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Exposure assessment using human biomonitoring for glyphosate and fluroxypyr users in amenity horticulture

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ABSTRACT

Background: Pesticides and their potential adverse health effects are of great concern and there is a dearth of knowledge regarding occupational exposure to pesticides among amenity horticulturalists.
Objective: This study aims to measure occupational exposures to amenity horticuturalists using pesticides containing the active ingredients, glyphosate and fluroxypyr by urinary biomonitoring.
Methods: A total of 40 work tasks involving glyphosate and fluroxypyr were surveyed over the period of June – October 2015. Workers used a variety of pesticide application methods; manual knapsack sprayers, controlled droplet applicators, pressurised lance applicators and boom sprayers. Pesticide concentrations were measured in urine samples collected pre and post work tasks using liquid chromatography tandem mass spectrometry (LC–MS/MS). Differences in pesticide urinary concentrations pre and post work task, and across applications methods were analysed using paired *t*-tests and linear regression.
Results: Pesticide urinary concentrations were higher than those reported for environmental exposures and comparable to those reported in some agricultural studies. Log-transformed pesticide concentrations were statistically significantly higher in post-work samples compared to those in pre-work samples (paired *t*-test, $p<0.001$; for both $\mu g\,L^{-1}$ and μmol/mol creatinine). Urinary pesticide concentrations in post-work samples had a geometric mean (geometric standard deviation) of 0.66 (1.11) $\mu g\,L^{-1}$ for glyphosate and 0.29 (1.69) $\mu g\,L^{-1}$ for fluroxypyr. Linear regression revealed a statistically significant positive association to exist between the time-interval between samples and the log-transformed adjusted (i.e. post- minus pre-task) pesticide urinary concentrations ($\beta=0.0039$; $p<0.0001$).
Conclusion: Amenity horticulturists can be exposed to pesticides during tasks involving these products. Further research is required to evaluate routes of exposure among this occupational group.
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1. Introduction

The use of pesticides and their possible health effects is of significant concern, especially with an estimated annual usage of 12.5 million tonnes of pesticides worldwide (Michael and Alavanja, 2009). Occupational exposures to pesticides have regularly been associated with adverse health effects such as cancer, respiratory diseases, detrimental reproductive health, and neurological diseases (Acquavella et al., 2003; Arbuckle et al., 2001; Bretveld et al., 2008; Guyton et al., 2014; Henneberger et al., 2014; Hernandez and Menendez, 2016; Lauria et al., 2006; Montano, 2014; Mostafalou and Abdollahi, 2013, 2016; Weidner et al., 1998; Ye et al., 2013).

Pesticide products must be approved before use and, dependent on their approval, they can be used in a variety of settings. However, occupational exposure assessment studies have largely been confined to agriculture for example, farm workers (Baharuddin et al., 2011; Blanco et al., 2005; Krenz et al., 2015; Lebailly et al., 2009; MacFarlane et al., 2008; Rubino et al., 2012; Singleton et al., 2015; Strong et al., 2008) and their families or residents near farmland (Curwin et al., 2007; Galea et al., 2015; Hanchenlaksh et al., 2011; Sams et al., 2016).

There is a comparably less research on occupational exposures to pesticides in horticulture; in greenhouses (Bouvier et al., 2006; Flores et al., 2011; Machera et al., 2003; Machera et al., 2002;

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Nuyttens et al., 2009; Ramos et al., 2010), florists (Bouvier et al., 2006) and amenity horticultural workers (Johnson et al., 2005). This sector contributes approximately €90 billion to the European economy (EPSO, 2014). The horticulture industry is diverse and can be divided into commercial (production of food and flowers for commercial gain) and amenity (for recreational or ornamental purposes) horticulture (The Chartered Institute of Horticulture, 2016).

Pesticide products are used to control weeds and scrub growth in amenity areas and for the preservation and growth of decorative plants or endangered species in ornamental horticulture (Illing, 1997). However, the use of pesticides in horticulture is different to agriculture, so results from agricultural based studies are not always applicable to the horticultural sector. Horticulturists regularly use a variety of pesticides for a range of applications from protecting indoor and outdoor plants from disease or insect infestations, to control nuisance and invasive weed growth in public areas. Horticultural workers tend to use a great variety of pesticides in a wider variety of applications throughout the year.

These differences in industrial use of pesticides can include the dose rate of pesticide products used, the type of work tasks and the application frequency. Use of pesticides is not always confined to the growing season and horticultural workers usually use a variety of application methods including boom sprayers, manual knapsack sprayers as well as battery operated sprayers on a more regular basis. In horticulture, a work task involving pesticide use typically involves: the collection of the product from the chemical store, mixing of the product to the correct dose rate, loading the applicator (mixing and loading), spraying the pesticide and finally cleaning the application equipment after use (MacFarlane et al., 2013).

Previous studies on horticultural workers spraying herbicides in residential and public areas, or on spraying pesticides in greenhouses suggest that dermal exposure is the prominent exposure route, accounting for up to 99% of the total body exposure (Aprea et al., 2004; Flack et al., 2008; Tuomainen et al., 2002; Vitali et al., 2009). Potential dermal exposure (PDE) levels tend to vary depending on the stage of the activity (i.e. mixing & loading, spraying or cleaning stage), application method used, condition of the applicator, human factors e.g. worker's attitude towards safety (Johnson et al., 2005; Ramos et al., 2010) and use of personal protective equipment (PPE) (Lander and Hinke, 1992; MacFarlane et al., 2013; Nigg et al., 1986). Higher exposures have been associated with the mixing and loading activity (Ramos et al., 2010) especially when more than one mix and load task is completed in a day (Flores et al., 2011). Higher exposures have also been associated with the use of high pressure application methods (Machera et al., 2003) or when using a hand held lance walking forward into the pesticide mist (Nuyttens et al., 2009) and from exposure due to surface transfer of previously applied pesticide contaminants (MacFarlane et al., 2013). Although PDE data provides useful information on exposure, PDE estimates do not always reflect the total exposure risk from other routes, such as, inhalation and inadvertent ingestion.

Biological monitoring, measuring pesticides or their biomarkers in samples such as blood or urine, can be used to evaluate total uptake of a pesticide by an operator by all exposure routes and can provide a more reliable estimate of pesticide exposure where information on pharmacokinetics is available (Acquavella et al., 2003; Chester, 2010). Biological monitoring has previously been used in studies evaluating occupational exposures to pesticides in both the agricultural (Acquavella et al., 2004; Aprea, 2012; Hanchenlaksh et al., 2011; Mesnage et al., 2012; Rubino et al., 2012; Singleton et al., 2015) and horticultural sectors (Aprea et al., 2005; Baldi et al., 2006; Bouvier et al., 2006; Flores et al., 2011). Recent papers have called for more exposure assessment studies, as exposure can vary greatly between workers in apparently similar conditions. This information would provide direct evidence of exposure levels to evaluate the range of human pesticide exposure, to advance epidemiology and provide more reliable data for predicting health effects, to provide regulatory estimates of exposure and to ensure safe and sustainable pesticide use (Acquavella et al., 2003; Gangemi et al., 2016).

Fluroxypyr and glyphosate are commonly used pesticides in amenity horticulture, fluroxypyr is an herbicide used to control broadleaf weeds and woody brush, glyphosate is a non-selective weed killer. Fluroxypyr is classified as a 'not likely' human carcinogen (US EPA, 1998) and has been assigned an Acceptable Daily Intake (ADI) and an Acceptable Operator Exposure Level (AOEL) of 0.8 mg/kg body weight per day (bw/day) (EFSA, 2011). Glyphosate, is the highest volume herbicide used worldwide, contained in over 750 pesticides (Guyton et al., 2014) and was classified as 'Group 2A – probably carcinogenic to humans' (IARC, 2015). The European Food Safety Authority conducted a peer review and concluded that it was 'unlikely to pose a carcinogenic hazard to humans' (EFSA, 2015). Glyphosate has an ADI of 0.5 mg/kg bw/day and an AOEL of 0.1 mg/kg bw/day (EFSA, 2015).

A small number of studies have used biological monitoring methods to evaluate human exposures to glyphosate (Acquavella et al., 2004; Conrad et al., 2017; Curwin et al., 2007; Jayasumana et al., 2015; Krüger et al., 2014; Mesnage et al., 2012) but none among amenity horticulture workers. In addition, to the author's knowledge, thus far, there have been no human biomonitoring studies on fluroxypyr.

The aim of the current study was therefore to characterise the occupational exposures via a newly developed biomonitoring method for both glyphosate and fluroxypyr in amenity horticultural workers.

2. Materials and methods

2.1. Site description and study population

This study was conducted over the period of June – October 2015 in the Republic of Ireland, at field sites managed by Commissioners for Public Works in Ireland – referred to hereafter as the Office of Public Works (OPW). The OPW manage and maintain a diverse portfolio, which includes national monuments and heritage services ranging from ornamental gardens to the Phoenix Park located in Dublin which is 707 ha and one of the largest walled city parks in Europe (OPW, 2010).

A walk through survey was performed by the researcher at the selected OPW sites including national parks, ornamental gardens and historic monuments, to collect information on the frequency of use of pesticides containing glyphosate and fluroxypyr and spraying methods used. Four similar exposure groups (SEGs) were defined based on the pesticide and application method used (Table 1). Biological monitoring study protocols were developed following a review of previous research protocols (Galea et al., 2011; Health and Safety Authority, 2011; Health and Safety Executive (HSE) UK, 1997). Study protocols were approved by the National University of Ireland, Galway Research Ethics Committee (Ref 15/May/04).

The lead researcher presented the project aims and objectives and distributed project information leaflets at different sites and events hosted by the OPW with the aim of informing potential study participants. Following these events, amenity horticulturists that used relevant pesticides and worked with the OPW at the designated sites, were invited to participate in the study. Recruitment was done in coordination with the OPW Health and Safety Unit, however participation was voluntary. Prior to the commencement of the study, the lead researcher met with the workers and presented an overview of the sampling protocols and meth-

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