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<td>Authors(s)</td>
<td>Zhao, Y.Q.; Singleton, P.; Meredith, S.; et al.</td>
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<tr>
<td>Publication date</td>
<td>2013-02</td>
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<tr>
<td>Publication information</td>
<td>International Journal of Environmental Studies, 70 (1): 59-72</td>
</tr>
<tr>
<td>Publisher</td>
<td>Informa UK (Taylor &amp; Francis)</td>
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<td>Item record/more information</td>
<td><a href="http://hdl.handle.net/10197/4864">http://hdl.handle.net/10197/4864</a></td>
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<td>Publisher's statement</td>
<td>This is an electronic version of an article published in International Journal of Environmental Studies (2013) 70(1): 59-72. International Journal of Environmental Studies is available online at: <a href="http://www.tandfonline.com">www.tandfonline.com</a>. DOI: <a href="http://dx.doi.org/10.1080/00207233.2012.752557">http://dx.doi.org/10.1080/00207233.2012.752557</a>. 10.1080/00207233.2012.752557</td>
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<td>Publisher's version (DOI)</td>
<td>10.1080/00207233.2012.752557</td>
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Current status of pesticides application and their residue in the water environment in Ireland

Y. Q. Zhao1*, P. Singleton1, S. Meredith1 and G.W. Rennick2

1 Centre for Water Resources Research, School of Civil, Structural and Environmental Engineering, University College Dublin, Newstead, Belfield, Dublin 4, Ireland
2 Pesticide Registration & Control Division, Department of Agriculture, Food and the Marine, Backweston Campus, Young's Cross, Celbridge, Co. Kildare, Ireland

*Corresponding author: E-mail: yaqian.zhao@ucd.ie

Abstract

Pesticides have been listed by the Irish EPA (Environmental Protection Agency) as potentially dangerous pollutants that may pose a significant risk to the water environment in the Republic of Ireland (ROI). Although this analysis of pesticides data was based on the existing pesticides application survey in ROI, this study aims to produce a GIS profile for the amount of pesticides used in agriculture and the distribution of their use in different parts of the country. The study identifies and reports on the six most widely used pesticides in ROI, they are MCPA, Glyphosate, Chlorothalonil, Mecoprop-P, Chlormequat and Mancozeb. More significantly, the study discusses the application of pesticides and the potential impact on the Irish water environment by examining the status of pesticide residue in the Irish water environment. Finally, the study surveys possible strategies for the removal of pesticides residues, with attention to some of the studies done worldwide.

Keywords: Pesticide, water environment, residue, Ireland

1. Introduction

The arable/horticulture industry in Republic of Ireland (ROI) is currently worth close to €1 billion per year and the total agrochemicals market in 2008 was valued at €48.5 million [1]. Crop production in ROI relies heavily on the use of Plant Protection Products (PPPs) to guarantee and maximise yields. This reliance on chemical treatment results in over 2 million kg of synthetic pesticides being applied to crops (both forage and arable) each year [2, 3]. Pesticide is a term used to describe both biocides (wood preservatives, rodenticides, human and veterinary disinfectants etc.) and PPPs. Pesticides are chemical substances or biological agents that are used to repel or destroy insects, plant pathogens, weeds, microbes or other organisms injurious to plants. This broad heading can be split into a number of categories including organic insecticides, herbicides, fungicides, nematocides, acaricides, algicides, rodenticides and slimicides. Other categories include plant growth regulators and seed treatments, both used in commercial crop production [4, 5].

The government body in ROI responsible for the regulatory system for PPPS is the Pesticide Registration & Control Division of The Department of Agriculture, Food and the Marine (DAFM). Water quality monitoring in Ireland is the responsibility of the Environmental Protection Agency (EPA). Although pesticides are a vital part of the agricultural systems, they also pose a threat to human and animal health via their off-site migration and detrimental effects on surface water and groundwater quality [5-7].
introduction of new ingredients in pesticides manufacture could result in diffuse inputs of new pollutants into the environment, for which the environmental fate is still poorly understood [8]. Walker et al have outlined the potential hazards and damage caused by pesticides [9]. Although the Irish EPA statistics on pesticide reported that in 2010 only 5 samples out of 1335 (<0.3%) [10] exceeded the drinking water limit of 0.1 µg/l, some balance must be struck between high yields, quality food production and safe use of pesticide. Controls and monitoring, though important, cannot ensure that all pesticides will be used correctly and safely. There must be systems in place to deal with toxic aspects of pesticides if they are found in the water environment. There should also be research into reliable, cost-effective removal techniques.

Based on an existing database of the national pesticides application survey in ROI, this study provides an entire GIS geographic distribution profile with reasonably accurate facts and figures for the ROI of the types and quantities of the pesticides used. Relevant information on the pesticide pathways for entry to the Irish aqueous environment is discussed before presenting the main findings. The paper also presents possible strategies for the removal of pesticide residues, by means of relevant data from published literature.

2. Status of pesticide application in ROI
2.1 Data collection, computation, field visit and GIS generation
The main aim of this study was to produce a GIS (Geographical Information System) survey of the current status of pesticide use in ROI. The first step was to source the required information. The DAFM in Ireland produces usage surveys with a database. Fortunately, the usage data for forage crops and grassland areas [2] as well as for arable crops [3] were, respectively, obtained from the DAFM. These data represent the first comprehensive national surveys of pesticide use associated with plant protection products in ROI. Further surveys are currently planned and the data will be made available in due course. The DAFM database provides details of the quantities of active substances/pesticides used; the reasons for their application; regions (but not the precise counties) where used; and the most widely used active substances. Many of these data were of little relevance to the aim of this study, but the total amounts of each active substance formed the basis of the study. The database gives the amounts of each active substance or active substance combination applied to each crop. It does not give the amount of the crops produced in the region.

This was not sufficient to create a GIS profile of pesticide use as the geographical data were absent. The Central Statistics Office (CSO) in ROI was an invaluable resource. Although CSO does not collect statistics on use of pesticides, it provided the data on the amount of each crop grown in each county. These figures for amounts provided a means to relate the amounts of pesticide used to specific counties. The DAFM figures for pesticides used on each crop type could thus be related to their county of use.

Using ‘Microsoft Excel’, it was relatively easy to compute kilograms of active substance per county based on the amount of crops grown in each. For example, say that there was ‘x kilograms’ of ‘pesticide A’ used on a crop in a year. Divide this figure by the total area of crop grown and multiply by the amount of spring barley in each county to give figures for kilograms of ‘pesticide A’ used on a specific crop in that county. This can be repeated for each crop that required ‘pesticide A’ and added together to give total values on a county by county basis. A number of site visits made it possible to form a general idea of the pesticide application in ROI. Field trips included a trip to a tillage farm at Ballytore, Athy, County Kildare where it could be seen, first hand, where pesticides are used. Some photographs of crops and chemical containers were taken. A predominantly grassland farm was also visited.
at Mountmellick, Count Laois. There was an opportunity to see an example of water courses in close proximity to agricultural land and the various drainage systems in use.

GIS profiles of the data gathered were created geographically using ArcGIS software. To give an accurate representation of the amounts of pesticide used in each county, the units used are kilograms per square kilometre.

2.2 Total pesticide usage

To date two pesticides usage surveys have been conducted and published by the Pesticide Registration & Control Division of DAFM [2, 3]. The first was on grassland and forage crops and the second on arable crops. These surveys were carried out to quantify the amounts and types of pesticides being used on different crops. The data generated provide critical information for use in assessing the impact of pesticide use on the environment and will be of great benefit in ensuring the sustainable development of the sector. Computed data show that Irish agriculture uses 2,089,287 kilograms of active substances each year across the country. These pesticides are applied to over 4.4 million hectares of agricultural land to a wide variety of crops. Figure 1 shows the percentage value of each PPP category in the context of the overall amount spent on pesticides in ROI. It shows that fungicides account for 60% of the total amount and herbicides account for a further 26%. In addition, according to the current PPPs marketing data, 877 PPPs have been registered for sale in the Irish market [11].

Figure 1 shows a geographical profile of the total quantity of pesticides used in each county in ROI. The GIS profile indicates that County Louth is the highest consumer of pesticides in ROI with an average active substance application rate of 146.7 kg/km². One reason is that due to an abundance of good farm land, the growth of crops needs a relatively high amount of treatment with PPPs. County Dublin is the second highest user of pesticides in ROI. Despite Dublin’s high levels of urbanisation, there is a large amount of high quality farmland in County Dublin area as a whole, resulting in high pesticide usage. In addition to this, North county Dublin is known for its very concentrated vegetable producing areas, crops which tend to require higher PPP input. In contrast, County Mayo is the lowest contributor with an average application rate of just 3.7 kg/km² probably due to its peat land nature. It is clear that the main areas of pesticide use are along the east coast of the country. This is due to better quality land, the growth of crops dependent on greater PPP usage and greater demand for food products in densely populated regions.
2.3. Top six widely used pesticides

Computed data show that MCPA, Glyphosate, Chlorothalonil, Mecoprop-P, Chlormequat and Mancozeb are the six most widely used pesticides in agriculture in ROI. Information on each of these is given by a GIS profile with geographical distribution in each county, as shown in figure 3. The GIS profiles provided a clear and easy way to understand how much of these six pesticides are used in ROI.
Figure 3 GIS profile of the top six most widely used pesticides of ROI
Figure 3 GIS profile of the top six most widely used pesticides of ROI (continued)
MCPA is the no. 1 most widely used pesticide in ROI. There appears to be a gradual change in use from the largest user in County Monaghan (3.9 kg/km²) to the smallest user in County Dublin (1.4 kg/km²). But the change does not seem to be significant, showing that MCPA is a general, widely used pesticide. Glyphosate is one of the six most widely used and common pesticides in the country. It is used extensively on both arable and grassland and fodder crops, showing high amounts of use per kilometre squared of intensive farming areas in County Louth (6.0 kg/km²). The farther west the county, the smaller the amount used (0.87 kg/km² in County Mayo). This reflects the poorer land found in the west of ROI as compared to the east. Chlorothalonil is a fungicide mainly used to treat diseases associated with cereal, fruit and vegetable growth. Spring barley and Winter wheat are the best examples of crops which require the use of Chlorothalonil. Figure 3, of amounts used per county, shows that County Wexford (8.1 kg/km²), County Louth (7.8 kg/km²) and County Carlow (6.4 kg/km²) are the highest users. As with other pesticides, there is low use in areas of poor land, such as Mayo (0.1 kg/km²). Mecoprop-P is mainly used for grass treatment as well as weed control in cereal crops. County Wexford is the largest user of Mecoprop-P (9.5 kg/km²) by a wide margin and as can be seen from the geographical representation of the data in figure 3, its use is heavily concentrated in the South-East, which is the ROI’s primary spring barley growing region.

Figure 3 also shows the usage of Chlormequat in ROI by county. There are only a small number of counties (highest user in County Louth of 5.7 kg/km²) that have high usage values. Figure 3 shows that they are all on the east coast of the country. This once again highlights the fact that better land is on Ireland’s east coast where higher yields can be expected, with the help of PPPs. Mancozeb is mainly used, in ROI, for treating fungal diseases that affect potatoes. It can be seen from figure 3 that the use of Mancozeb shows a very large difference in usage patterns throughout the country. Potatoes are highly susceptible to fungal diseases, and so if the crop is grown, large amounts of pesticide are often required. County Dublin (24.2 kg/km²), County Louth (22.3 kg/km²) and County Meath (14.9 kg/km²) have very high levels of Mancozeb use with usage levels dropping rapidly outside the Leinster region. Mancozeb’s specific use is shown by the largest range of usage figures compared to the other pesticides.

3. Pesticide residue in water environment in ROI
3.1 Entry routes
Large areas of land in ROI are sprayed with PPPs to control various pests. The problems for water can arise from spray drift to areas outside the target site, movement through soil into groundwater and flow into drains and watercourses after heavy rain. Thus, a major pathway for pesticide transport to surface water and groundwater is leaching from soils following application [12, 13]. Figure 4 shows a stream adjacent to agricultural land on a farm near Mountmellick, County Laois, during a recent visit. It demonstrates the proximity of rivers to land which could potentially be receiving pesticide leaching. In ROI, the majority of drinking water originates from surface water (81.9%) and the remainder originates from groundwater (10.3%) and springs (7.8%) [10]. In most cases, there is no drain, but the water with pesticide content infiltrates the soil and passes into the stream. Therefore, surface water quality is a main concern in relation to its suitability for use as a source for drinking water supply, in food processing and related industrial operations and in the bottled water industry.
Figure 4 An example of land drains entering a water course on a farm near Mountmellick, County Laois.

Figure 5 shows a model by which pesticides enter waters via diffuse and point pathways. Diffuse pathways comprise the natural fissures of the sub-surface, artificial drainage systems, surface or sub-surface, leaching or by-pass flow. Point entry pathways are direct. The contaminating effect may come from tank filling, spillages, faulty equipment, washing of equipment, and improper cleaning and disposal of pesticide containers. Application to crops may lead to contamination by diffuse pathways. It may also lead to contamination by point pathways. In either case, the problem of pesticides is that they go beyond the target crops.

3.2 Legislation
Pesticides enhance agricultural productivity, but the possible environmental and health side effects justify very stringent government regulation. Very few pesticides in use are known to be mutagenic, carcinogenic or teratogenic [14]. Nevertheless, there is no room for complacency; the environmental fate of any pesticide must cause concern when it is poorly
understood. [8]. The correct amount of pesticides must be chosen to destroy the target pests while leaving the affected crop intact. In ROI, pesticide use is governed mainly by EU regulations and the DAFM monitors usage throughout the country, as well as monitoring sales and marketing.

3.2.1 EU pesticides directive: It has been estimated that recent amendments to the European regulations on pesticide registration could decrease the number of active ingredients available by up to 15% [8]. Thus, the aim is to have in use only those products which have minimal adverse impact for maximum effect. The current EU Regulation No. 1107/2009 [15] states that Member States may only authorise the use of products containing active substances once these active substances have been first approved. This regulation states that PPPs should be applied safely, only where absolutely necessary and in the least possible amounts. In the EU, the evaluation of active substances is divided into two categories, hazard and risk. In addition, Directive 98/8/EC [16] on the placing of biocidal products on the market and Directive 1999/45/EC [17] on the classification, packaging and labelling of dangerous preparations have been implemented by DAFM in ROI.

Directive 98/8/EC [16] is given effect in Irish law by Statutory Instrument S.I. No. 625 of 2001[18]. In addition, the new EU Sustainable Use Directive 2009/128/EC [19] concerns significant reductions in risks associated with pesticide use in the short to medium term. Decreased use in certain areas will also have to be implemented, for example, outside specific buffer zones of major waterways. Active substances are not authorised for marketing and use unless it has been shown that there will be no harmful effects on human health and no unacceptable impact on the environment. The implementation of the new regulations in ROI requires significant changes in how PPPs are stored and distributed.

3.2.2 EU drinking water directive: The legislation that governs the permitted amount of pesticides allowed in water intended for human consumption is the Drinking Water Directive 98/83/EC [20]. The main points include setting quality standards for water at the tap, requiring Member States to check their own drinking water standards regularly and provide consumers with regular and accurate information on drinking water quality. The directive states that the quantity of pesticides present cannot exceed 0.10 µg/l. These are defined as including organic insecticides, herbicides, fungicides, nematocides, acaricides, algicides, rodenticides and slimicides. Certain active substances have been given specific parametric values including aldrin, dieldrin, heptachlor and heptachlor epoxide, permitted levels for which have been reduced to 0.030 µg/l. Where more than one active substance is monitored, the parametric value for the sum of all individual pesticides detected and quantified in the monitoring procedure is 0.50 µg/l.

3.3 Pesticides in water samples

Individual pesticides have long been detected in drinking water throughout ROI. According to the Irish EPA’s report on drinking water quality, in recent years, the number of samples exceeding the 0.1µg/L value is very low [10]. Table 1 summarises the overall results of the total pesticides in Irish drinking water samples. This suggests that a relatively small number of pesticides are being detected and the trend seems to be going down. This reflects the efforts made by DAFM, EPA, county councils, water treatment plants etc. Undoubtedly, new EU legislation 2009/128/EC [19] is playing an important role in ensuring safe distribution and use of pesticides. [21]. Because some pesticides are toxic and drinking water must be safe, pesticide monitoring, control and removal technology should deserve high attention.

The Irish EPA published a Dangerous Substances Regulations National Implementation Report 2005 [6]. The EPA tested a large number of rivers, lakes and tidal waters for the
presence of a wide range of dangerous substances, Atrazine being one of these. According to the EPA report, out of 299 test sites, 12 exceeded the drinking water limit [6].

### Table 1 Pesticides monitoring in Irish drinking water samples

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4. Removal and remediation

To regulate pesticides by restricting their use reduces the pesticide residues in the water environment and reduces the risk of the environmental impact. Removal/treatment technologies are also necessary to secure the water environment from pesticide application. Studies have shown that conventional drinking water treatment processes are not effective in removing certain types of pesticides [25, 26]. The chlorination process in disinfection of drinking water guarantees the removal of many of the pesticides that might be present in raw waters. Despite this, the recent use of a wide variety of pesticides in agriculture means that it is difficult to adopt a simple method for the removal of pesticides since the process of separation depends on the chemical nature of the pesticide itself. Some of the methods currently being employed throughout Europe include preoxidation by chlorine, preoxidation by ozone, coagulation with aluminium sulphate, activation carbon filtration, nanofiltration and combinations of these techniques [4,5,7,25-29].

Thuy et al., [29] investigated the extent to which pesticides are removed from water by coagulation-flocculation. These researchers used aluminium sulphate as coagulant to remove four pesticides (Aldrin, Dieldrin, Atrazine and Bentazon) during jar test experiments in surface water from a Belgian river. The study showed that the removal rate is always below 50%. Testing whether nanofiltration can remove pesticides from drinking water shows that this process has a good capacity to remove some pesticides from water and the membrane material used in the filtration process greatly influences the percentage of pesticides removed [25, 27]. Ormad et al Conducted a comprehensive study aimed to explore the effectiveness of the treatments commonly used in drinking water plants in Spain to degrade 44 pesticides systematically detected in the Ebro River Basin [6]. These researchers used preoxidation by chlorine or ozone, chemical precipitation with aluminium sulphate and activated carbon adsorption. The results showed that the oxidation by chlorine removes 60% of the studied pesticides, although combining oxidation with a coagulation-flocculation-decantation process is more effective. Oxidation by ozone removes 70% of the studied pesticides. Although combination with a subsequent coagulation-flocculation-decantation process does not improve the efficiency of the process, combination with an activated-carbon absorption process gives rise to 90% removal of the studied pesticides. In a recent review study, Stuart [5] provides a summary on efforts to remove pesticides in drinking water treatment and wastewater treatment processes.

Regarding pesticides removal in water environment in ROI, O’Dwyer et al. [30] study the advanced oxidation process for pesticide removal in water and wastewater. Three TiO2-containing composites have been developed and trials have been conducted for MCPA and 2,
4-D removal in the presence of UV light. Results show that the pesticides can be successfully removed mainly by the integrated adsorption and the enhanced superior photocatalyst. Hu et al. [31] have studied pesticide removal by using water treatment residual (WTR) as a low cost-effective approach. WTR refers to the by-product generated inevitably from the production of drinking water in water treatment plants. Al-WTR represents the residuals when aluminium sulphate is used as coagulant. Generally, in Ireland and most places worldwide, the Al-WTR is regarded as a waste and consequently landfilled. Al-WTR is predominantly composed of Al of 29.7±13.3% dry weight [32]. Al holds great promise of adsorption affinity with phosphorus; which is the basic monomer of most pesticides. Hu et al. [31] have advocated the reuse of Al-WTR as pesticide pre-treatment to prevent their entry to water bodies in various runoffs.

5. Discussion

Although pesticides are a vital part of agricultural systems, they also pose a threat to human and animal health if used incorrectly. Some would say that no pesticide is completely safe. Therefore, some balance must be struck between high yields, quality food production and safe use. There are a large number of different types of pesticides used in ROI and the control of marketing and use is a major issue. DAFM’s annual control programme includes nearly 2,000 inspections of retail, wholesale and end user premises. The GIS profiles are useful in giving a clear and easy way on how the pesticides are used in Ireland (Figs. 2 and 3). It is clear that the main areas of concern are along the east coast of the country (Fig. 2). This is due to better quality land and the more extensive cultivation of crops as well as greater demand for food products in densely populated regions. The high populations of the Greater Dublin Area, including counties Kildare, Meath and Louth, mean that a major pesticide incident could cause potential danger. In Dublin, water resources are already stretched and the Local Authorities could not afford a serious contamination by pesticides of drinking water supply.

It is worth noting that ROI only has two national pesticides usage surveys so far, and will establish an annual survey; but Northern Ireland has 11 (by 2010) pesticides usage surveys on arable crops and 6 surveys (by 2009) on grassland and fodder crops [33, 34]. These surveys provide information for consideration by the government, but this can be used by those engaged in residue testing. The surveys are also useful for evaluating the impact of policy and trends in pesticide usage. The 11th survey (in 2010 for arable crops) of pesticide usage in Northern Ireland showed that the total weight of pesticides applied to arable crops in 2010 decreased to 136 tonnes of active ingredients, a reduction of 19% compared with 2008 and 43% compared with 2006 [33]. The 6th survey (in 2009 for grassland and fodder crops) examining pesticide usage in Northern Ireland, indicated that the area of grassland and fodder crops receiving pesticide treatment, decreased by 21% when compared to that recorded in 2005. A total of 75 tonnes of pesticides was applied to 101,998 spray hectares of grassland and fodder crops during 2009 [34]. Although accurate estimation of pesticides usage is difficult, there is a global trend that the usage of pesticides is decreased by time.

Direct comparison of pesticide usage between countries is difficult, but useful. Currently, more than 300,000 tonnes of pesticides are used in agriculture each year in China [35] while more than 100,000 tons of pesticides are imported into Thailand, ranked fourth out of 15 Asian countries in annual pesticide use [36]. A study in Palestine has identified 217 pesticides including 13 soil sterilizers, while 134 kinds with different active ingredients (insecticides 62; fungicides 45; herbicides 20) were applied and the quantity was 254 tonnes in 2006 [37].
Pesticide residues have long been detected in surface water and groundwater, with many cases reported worldwide including ROI (see Table 1). Pandey et al. [38] reported that although there are currently 179 pesticides registered for use in India, their use has caused both surface sediment and river water pollution as several pesticides have been detected in the river Yamuna in Delhi. Similar studies also reported the pesticides detection in other rivers in India. In addition, Pandey et al. [38] also reported some cases of pesticides monitoring in other places in the world, such as: (1) coastal marine sediment in Singapore; (2) Ebro river delta, Mediterranean Sea; (3) Paranoa lake in Brazil; (4) Coastal lagoon watershed in Argentina; (5) Bay of Ohuira in Mexico; (6) Haleji lake in Pakistan; (7) some stream sediment in Spain; (8) Lake Orta sediments in Italy; (9) Uluabat lake in Turkey and (10) Pearl river estuary in China etc. Pesticides are in detectable level in the UK groundwater [5] while, in the US, it has been reported that 100% of major rivers and streams and 33% of major aquifers contained at least one pesticide at detectable levels [39].

Although quantity control and residues monitoring are important, these cannot ensure that all pesticides will be used correctly and safely. There must also be systems in place to deal with toxic chemicals if they are found in drinking water although there seems to be no health risks in ROI arising from pesticide misuse. No doubt, strict control measures by bodies such as the DAFM, regular testing by county councils on behalf of the EPA and new EU legislation banning hazardous pesticides have prevented this. It does not appear, however, that ROI has used any remediation measures other than normal water treatment procedures, such as coagulation-flocculation and disinfection.

With regard to recommendations for the future, some investment is required in training farmers on correct application methods for pesticides. Otherwise, potential dangers to drinking water can be ignored. It may be appropriate to sell pesticides only to those who can produce written evidence of having received the necessary safety training. DAFM is at present introducing training requirements for all professional end users such as farmers. In addition, an existing risk assessment established by DAFM should further be enhanced by which the pesticides entering groundwater, their toxicity and potential risks to drinking water and the environment can be assessed. Zhao and Pei [40] have reviewed the four major methodologies of such risk evaluation.

In relation to drinking water quality assurance, there should be an increase in the sampling rates of water supplies, especially during times of maximum pesticide application. Even the EPA has suggested that the point sampling of water supplies, which provides only a snapshot of the quality of the water on the day it is taken, is an inadequate method of ensuring that drinking water meets the necessary standards. To ensure the water is adequately clean, the water supplier must have in place a management system that identifies all potential risks and includes reduction measures to manage these. There is no specific treatment process being employed at present to remove pesticide in Ireland. The reason might be that it appears that pesticides are not considered to be a significant risk. The use of chlorine is widespread in ROI and this disinfection reaction has an average pesticide removal rate of 60% [7]. Nevertheless, it seems highly desirable to conduct research and development of practical technologies for pesticides control/removal.

ROI is at a critical point in its water reform strategy, in a difficult economic climate. Despite this, the Irish government decided to establish Irish Water, a state body. This will become responsible for the delivery of water services currently managed by local authorities. This significant reform in the Irish water sector aims to develop one single water utility company and to invest in infrastructure.

6. Conclusions
There are 877 different types of pesticides used in ROI and the control of their use is a major issue in terms of the water environment, especially the safety of drinking water. In total, about 2 million kilograms of active substances are used each year in ROI. The GIS profiles indicate that County Louth is the greatest contributor of pesticides in ROI with an average pesticides application rate of 146.7 kg/km². The main areas of pesticides usage are along the east coast of the country. The top six pesticides used in ROI are MCPA, Glyphosate, Chlorothalonil, Mecoprop-P, Chlormequat and Mancozeb. Pesticides become a potential pollutant in water environment following agricultural usage or accidental spills. The rates at which pesticides enter water courses vary according to soil properties, prevailing weather conditions, aspect and slope of the area of application and the chemical makeup of the individual pesticide. As a number of cases of pesticide contamination have been monitored in drinking water samples across the country, Ireland should reflect further on the actions which should be considered necessary to reduce the risks associated with PPP application. These may include training of pesticide operators in proper use, adequate pesticide monitoring, and development of practical pesticide removal technology. Application of water treatment residual as a low cost adsorbent for pesticide immobilization may be a promising and cost-effective approach in Ireland.

Acknowledgement
The authors acknowledge the valuable review comments and suggestions from the anonymous reviewers and the editor to improve the quality of the paper.

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