of national or European total PPP use challenging and hampers the development of holistic risk assessments at a landscape level that needs the integration of spatial-explicit PPP use information with other data sets (e.g. soil type maps) at national scale.

Concisions

Recommendations to improve national statistics of PPP use in EU

Our study demonstrates the current gaps in data collection and reporting on national PPP use across the EU, particularly for indicators of potential PPP exposure of non-target organisms, such as the area of PPP application. Unlike sales data for EU Member States, PPP usage data are not publicly available for all countries. When available, these data are presented in heterogeneous format, different units, and the crop classifications and PPP groups are not systematic. Our results demonstrate that the area of PPP application can serve as a proxy of PPP residues present across the European countries and hence, as a first crucial step to upscale potential risks on non-target organisms and ecosystems to national and international scales. While we have shown that it is possible for different countries to record comparable PPP usage data, the





current regulations governing pesticide usage data reporting do not result in consistent and accessible data 421 422 at EU scale. The recently published EU "Farm to fork strategy for a fair, healthy and environmentally-friendly food 423 system" states that the European Commission will also propose changes to the 2009 regulation concerning 424 425 statistics on PPPs to overcome data gaps and promote evidence-based policymaking, and this would be an 426 important step in ensuring the usability of these data for multiple purposes (see also Mesnage et al. 2021). 427 Based on our results, our main recommendations are: a) Consistent reporting requirements around data format, disaggregation levels of PPP groups and 428 crop types and reporting years. 429 b) Mandatory reporting of basic and treated areas. Basic areas tell us about the geographical extent of 430 431 PPP and respective AI use, however, treated areas give us information about how frequently these PPPs and AIs are applied in a given crop type or country. Both metrics are useful as the information 432 they carry is complementary. 433 c) Mandatory reporting of summary data. On the basis of this review the most appropriate estimate to 434 435 accurately assess the country area exposed to the different PPP groups is the national basic area. d) Controlled vocabularies (e.g. same PPP groups for all countries) to assure harmonisation among 436 national databases. Additionally, in the case of crop types, these should be named in accordance 437 438 with the cultivation types shown in EUROSTAT database (Table 3). 439 e) Consideration of all sectors that use PPPs, including sectors such as forestry and amenity use. Accessibility and usability of all these data should be guaranteed since current data repository 440 441 platforms are already available. 442 The EU has one of the most developed systems for collection of national pesticide usage statistics globally. 443 Increasing the uniformity and accessibility of pesticide usage data in the EU would further enhance their



444	usefulness to design and evaluate policy actions that pursue a more sustainable agriculture that minimizes
445	biodiversity decline.
446	
447	Acknowledgements
448	This work was funded by an Irish Department of Agriculture, Food and the Marine award (PROTECTS
449	Project Ref: 17/S/232). ALB was also supported by a Juan de la Cierva-Formación postdoctoral contract
450	from the Spanish Ministry of Science, Innovation and Universities (FJC2018-038192-I) and through the
451	BC3 María de Maeztu excellence accreditation (MDM-2017-0714).
452	Data Availability
453	Data used in this study is publicly available at the URLs provided in Table 3.
454	
455	Author Contribution
456	All authors contributed to the conception of the study, revised the manuscript and proposed
457	improvements. ALB and AD collected and assessed the data and wrote the manuscript.
458	
459	References
460	Alavanja, M.C.R., Bonner, M.R., 2012. Occupational pesticide exposures and cancer risk: A review. J.
461	Toxicol. Environ. Heal Part B Crit. Rev. https://doi.org/10.1080/10937404.2012.632358
462	Anderson, S.E., Meade, B.J., 2014. Potential Health Effects Associated with Dermal Exposure to
463	Occupational Chemicals. Environ. Health Insights 8s1, EHI.S15258.
464	https://doi.org/10.4137/EHI.S15258
465	Arena, M., Sgolastra, F., 2014. A meta-analysis comparing the sensitivity of bees to pesticides.
466	Ecotoxicology 23, 324–334. https://doi.org/10.1007/s10646-014-1190-1



- Barzman, M., Bàrberi, P., Birch, N.A.E., Boonekamp, P., Dachbrodt-Saaydeh, S., Graf, B., Hommel, B.,
- Jensen, J.E., Kiss, J., Lamichhane, J.R., Messéan, A., Moonen, A.-C., Ratnadass, A., Ricci, P., Sarah,
- J.-L., Sattin, M., 2015. Eight principles of integrated pest management. Agron. Sustain. Dev 35, 1199–
- 470 1215. https://doi.org/10.1007/s13593-015-0327-9
- Baur, M., Beckie, H.J., Branch, T.-O., Canada, A.-F., Giesler, L.J., Leppla, N.C., Schroeder, J., Bradley,
- 472 C., 2017. Crop Protection Contributions toward Agricultural Productivity.
- 473 Bernauer, O., Gaines-Day, H., Steffan, S., 2015. Colonies of Bumble Bees (Bombus impatiens) Produce
- Fewer Workers, Less Bee Biomass, and Have Smaller Mother Queens Following Fungicide Exposure.
- 475 Insects 6, 478–488. https://doi.org/10.3390/insects6020478
- Boutin, C., Strandberg, B., Carpenter, D., Mathiassen, S.K., Thomas, P.J., 2014. Herbicide impact on non-
- 477 target plant reproduction: What are the toxicological and ecological implications? Environ. Pollut.
- 478 185, 295–306. https://doi.org/10.1016/j.envpol.2013.10.009
- 479 Cooper, J., Dobson, H., 2007. The benefits of pesticides to mankind and the environment. Crop Prot.
- 480 https://doi.org/10.1016/j.cropro.2007.03.022
- de Snoo, G.R., Luttik, R., 2004. Availability of pesticide-treated seed on arable fields. Pest Manag. Sci. 60,
- 482 501–506. https://doi.org/10.1002/ps.824
- 483 Eng, M.L., Stutchbury, B.J.M., Morrissey, C.A., 2017. Imidacloprid and chlorpyrifos insecticides impair
- migratory ability in a seed-eating songbird. Sci. Rep. 7. https://doi.org/10.1038/s41598-017-15446-x
- 485 European Commission, 2017. REPORT FROM THE COMMISSION TO THE EUROPEAN
- PARLIAMENT AND THE COUNCIL on the implementation of Regulation (EC) No 1185/2009 of
- the European Parliament and of the Council of 25 November 2009 concerning statistics on pesticides.
- 488 Brussels.
- 489 European Commission, 2003. Presence of persistent chemicals in the human body results of Commissioner



- Wallstrom's blood test MEMO/03/219. Bruxelles.
- 491 EUROSTAT, 2019. Data product with code "tai02". Available at
- https://ec.europa.eu/eurostat/databrowser/view/tai02/default/table?lang=en).
- 493 EUROSTAT, 2019b. Data product with code "aei_fm_salpest09". Available at
- https://ec.europa.eu/eurostat/product?code=AEI FM SALPEST09&mode=view).
- Evenson, R.E., Gollin, D., 2003. Assessing the Impact of the Green Revolution, 1960 to 2000. Science (80-.
- 496). 300, 758–763. https://doi.org/10.1126/science.1078710
- 497 FAO. 2006. International Code of Conduct on the Distribution and Use of Pesticides. Guidelines on
- 498 Efficacy Evaluation for the Registration of Plant Protection Products. Available at
- http://www.fao.org/fileadmin/templates/agphome/documents/Pests Pesticides/Code/Efficacy.pdf.
- 500 FAO, 2018. FAOSTAT, Agri-environmental Indicators / Pesticides. Available at
- 501 http://www.fao.org/faostat/en/#data/EP
- 502 FAO, 2018b, FAOSTAT Land Use domain. Available at http://www.fao.org/faostat/en/#data/RL
- 503 FAO, 2019. FAOSTAT Pesticides Use Dataset. Available at http://www.fao.org/faostat/en/#data/RP
- Hallmann, C.A., Foppen, R.P.B., Van Turnhout, C.A.M., De Kroon, H., Jongejans, E., 2014. Declines in
- insectivorous birds are associated with high neonicotinoid concentrations. Nature 511, 341–343.
- 506 https://doi.org/10.1038/nature13531
- 507 Jess, S., Matthews, D., Murchie, A., Lavery, M., 2018. Pesticide Use in Northern Ireland's Arable Crops
- from 1992–2016 and Implications for Future Policy Development. Agriculture 8, 123.
- 509 https://doi.org/10.3390/agriculture8080123
- 510 Kim, K.-H., Kabir, E., Jahan, S.A., 2017. Exposure to pesticides and the associated human health effects.
- 511 Sci. Total Environ. 575, 525–535. https://doi.org/10.1016/J.SCITOTENV.2016.09.009



- 512 Maggi, F., Tang, F.H.M., la Cecilia, D., McBratney, A., 2019. PEST-CHEMGRIDS, global gridded maps
- of the top 20 crop-specific pesticide application rates from 2015 to 2025. Sci. Data 6, 1–20.
- 514 https://doi.org/10.1038/s41597-019-0169-4
- 515 Main, A.R., Webb, E.B., Goyne, K.W., Mengel, D., 2018. Neonicotinoid insecticides negatively affect
- performance measures of non-target terrestrial arthropods: a meta-analysis. Ecol. Appl. 28, 1232–
- 517 1244. https://doi.org/10.1002/eap.1723
- 518 Mesnage, R., Antoniou, M.N., 2018. Ignoring Adjuvant Toxicity Falsifies the Safety Profile of Commercial
- Pesticides. Front. Public Heal. 5, 1. https://doi.org/10.3389/fpubh.2017.00361
- 520 Mesnage, R., Benbrook, C., Antoniou, M.N., 2019. Insight into the confusion over surfactant co-formulants
- in glyphosate-based herbicides. Food Chem. Toxicol. https://doi.org/10.1016/j.fct.2019.03.053
- 522 Mesnage, R., Straw, E.A., Antoniou, M.N., Benbrook, C., Brown, M.J.F., Chauzat, M.-P., Finger, R.,
- Goulson, D., Leadbeater, E., López-Ballesteros, A., Möhring, N., Neumann, P., Stanley, D., Stout,
- J.C., Thompson, L.J., Topping, C.J., White, B., Zaller, J.G., Zioga, E., 2021. Improving pesticide-use
- data for the EU. Nature Ecology & Evolution 5, 1560-1560. https://doi.org/10.1038/s41559-021-
- 526 01574-1
- 527 Morrissey, C.A., Mineau, P., Devries, J.H., Sanchez-Bayo, F., Liess, M., Cavallaro, M.C., Liber, K., 2015.
- Neonicotinoid contamination of global surface waters and associated risk to aquatic invertebrates: A
- review. Environ. Int. https://doi.org/10.1016/j.envint.2014.10.024
- 530 Oerke, E.-C., 2006. Crop losses to pests. J. Agric. Sci. 144, 31–43.
- 531 https://doi.org/10.1017/S0021859605005708
- 532 Pimentel, D., 2009. Pesticides and pest control, in: Integrated Pest Management. Springer Netherlands, pp.
- 533 83–87. https://doi.org/10.1007/978-1-4020-8992-3 3
- Puglisi, E., 2012. Response of microbial organisms (aquatic and terrestrial) to pesticides. EFSA Support.



535 Publ. 9. https://doi.org/10.2903/sp.efsa.2012.en-359 Relyea, R.A., 2009. A cocktail of contaminants: How mixtures of pesticides at low concentrations affect 536 aquatic communities. Oecologia 159, 363-376. https://doi.org/10.1007/s00442-008-1213-9 537 Rivera-Becerril, F., van Tuinen, D., Chatagnier, O., Rouard, N., Béguet, J., Kuszala, C., Soulas, G., 538 Gianinazzi-Pearson, V., Martin-Laurent, F., 2017. Impact of a pesticide cocktail (fenhexamid, folpel, 539 540 deltamethrin) on the abundance of Glomeromycota in two agricultural soils. Sci. Total Environ. 577, 541 84–93. https://doi.org/10.1016/j.scitotenv.2016.10.098 Sgolastra, F., Medrzycki, P., Bortolotti, L., Renzi, M.T., Tosi, S., Bogo, G., Teper, D., Porrini, C., 542 543 Molowny-Horas, R., Bosch, J., 2017. Synergistic mortality between a neonicotinoid insecticide and 544 an ergosterol-biosynthesis-inhibiting fungicide in three bee species. Pest Manag. Sci. 73, 1236–1243. 545 https://doi.org/10.1002/ps.4449 Sharma, A., Kumar, V., Shahzad, B., Tanveer, M., Sidhu, G.P.S., Handa, N., Kohli, S.K., Yadav, P., Bali, 546 A.S., Parihar, R.D., Dar, O.I., Singh, K., Jasrotia, S., Bakshi, P., Ramakrishnan, M., Kumar, S., 547 Bhardwaj, R., Thukral, A.K., 2019. Worldwide pesticide usage and its impacts on ecosystem. SN 548 549 Appl. Sci. 1, 1–16. https://doi.org/10.1007/s42452-019-1485-1 550 Silva, V., Mol, H.G.J., Zomer, P., Tienstra, M., Ritsema, C.J., Geissen, V., 2019. Pesticide residues in 551 European agricultural soils - A hidden reality unfolded. Sci. Total Environ. 653, 1532-1545. https://doi.org/10.1016/j.scitotenv.2018.10.441 552 553 Soil Association, Pesticide Action Network UK, 2019. The cocktail effect. How pesticide mixture may be 554 harming human health and the environment. Sponsler, D.B., Grozinger, C.M., Hitaj, C., Rundlöf, M., Botías, C., Code, A., Lonsdorf, E. V., 555 556 Melathopoulos, A.P., Smith, D.J., Suryanarayanan, S., Thogmartin, W.E., Williams, N.M., Zhang, 557 M., Douglas, M.R., 2019. Pesticides and pollinators: A socioecological synthesis. Sci. Total Environ.



- 558 https://doi.org/10.1016/j.scitotenv.2019.01.016
- 559 Schulz, R., Bub, S., Petschick, L.L., Stehle, S., Wolfram, J., 2021. Applied pesticide toxicity shifts toward
- plants and invertebrates, even in GM crops. Science (80-.). 372, 81–84.
- 561 https://doi.org/10.1126/science.abe1148
- 562 Takács, E., Klátyik, S., Mörtl, M., Rácz, G., Kovács, K., Darvas, B., Székács, A., 2017. Effects of
- neonicotinoid insecticide formulations and their components on Daphnia magna–the role of active
- ingredients and co-formulants. Int. J. Environ. Anal. Chem. 97, 885–900.
- 565 https://doi.org/10.1080/03067319.2017.1363196
- Tanner, C.M., Kamel, F., Ross, G.W., Hoppin, J.A., Goldman, S.M., Korell, M., Marras, C., Bhudhikanok,
- G.S., Kasten, M., Chade, A.R., Comyns, K., Richards, M.B., Meng, C., Priestley, B., Fernandez, H.H.,
- 568 Cambi, F., Umbach, D.M., Blair, A., Sandler, D.P., Langston, J.W., 2011. Rotenone, Paraquat, and
- Parkinson's Disease. Environ. Health Perspect. 119, 866–872. https://doi.org/10.1289/ehp.1002839
- 570 Ware, G.W., 1975. Effects of DDT on reproduction in higher animals. Residue Rev.
- 571 https://doi.org/10.1007/978-1-4612-9863-2 5
- 572 Zhang, W., 2018. Global pesticide use: Profile, trend, cost / benefit and more. Proc. Int. Acad. Ecol.
- 573 Environ. Sci. 8, 1–27.
- 574 Zhao, Y.Q., Singleton, P., Meredith, S., Rennick, G.W., 2013. Current status of pesticides application and
- 575 their residue in the water environment in Ireland. Int. J. Environ. Stud. 70, 59–72.
- 576 https://doi.org/10.1080/00207233.2012.752557
- 577 Zimmermann, J., Stout, J.C., 2016. Underestimating neonicotinoid exposure: how extent and magnitude
- 578 may be affected by land-use change. Environ. Sci. Pollut. Res. 23, 7050–7054.
- 579 https://doi.org/10.1007/s11356-016-6159-6
- 580 Zioga, E., Kelly, R., White, B., Stout, J.C., 2020. Plant protection product residues in plant pollen and





nectar: A review of current knowledge. Environ. Res. https://doi.org/10.1016/j.envres.2020.109873



Figure 1

Basic area of application of top PPPs in Ireland

Estimated national basic area (1 pma) for the main groups of plant protection products (PPP) used in Ireland (A) together with the percentage applied to the different crop type areas (B). Soft and top fruit crops are not visible; while PPP usage is high in these systems, they represent a very small geographic area nationally. Data source: Irish Government Department of Agriculture, Food and the Marine's Pesticide (DAFM) usage reports.

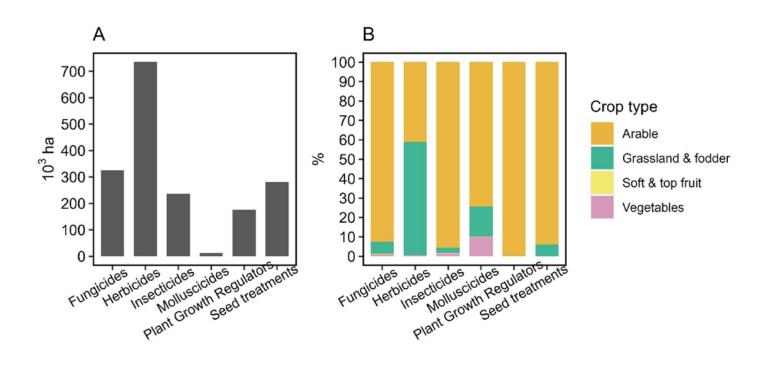




Figure 2

Estimated national basic area of PPP groups in Ireland

Estimated national basic area (10³ ha) of the most widely used active ingredients applied in Irish agriculture. Agricultural area (grey) includes grassland and fodder, soft and top fruits, vegetables and arable systems. Data source: Irish Government Department of Agriculture, Food and the Marine's Pesticide (DAFM) usage reports.



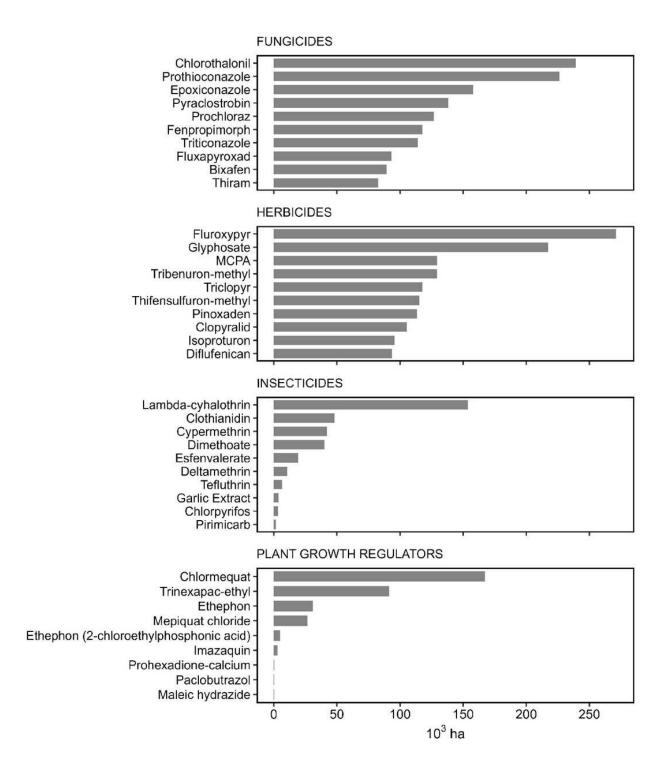




Figure 3

Minimum and maximum potential areas of PPP group application per country

Minimum (black) and maximum potential areas (grey) where PPP active ingredients were applied in thousands of hectares and as a percentage of the total non-organic cultivated area per country and per PPP group. For all countries except for Finland, the minimum area is equal to the basic area of the most widely applied AI for each PPP group because it was not possible to determine the degree of overlap PPP application for the different active ingredients belonging to the same PPP group. In case of Finish database, basic areas provided are disaggregated by PPP group but not by active ingredients. The maximum potential area represents the entire national non-organic cultivated areas for all countries. Data sources: public accessible data of national plant protection products use, and EUROSTAT TAG00025 and org_cropar databases.



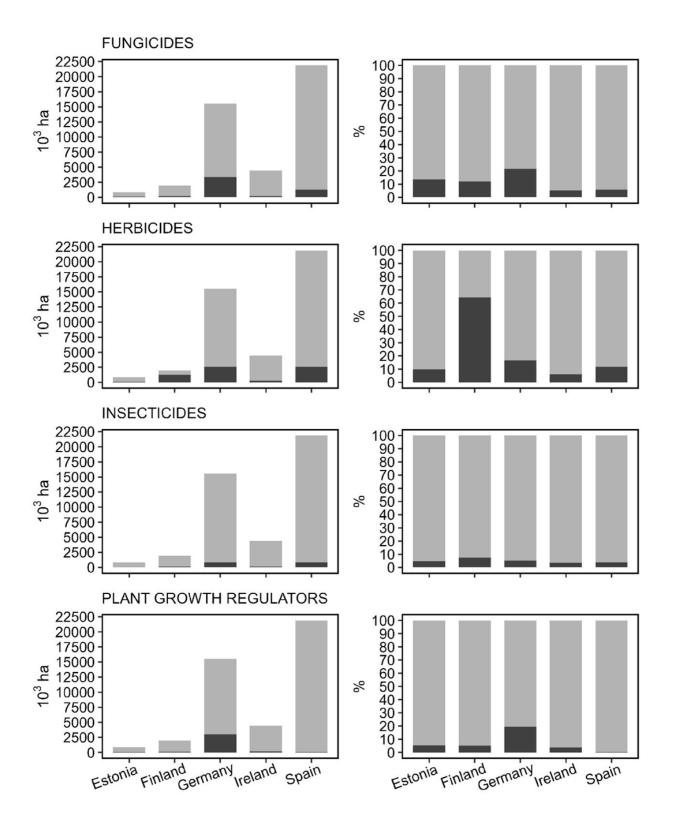




Table 1(on next page)

Summary of Irish pesticide usage data used

Crop type, the year for which data were collected and sample size for each of the Irish pesticide usage surveys included in this report. (Sample size includes the number of farms, area surveyed and proportion of each crop type surveyed)



Crop type	Survey year	Number of farms surveyed	Area surveyed (ha)	Area surveyed (% of total crop area)
Top fruit	2014	23	492	79.7
Soft fruit	2014	26	187	56.0
Vegetable	2015	109	2902	61.2
Arable	2016	260	23,199	7.6
Grassland and fodder	2017	530	33,187	0.7



Table 2(on next page)

Areas related to PPP application in Ireland

Sample, national cultivated and national basic areas where PPPs were applied (in ha and % of total cultivated area) by crop type for Ireland. Data source: Irish Government Department of Agriculture, Food and the Marine's Pesticide (DAFM) usage reports.





Crop type	Year sampled	Sample area (ha)	National cultivated area (ha)	National basic area of PPP application (ha)	% total cultivated area in which PPPs are applied
Arable	2016	23,199	306,092	305,744	99.89
Soft & top fruits	2014	679	951	924	97.18
Vegetables	2015	2,831	4,635	4,314	93.07
Grassland & fodder	2017	33,187	4,652,044	431,154	9.27



Table 3(on next page)

Summary of PPP usage data for EU countries used

Accessed PPP usage public data for each country together with the disaggregation levels utilized to classify PPP groups (i.e. disaggregation levels 1 and 2 corresponding to major groups, and active ingredients, respectively) and crop types (disaggregation levels from 1 to 3 corresponding to broader to more specific crop types). Blank cells mean that no data was available for a given disaggregation level.



]	PPP groups Crop types			es				
Country	Year	Disaggregation Disaggregation levels levels		tion	Area	Uncertainty indicator provided			
		1	2	1	2	3			
Estonia ¹	2015	4	140	11	8	45	Basic	None	
Finland ²	2018	4	-	19	-	-	Basic	None	
Ireland ³	2014-2017	6	215	5	-	56	Basic & treated	None	
Germany ⁴	2017	5	222	9	-	-	Basic	Confidence interval	
Spain ⁵	2013	6	272	7	-	-	Basic & treated	None	

¹Data source: http://pub.stat.ee/px-web.2001/Dialog/varval

.asp?ma=EN2081&ti=QUANTITY+OF+PESTICIDES+USED+AND+THE+BASIC+AREA+TREATED+IN+AGRICULTUR AL+HOLDINGS+BY+ACTIVE+SUBSTANCE+AND+CROP&path=../I_Databas/Environment/01Agri_environmental_indica tors/&lang=1

3 4 5 6 7 8 ²Data source:

https://statdb.luke.fi/PXWeb/pxweb/en/LUKE/LUKE 02%20Maatalous 04%20Tuotanto 34%20Kasvinsuojeluaineiden%2 0kaytto%20maataloudessa/02 Kasvinsuojeluainekaytto,px/table/tableViewLayout2/?rxid=fe4b917d-bf2e-4897-b62dd6b93e4d69ef

9 ³Data source: http://www.pcs.agriculture.gov.ie/sud/pesticidestatistics/

10 ⁴Data source: https://papa.julius-kuehn.de/index.php?menuid=33

11 ⁵Data source: https://www.mapa.gob.es/es/estadistica/temas/estadisticas-agrarias/agricultura/estadisticas-medios-12

produccion/fitosanitarios.aspx

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Table 4(on next page)

Breakdown of cultivated areas of countries used

Country and cultivated (10³ ha) areas, together with the fraction of cultivated area (CA) designated to the distinct crop types. Data source: EUROSTAT TAG00025 and org_cropar databases.



Country	Country area (10³ ha)	National cultivated area (10³ ha)	Organic crops (% CA)*	Arable (% CA)	Permanent grasslands (% CA)	Permanent crops (% CA)	Kitchen crops (% CA)
Estonia	4,522.70	993.60	15.68	67.02	31.69	0.33	0.97
Finland	33,844.00	2,271.90	13.09	98.71	1.06	0.15	0.06
Germany	35,737.60	16,687.30	6.82	70.30	28.25	1.19	0.01
Ireland	6,979.70	4,489.21	1.66	10.02	90.53	0.04	0.00
Spain	50,594.40	23,494.57	6.85	52.40	27.20	19.93	0.47

^{*} This value includes agricultural areas that are fully converted and under conversion to organic farming. This value excludes kitchen gardens.



Table 5(on next page)

Basic area treated with top five active ingredients

National basic areas (ha) treated with the top five most widely used active ingredients in Estonia, Germany, Ireland and Spain, together with the proportion these represent of the total national non-organic cultivated area (CA) and country areas. Data sources: public accessible data of national plant protection products use, and EUROSTAT TAG00025 and org_cropar databases.



Country	PPP group	Active Ingredient	Basic area (ha)	% non-organic CA	% country area
	Fungicides Tebuconazole		114,779.02	13.70	2.54
	Herbicides	Glyphosate	83,335.97	9.95	1.84
Estonia	Herbicides	Florasulam	79,537.10	9.49	1.76
Estonia	Fungicides	Prothioconazole	73,079.43	8.72	1.62
	Herbicides	Iodosulfuron- methyl-sodium	70,017.82	8.36	1.55
	Fungicides	Prothioconazole	3,383,028.00	21.76	9.47
	Fungicides	Tebuconazole	3,251,387.00	20.91	9.10
Commons	Plant Growth	Trinexapac	2 022 200 00	19.51	8.49
Germany	Regulators		3,033,290.00		8.49
	Fungicides	Epoxiconazole	2,963,429.00	19.06	8.29
	Herbicides	Flufenacet	2,585,973.00	16.63	7.24
Herbicides		Fluroxypyr	271,137.11	6.14	3.88
	Fungicides	Chlorothalonil	239,146.51	5.42	3.43
Ireland	Fungicides	Prothioconazole	219,424.13	5.12	3.14
Heland	Herbicides	Glyphosate	218,858.60	4.92	3.14
	Plant Growth Regulators	Chlormequat	163,052.85	3.79	2.34
	Herbicides	Glyphosate	2,588,693.10	11.83	5.12
Spain	Herbicides	2,4-D acid	1,550,798.90	7.06	3.07
	Herbicides	Tribenuron-methyl	1,419,758.40	6.49	2.81
_	Fungicides	Copper oxychloride	1,287,566.80	5.88	2.54
	Insecticides	Dimethoate	826,432.00	3.78	1.63



Table 6(on next page)

Risks associated with most widely used active ingredients



Risks associated (L-Low, M-Moderate, H-High) with the contact or oral exposure of non-target organisms to the most widely used active ingredients (Als) among the analysed countries. For more detailed information, see Table S2 of supplementary material. Blank cells mean no data available.



Country	AI	% non- organic CA	% country area	Mammals	Birds	Fish	Aquatic invertebrates	Crustaceans	Apis spp. Contact	Apis spp. Oral	Bombus spp. Contact	Bombus spp. Oral	Osmia spp. Contact	Osmia spp. Oral	Earthworms
Estonia	Tebuconazole	13.7	2.54	M	M	M	M	M	L	M	-	-	-	-	L
Estonia	Glyphosate	9.95	1.84	L	L	M	M	M	L	M	-	-	-	-	L
Estonia	Florasulam	9.49	1.76	L	M	L	L	-	L	L	-	-	-	-	L
Estonia	Prothioconazole	8.72	1.62	L	L	M	M	L	-	L	M	L	-	-	L
Estonia	Iodosulfuron- methyl-sodium	8.36	1.55	L	L	L	L	-	L	M	L	-	-	-	L
Germany	Prothioconazole	21.76	9.47	L	L	M	M	L	-	L	M	L	-	-	L
Germany	Tebuconazole	20.91	9.1	M	M	M	M	M	L	M	-	-	-	-	L
Germany	Trinexapac	19.51	8.49	L	L	M	L	M	L	L	-	-	-	-	M
Germany	Epoxiconazole	19.06	8.29	L	L	M	M	-	L	M	L	L	-	-	M
Germany	Flufenacet	16.63	7.24	M	M	M	M	M	L	L	L	-	-	-	M
Ireland	Fluroxypyr	6.14	3.88	L	L	M	L	-	L	M	-	-	-	-	M
Ireland	Chlorothalonil	5.42	3.43	L	L	Н	Н	-	M	M	L	M	-	-	M
Ireland	Prothioconazole	5.12	3.14	L	L	M	M	L	-	L	M	L	-	-	L
Ireland	Glyphosate	4.92	3.14	L	L	M	M	M	L	M	-	-	-	-	L
Ireland	Chlormequat	3.79	2.34	M	M	L	M	-	M	M	-	-	-	-	M
Spain	Glyphosate	11.83	5.12	L	L	M	M	M	L	M	-	-	-	-	L
Spain	2,4-D acid	7.06	3.07	M	M	M	L	-	L	M	-	-	-	-	M
Spain	Tribenuron- methyl	6.49	2.81	L	L	L	L	-	-	M	-	-	-	-	L
Spain	Copper oxychloride	5.88	2.54	M	M	M	M	-	M	M	-	-	-	-	M
Spain	Dimethoate	3.78	1.63	M	Н	M	M	M	Н	Н	Н	Н	M	Н	M