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# Human health risk assessment of lead (Pb) through the environmental-food pathway

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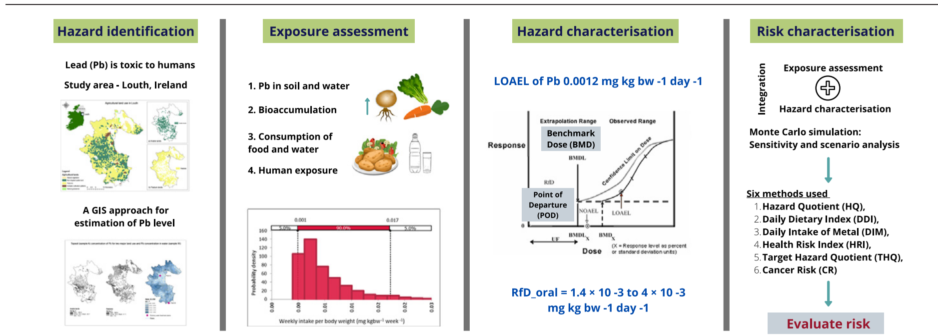
## HIGHLIGHTS

- A probabilistic farm-to-fork human health risk assessment for Pb was developed.
- Spatial and anthropogenic concentration data were collated.
- Tubers are prone to higher bioaccumulation of Pb.
- Biosolids may be restricted where elevated levels of Pb are found.
- Back-calculation suggests the permissible limit of 51 mg $kg^{-1}$ in the soil.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Drinking water and farm-to-fork pathways have been identified as the predominant environmental pathways associated with human exposure (HE) to Pb. This study integrates a GIS-based survey of metal concentrations in soil and a probabilistic quantitative risk assessment of Pb through the food chain. The case study area was selected in the east of Ireland. A step-wise exposure assessment collated the data for Pb concentration in soil and water media, bioaccumulation of Pb in unprocessed food products, such as potatoes, carrots, green vegetables, and salad vegetables. The daily mean HE to Pb through selected food products was found to be 0.073 mg $day^{-1}$, where a mean weekly exposure was estimated as 0.0065 mg kg body $weight^{-1}$ $week^{-1}$. Multiple risk estimates were used. Hazard Quotient (HQ), Daily Dietary Index (DDI), Daily Intake of Metal (DIM), Health Risk Index (HRI), Target Hazard Quotient (THQ) and Cancer Risk (CR) were found as 0.234 to 0.669, 0.002, 0.0002, 0.020 to 0.057, 0.234 to 0.669, and 0.00001, respectively which signify a low to moderate risk. A sensitivity analysis revealed that intake of potato is the most sensitive parameter of the model, which is positively correlated (coeff. + 0.66) followed by concentration of Pb in the arable soil (+0.49), bioaccumulation in tubers (+0.37), consumption of salad vegetables (+0.20), and consumption of green vegetables (+0.13) (top 5). A back-calculated limit of Pb in the soil (51 mg $kg^{-1}$) justifies the lower threshold limit of Pb (50–300 mg $kg^{-1}$) in agricultural soil set by the European Union to mitigate potential bio-transfer into food products. The study concludes there is a low to moderate risk posed by Pb, within the system boundary of the probabilistic model, and highlights the significance of limiting Pb concentrations in the vegetable producing agricultural soil.



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## 1. Introduction

### 1.1. Hazard identification: Global burden of Pb

Several metals and metalloids (elements whose properties are intermediate between metals and non-metals) (metal(loid)s) are essential for living organisms, with particular roles in cell division and metabolism while also facilitating endocrine signals between organs. Excessive concentrations of certain metal(loid)s may lead to serious health issues (Nag et al., 2022a). Lead (Pb) has been identified as a toxic metal at elevated concentrations, and extensive use of Pb is reported to result in widespread environmental contamination and health problems globally (Li et al., 2019). Being a cumulative toxicant, Pb can influence the neurologic system, kidneys, and blood circulation, especially in children, infants, and foetuses (Guo et al., 2018). Pb is distributed in the brain, liver, kidney and bones (Zwolak et al., 2019), and it. It may accumulate over time in teeth and bones, reflecting a cumulative human exposure. Pb may also affect brain and intellectual development in children, inducing apoptosis in organ tissues (FSAI, 2009; Mani et al., 2019); in some cases, irreversible neurological damage occurs. Pb was considered to be responsible for 540,000 deaths worldwide in 2016 (Li et al., 2019), and Pb exposure is estimated to account for 0.6% of the global burden of disease (expressed in disability-adjusted life years, or DALY) with the highest burden in developing regions (WHO, 2010). Recent reductions in Pb-use in petrol (gasoline), paint, plumbing, and solder are reported to have resulted in substantial reductions in Pb exposure levels (Nag et al., 2022a; WHO, 2010).

### 1.2. Sources of Pb

Pb is found at low levels in the Earth's crust, mainly as lead sulphide (PbS) (WHO, 2010). Hence, background levels of Pb may be present in soil depending on geological sources. For example, natural weathering processes releases metal(loid)s from rocks to constitute part of the soil, making nutrients more available for plants (Ling et al., 2014). However, there is also regularly anthropogenic influences. Therefore, the term 'background' may be replaced by the term 'baseline', which included the background concentration in addition to a nominal anthropogenic concentration (Nag and Cummins, 2021). Mining activity is reported as one of the most significant anthropogenic sources of heavy metal contamination and the reason for many health-related issues; for example, in the Zamfara State (Nigeria), a Pb poisoning epidemic caused over 40 children deaths in 2010 (Lo et al., 2012). Other anthropogenic activities and sources which can contribute to Pb levels in soils around industrial areas include emission from vehicles (mainly use of leaded petrol/gasoline), smelting, electrical waste dismantling, paint, glass industries, agricultural practices and waste disposal (Zwolak et al., 2019) with evidence reported in China, Sweden, France, and Germany (Guo et al., 2019). Furthermore, fixtures and plumbing work, either made of Pb or with Pb solder, of drinking water distribution systems potentially increase the burden of Pb (Li et al., 2019). Different biosolid production methods can also result in high Pb levels in the biosolids (Healy et al., 2016a).

### 1.3. Pathways of human exposure

For the non-smoking general population, the largest contributor to the daily intake of Pb is through ingestion of food including cereals and vegetables. Other sources include the effect of packaging (use of Pb-soldered food and beverage cans; which is now diminishing), use of Pb-glazed ceramic or pottery dinnerware, household plumbing systems containing Pb pipes, solders and fittings, dirt and dust (WHO, 2010). Smoking tobacco increases Pb intake. Consumption of fruit and vegetables (excluding potatoes and other starchy tubers) is recommended to prevent heart diseases, cancer, diabetes, obesity, several micronutrient deficiencies, especially in less developed countries. However, it has been shown that leafy vegetables and rootstalk vegetables have the greatest ability to accumulate heavy metals. Hence, measures must be taken to minimise heavy metal accumulation through such foodstuffs, for example, by restricting the growth of certain crops on contaminated soils (Zwolak et al., 2019). Wheat (*Triticum aestivum* L.) is the third most crucial cereal worldwide after the rice and maise, and the concentration of heavy metals, including Pb, is reported to decrease in the order of root > leaf > stem > grain in a wheat plant (Guo et al., 2018). Principal crops of interest produced in Ireland include barley, wheat, potatoes, and oats (CSO, 2020). The main vegetables include carrots, cabbage, broccoli, cauliflowers, swedes and parsnips, lettuce, onions, scallions, leeks (Bord Bia and DAFM, 2015). The density of arable lands in the 26 counties in the Republic of Ireland (CSO, 2012) is highlighted in green shades in Fig. A1. Predominantly arable lands, which produce significant vegetable crops in the Republic of Ireland, are highlighted in yellow in Fig. A1. The top 5 counties with intense arable lands are Carlow, Louth, Kildare, Dublin, Wexford. Also, the East Coast of Ireland is reported to have elevated levels of certain metal (loid)s in the soil (Nag et al., 2022b).

### 1.4. Guidelines and legislation around the maximum permissible levels of Pb in environmental media

Directive of 1986/278/EEC (The European Commission, 1986) limits the level Pb in the agricultural soil to between 50 and 300 mg kg$^{-1}$. Similarly, more refined allowable limits of metal(loid)s in the soil can be found in Irish guidelines. In the Codes of Good Practice for the Use of Biosolids in Agriculture (Department of the Environment and Local Government, 2009), the maximum permissible concentrations of Pb in the soil before the application of biosolids is reported as 80 mg kg$^{-1}$ (soil pH > 5.0 and clay content ≥ 15%). Based on the Water Framework Directive, Irish legislation S.I. No. 77 of 2019 (The European Union, 2019), the permissible limit of Pb in the surface water is 1.2 μg l$^{-1}$ whereas the limit of Pb in drinking water is set as 10 μg l$^{-1}$ Irish legislation S.I. No. 122 of 2014 based on The European Union (2014). Likewise, the threshold of Pb in drinking water in the United States is 5 μg l$^{-1}$ (U.S. Code of Federal Regulations, 2019) 5, while WHO (2017) recommends the limit as 10 μg l$^{-1}$. The limit of Pb in the sediments of streams is set as 1000 mg kg$^{-1}$ based on the regulation for livestock (the Irish EPA and GSI, 2009). A summary of the threshold limits for a suite of metal (loid)s including Pb can be found in Nag et al. (2022a) and Nag and Cummins (2021).

### 1.5. Relevant Irish studies around the levels of Pb in different types of biosolids

The range of Pb concentration in Irish pastures is typically 0.5–20 mg kg$^{-1}$ dry matter (Healy et al., 2016b). Metal(loid)s are typically important as a nutrient for plant growth; however, Pb has no function for such growth. According to 2002/32/EC, the maximum allowable concentration of Pb in animal feed is 30 mg kg$^{-1}$ (The European Union, 2002). Directive of 1986/278/EEC recommends the permissible concentration of Pb in biosolid sludge as 750–1200 mg kg$^{-1}$; and based on a ten-year average application, the maximum amount of Pb in the biosolids to be added to agricultural land is 15 kg ha$^{-1}$ year$^{-1}$ (The European Commission, 1986). Table A1 shows reported levels of Pb in a range of biosolids and soil in the Irish context. Lime stabilisation (LS) biosolids report lower Pb concentrations of 10.7 (SD ± 1.0) mg kg$^{-1}$ after raw dairy cattle slurry (DCS), which mean Pb concentration of <0.25 mg kg$^{-1}$. Bioaccumulation in Ryegrass is also reported (Healy et al., 2016b). Healy et al. (2016b) reported the limit values for metal concentrations in sludge for use in agriculture as 300 (Brazil), 300–1000 (China), 750–1200 (EU), 100 (Japan), 300 (Jordan), 250 (Russian FEd.), 300–840 (USA) mg kg$^{-1}$ dry weight (=ppm). AD treated sludge shows a very high (mean 791, ±SD 1625 mg kg$^{-1}$) concentration of Pb compared to TD (mean 54, ±SD 30 mg kg$^{-1}$) and LS

(33, ±SD 25 mg $kg^{-1}$) sludge (Healy et al., 2016b). A summary of the worldwide distribution of Pb variability in biosolids used in agriculture is captured in Fig. A2 (Healy et al., 2016a; LeBlanc et al., 2008; The European Commission, 1986; US EPA, 1993a). The wide-ranging Pb levels in different media trigger the need for risk assessment of Pb with a focus on the influence of anthropogenic sources (biosolids). The measured mean concentration of Pb in all types of treated sludge mentioned in Healy et al. (2016b) was 252 mg $kg^{-1}$. Out of 16 wastewater treatment plants (WWTPs) in Ireland studied by Healy et al. (2016b), one WWTP indicated a higher concentration of Pb (3696 mg $kg^{-1}$) in biosolids which exceeded the EU limit of 1200 mg $kg^{-1}$. Also, it must be noted that organic Pb is more toxic than inorganic Pb due to its lipid solubility; however, inorganic Pb is associated with an increased risk of cancer (Mani et al., 2019).

### *1.6. Tolerable intake level*

The tolerable level of metal intake is determined by dose-response relationships (Astolfi et al., 2019). The previously established provisional tolerable weekly intake (PTWI) of 25 μg kg body $weight^{-1}$ $week^{-1}$ was no longer considered appropriate and, therefore, withdrawn (WHO, 2010). Conversely, the Lowest Observed Adverse Effect Level (LOAEL) of Pb is 0.0012 mg kg body $weight^{-1}$ $day^{-1}$ (USEPA, 1987). Therefore, the daily tolerable limit of Pb can be calculated as 93 μg $day^{-1}$ for adults (for average body weight 78.1 kg) and 30 μg $day^{-1}$ for children (for body weight 25 kg) (Sharma et al., 2005). As indicated previously, the threshold of Pb in drinking water in the United States is 5 μg $l^{-1}$ (U.S. Code of Federal Regulations, 2019) while WHO (2010) sets the drinking water and air (inhalation) limits as 10 μg $l^{-1}$, and 0.5 μg $m^{-3}$ (annual average), respectively. There is a recommendation of initiating public health actions if blood Pb level (BLL) in children ≥ 5 μg $dl^{-1}$ (micrograms per decilitre) (Wu et al., 2020) (previously 10 μg $dl^{-1}$) and BLL > 25 μg $dl^{-1}$ for adults (Lo et al., 2012). Intensive medical management and chelation therapy are recommended if BLL is greater than 45 μg $dl^{-1}$ (Lo et al., 2012). As a preventive measure, the permissible concentration of Pb in foodstuff (The European Union, 2006) is documented in Table A2.

### *1.7. Risk assessment*

Exposure assessment and hazard characterisation are important parameters of human health risk assessment (Li and Cummins, 2020). A farm-to-fork exposure assessment has been carried out in this study, and the LOAEL was considered for hazard characterisation. A comparison between the methodologies (Gomes et al., 2019; Kebonye et al., 2017; Ramírez et al., 2020) suggested that risk indicator parameters and measures such as the Enrichment Factor (EF), Geochemical Mass Balance (GMB), Geoaccumulation Index ($I_{geo}$), Contamination Factor (CF), Pollution Load Index (PLI) can all be used in an initial hazard identification process (Nag et al., 2022a). Risk measures such as the Pollution Index (PI), Integrated Pollution Index (IPI) and Potential Ecological Risk Index (PERI) are based on traditional threshold values of metal(loid)s in the environmental media set by health authorities in different countries (Roşca et al., 2020); therefore, supporting them as mid-point analysis measures (Nag et al., 2022a). The other metal pollution assessment methods, such as the heavy metal index (HPI), heavy metal evaluation index (HEI), the degree of contamination (Cd), and water quality index (WQI) are used to determine the pollution status of water ecosystems to the heavy metals content (Roşca et al., 2020). Hazard Quotient (HQ), Hazard Index (HI), Risk Quotient (RQ), Daily Dietary Index (DDI), Daily Intake of Metal (DIM), Health Risk Index (HRI), Target Hazard Quotient (THQ), Cancer Risk (CR), Total Cancer Risk ($CR_{total}$) can be classified as endpoint analysis (Gupta et al., 2019; Mehta et al., 2018; Nag et al., 2021; Nain and Kumar, 2019; Zhuang et al., 2009) where exposure assessment and hazard characterisation concepts are combined to assess the overall potential human risk to pollutant metal(loid)s. There is no gold standard risk assessment method for evaluating risk from metal(loid)s. Methods in this area such as HQ, DDI, DIM, HRI and THQ need further development to reflect evolving risk assessment methodologies and evolution of dynamic monitoring databases. However, HQ, DDI, DIM, HRI, THQ, and CR have been identified as suitable for Pb risk assessment; hence, this study investigates these methods further. Also, the use of GIS in heavy metal risk assessment is widely used (Alavi et al., 2016; Dippong et al., 2020). To the best of the authors' knowledge, this probabilistic farm-to-fork quantitative human health risk assessment is the first study in the Irish context.

The overall aim of the study was

(i) To quantify the level of metal(loid)s arising from various sources (baseline levels and anthropogenic activities)
(ii) To develop an environmental exposure assessment for Pb contamination and
(iii) To identify predominant exposure pathways and pollutant controls to reduce the risks of detrimental health effects from selected metal(loid)s.

## 2. Materials and methods

A probabilistic human health risk assessment of lead (Pb) through the environmental-food pathway has been conducted. The methodology is based on the proposed risk assessment framework by Nag et al. (2022a). The development of a quantitative risk assessment framework model is exhibited in Fig. 1, and the schematic presents the steps required to build a risk assessment model. First, the baseline concentration (Nag and Cummins, 2021) of Pb in shallow (A) and deep (S) soil, water (W), and sediment (C) samples were collated from open source spatial data provided by the Tellus project (GSI, 2019). The raw data were treated and analysed by ArcGIS (ArcMap version 10.7). Next, the anthropogenic concentration of Pb (i.e. in biosolids) was collated from the designated authority. Next, land use classification, such as arable lands and pastures, was determined using the CORINE land use map (EPA Ireland, 2018). These steps predicted the maximum metal(loid)s concentration in soil and surface water, including surface water near the drinking water treatment plants' locations. Next, the variability of Pb baseline levels in soil and water was captured using a fitted lognormal distribution. This step was followed by the bioaccumulation of Pb (from soil to vegetables) and, finally, potential uptake by humans through crops and/or water. The consumption of different categories of food, water and beverages can be found using the Irish Universities Nutritional Alliance Survey (IUNA, 2011) for the consumers with extrapolation to the entire population. Finally, the daily human exposure ($HE_{daily}$) of Pb was calculated by multiplying the concentration of Pb in food/water by the daily ingestion of specified foodstuffs. This stepwise probabilistic model was run by Monte Carlo simulation (10,000 iterations) with @RISK 7.5 software (Palisade inc.), an add-in to Microsoft Excel version 2016. The simulated mean $HE_{daily}$ was used to evaluate human health risk using established risk assessment measures such as HQ, DDI, DIM, HRI, THQ, and CR, most relevant for Pb risk assessment.

### *2.1. Selection of study area*

The spatial (Tellus project) data on A, S, W and C samples is available for northern counties of the Republic of Ireland (GSI, 2019). These counties are Donegal, Sligo, Leitrim, Cavan, Monaghan, Louth, Mayo, Galway. As, Cd, Hg, and Pb are identified as the most toxic metal(loid)s to humans and the ecosystem (ECHA, 2020). The Soil Geochemical Atlas of Ireland (Fay et al., 2007), which is lower resolution data and available for the entire country of Ireland, indicates the elevated levels of As, Cd, Hg and Pb are observed along the Eastern coast of Ireland. Within the available higher resolution Tellus data, elevated Pb levels have been noticed in some areas of Co. Louth. Furthermore, Co. Louth

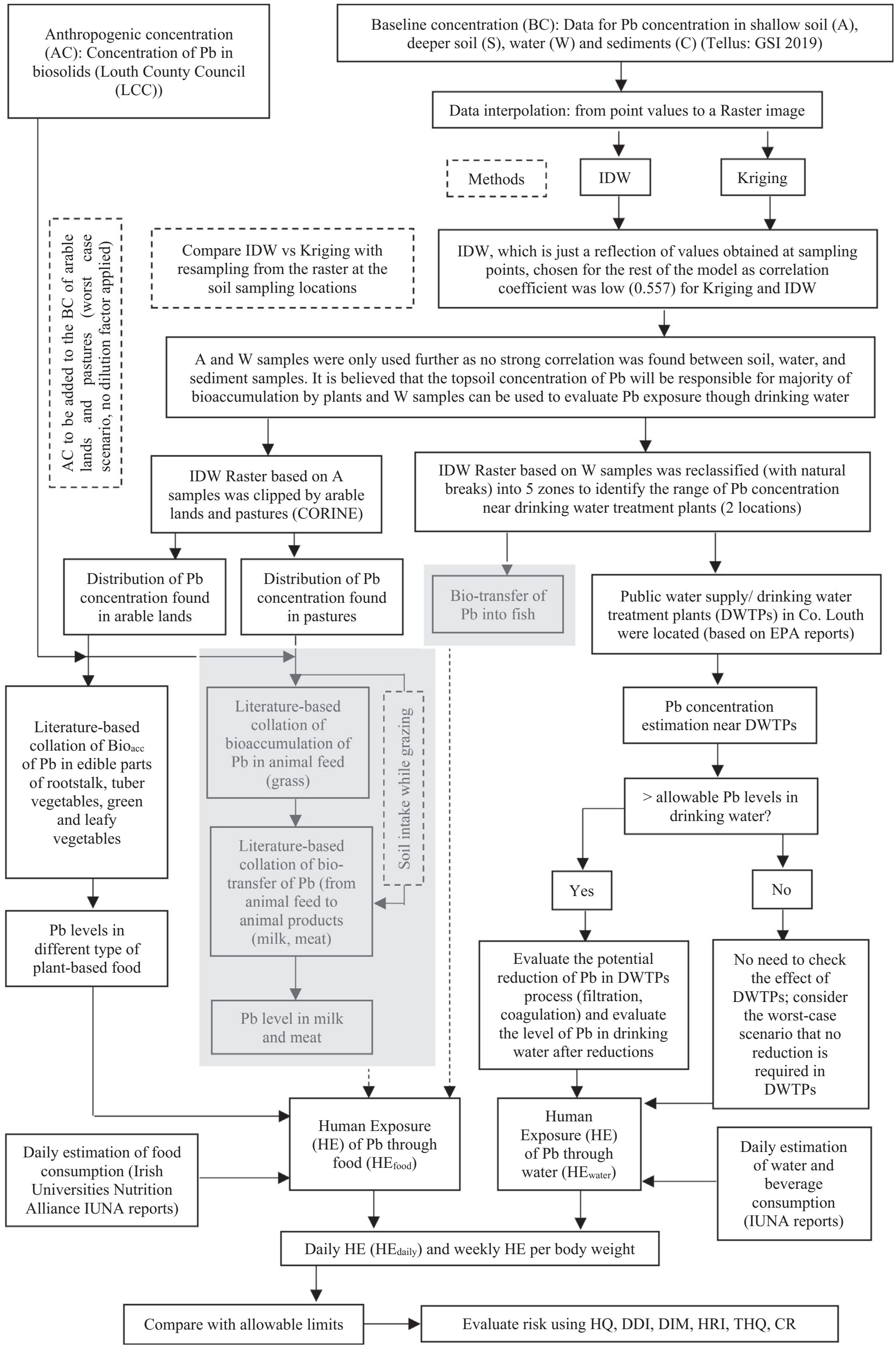



**Fig. 1.** A schematic presentation of risk assessment methodology; greyed pathway not considered in this study.

is a county that holds one of the densest arable lands (Fig. A1). Therefore, Co. Louth is the county of priority for which spatial data is available and therefore, selected as the study area. Co. Louth is stretched between N 54° 06′ 50″ (North) to N 53° 41′ 54″ (South) and between E - 6° 06′ 10″ (East) to E - 6° 41′ 42″ (West). The spatial distribution of land use is presented in Fig. 2a. The total population of Co. Louth was 122,897 in 2011 (CSO, 2012). The total area of agricultural land in Co. Louth is 709.83 km$^2$, which is 85.96% of the total land use (Fig. 2b), including 428.59 km$^2$ of pastures (60.37% of total agricultural land) and 238.9 km$^2$ of arable lands (33.65% of total agricultural land).

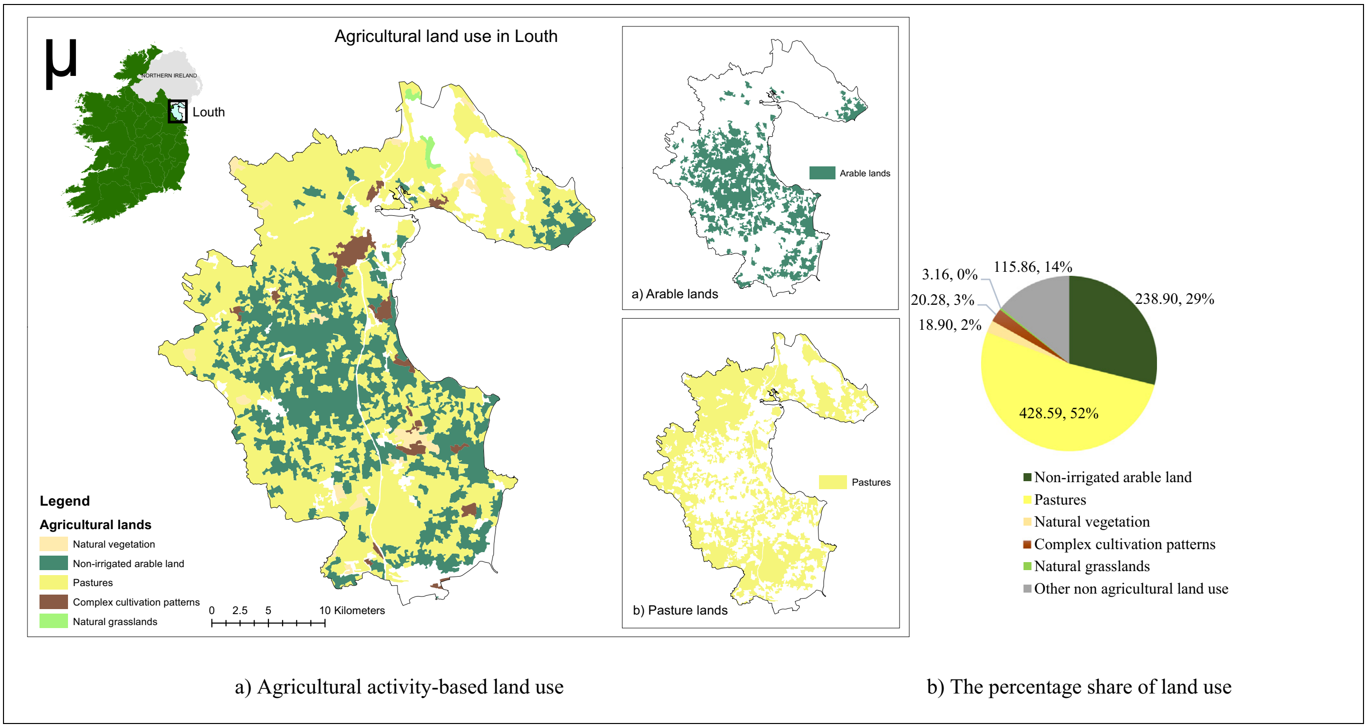



**Fig. 2.** Agricultural activity-based land (Co. Louth) and the percentage share of land use where the total land area is 825.69 $km^2$ (=100%).

### *2.2. Interpolation of Tellus-project raw data with IDW and Kriging*

The interpolation objective was to convert discrete data points (sample A, S, W, and C) to a continuous raster image for analysis. According to the ArcGIS manual, "*Inverse Distance Weighting (IDW) is a quick deterministic interpolator that is exact. There are very few decisions to make regarding model parameters, and Kriging is an interpolator that can be exact or smoothed depending on the measurement error model.*" Both IDW and Kriging were performed, and a correlation was checked to see which interpolation method performed better to represent the sampled data points. IDW was found more realistic to signify the data points, and sample A and W were found most appropriate for food (bioaccumulation) and drinking water pathway, respectively.

### *2.3. Anthropogenic source: biosolids*

Agricultural practices were evaluated in the study area, with the rate of biosolids applied in the area noted. Out of seven observations, biosolids' application rate on arable land in the area varied between 26.8 and 34.8 ton $ha^{-1}$. Biosolids' density was 4 ton $m^{-3}$ (dry solid DS) with organic matter of 3% (of DS). The level of Pb in biosolids was found as 7.9 mg $kg^{-1}$ for all study sites.

### *2.4. Bioaccumulation of Pb*

The bioaccumulation factor ($Bio_{acc}$) is a ratio (Eq. (1)) of the concentration of pollutants found in the plant tissue ($C_{Pb\_plant}$) and the concentration of pollutants in the soil ($C_{Pb\_soil}$) (Guo et al., 2019). $Bio_{acc}$ has been reported to significantly differ among wheat species tested for As, Cd, and Pb. Cultivars reportedly accumulate Cd more readily than As and Pb in wheat (Guo et al., 2018). The $Bio_{acc}$ of metals in a wheat plant body was found to be in the order $Bio_{acc\_root}$ > $Bio_{acc\ leaf}$ > $Bio_{acc\ stem}$ > $Bio_{acc\_grain}$. The typical order of bioaccumulation of metals in plants is found to be in the order of $Bio_{acc\_root}$ > $Bio_{acc\_stem}$ > $Bio_{acc\ leaf}$ > $Bio_{acc\_fruit}$ > $Bio_{acc\_seed}$, unless there is an abnormally high concentration of volatile metals (e.g. volatile mercury Hg) in the air, which may increase the concentration of metals in leaves due to the presence of volatile metal pollutants during the stomatal gas exchange (Beckers and Rinklebe, 2017). A review of the studies reporting metal bioaccumulation in crops and their components (rootstalk, tuber vegetable, leafy vegetables, grain crops, fruit crops, oil crops) is presented in Table 1.

$$Bio_{acc} = \frac{C_{Pb\_plant}}{C_{Pb\_soil}} \tag{1}$$

A positive correlation (steep linear slope initially) between the concentration of metal(loid)s in soil, and the bioaccumulation has been observed (Dudka et al., 1996). While intact potato tubers showed a similar relationship for Pb, there is a non-linear relationship between the concentration of Pb in soil and bioaccumulation of Pb in peeled potato tubers; which means a particular $Bio_{acc}$ is dependent upon the initial level in the soil ($C_{Pb\_soil}$) and also, the portion of the plant that is eaten. Guo et al. (2019) discovered that the $Bio_{acc}$ of As and Cd were in the order of leafy > rootstalk > fruit > legume vegetables, and the $Bio_{acc}$ of Cu, Pb, and Zn were in the order of leafy > rootstalk > legume > fruit vegetables. Also, Zhuang et al. (2009) discovered that the average $Bio_{acc}$ values (Cu, Zn, Pb, Cd) of leaf vegetables were significantly higher than non-leafy vegetables. For grain crops such as rice, the average concentration of Cu, Zn, Pb, and Cd decreases in the order of stalk > husk > grain, except that concentrations of Cu and Cd in the grain were higher than those in the husk. The range of $Bio_{acc}$ of heavy metals in the vegetables was reported to decrease in the order of Cd (0.02 to 0.08, mean 0.055), Zn (0.018 to 0.035, mean 0.024), Cu (0.015 to 0.022, mean 0.018), As (0.0025 to 0.012, mean 0.007), and Pb (0.001 to 0.004, mean 0.002) and the daily ingestion rate of vegetables was reported as the most sensitive parameters of the model followed by the concentration of Pb in vegetables (Guo et al., 2019). Also, Zhuang et al. (2009) found that $Bio_{acc}$ of heavy metals in different vegetables were in descending order of Cd > Zn > Cu > Pb with Pb $Bio_{acc}$ ranging 0.007–0.016 (leafy vegetables), 0.0035–0.0085 (fruit vegetables), 0.002–0.009 (root vegetables), 0.003–0.01 (rice) and having a $C_{Pb\_soil}$ range between 6.8 and 127 (mean 8.4 to 75) mg $kg^{-1}$ (Table 1). The Pb $Bio_{acc\_root}$, $Bio_{acc\_leaf}$, $Bio_{acc\_stem}$, $Bio_{acc\_grain}$

**Table 1**
Estimated bioaccumulation factors ($Bio_{acc}$) of Pb for different types of crops.

| Plant | Scientific name | Type of crop | Concentration in soil (mg kg$^{-1}$) | Pb $Bio_{acc}$ | Pb $Bio_{acc}$ adopted | Reference |
|---|---|---|---|---|---|---|
| Carrots | *Daucus carota* subsp. *Sativus* | Root/rootstalk vegetables | 6.8–127 (mean 8.4–75) | 0.002–0.009 | Uniform (0.002,0.009) | (Zhuang et al., 2009) |
| Potatoes | *Solanum tuberosum* | Tuber vegetable | 18.03–24.90 | 0.005–0.034 | Uniform (0.005,0.034) | (Musilova et al., 2016) |
| Green vegetables | – | Green vegetables | 6.8–127 (mean 8.4 to 75) | 0.007–0.016 | Uniform (0.007,0.016) | (Zhuang et al., 2009) |
| Leafy vegetables | – | Leafy vegetables | 6.8–127 (mean 8.4 to 75) | 0.007–0.016 | Uniform (0.007,0.016) | (Zhuang et al., 2009) |

values ranged from 0.23–0.89, 0.021–0.044, 0.0038–0.016, and 0.0007–0.0024, respectively, for 16 wheat cultivars (Guo et al., 2019). In the experiment, the $C_{Pb\_soil}$ ranged from 23.24 to 609.49 mg kg$^{-1}$ (median 122.2, mean 196.96, SD 165.11 mg kg$^{-1}$), which is slightly higher than the $C_{Pb\_soil}$ in the study area stretching from 7 to 264 (mean 43) mg kg$^{-1}$. Therefore, $C_{Pb\_soil}$ indicated in these studies (Guo et al., 2019; Zhuang et al., 2009) has a close match with the $C_{Pb\_soil}$ in the current study area can be used as the most appropriate Pb $Bio_{acc}$ for the probabilistic model.

### 2.5. Daily estimation of daily dietary intake (DDI): Food and water consumption

An estimated daily dietary intake (DDI) of food products was considered from the Irish Universities Nutritional Alliance Survey (IUNA, 2011). Water consumption is considered as lognormally distributed (Clarke et al., 2017), with mean daily consumption of 564 g (SD 617) (IUNA, 2011). This study's selected food products are potatoes (boiled, mashed, baked), green vegetables, carrots, and salad vegetables (e.g. lettuce). Based on the IUNA (2011), the daily food intake (unit g day$^{-1}$) in the Irish population (18–64-year-olds) are considered as; potato (mean 71, SD 74), green vegetables (mean 13, SD 23), carrots (mean 13, SD 19), and salad vegetables (mean 21, SD 28). All mean and standard deviation values were used to generate lognormal distribution for all food products.

### 2.6. HQ, DDI, DIM, HRI, THQ, CR for Pb risk assessment

The reference dose RfD is a benchmark dose derived from the No Observed Adverse Effect Level (NOAEL) divided by an uncertainty factor (UF) and a modifying factor (MF) (Eq. (2)). Benchmark Dose (BMD) is an exposure due to a dose of a substance associated with a specified low incidence of risk, generally in the range of 1% to 10%, of a health effect; or the dose associated with a specified measure or change of a biological effect (EPA, n.d.). The BMD approach aims to define a starting point of departure (POD) for computation an RfD. There is no recommendation of NOAEL for Pb by the US EPA; instead, the POD of Pb is recommended in terms of 'Lowest Observed Adverse Level' (LOAEL) of Tetraethyl Pb and is recommended as 0.0012 mg kg$^{-1}$ day$^{-1}$ (0.0017 mg kg$^{-1}$ day$^{-1}$ × 5 days / 7 days) (USEPA, 1987). UF and MF values are recommended as 10,000 and 1, respectively, for Pb (US EPA, 1993b). Therefore, reference Dose for Oral Exposure ($RfD_o$) is calculated as $1 \times 10^{-7}$ mg kg$^{-1}$ day$^{-1}$. However, $RfD_{ingestion}$ and $RfD_{skin-contact}$ were referred to as $1.4 \times 10^{-3}$ and $0.42 \times 10^{-3}$ mg kg$^{-1}$ day$^{-1}$, respectively (Nguyen et al., 2019). RfD due to ingestion is referred to as $4 \times 10^{-3}$ mg kgbw$^{-1}$ day$^{-1}$ (Aendo et al., 2019; Guo et al., 2018; Pipoyan et al., 2019; Zhuang et al., 2009). Also, RfD and cancer slope factor (CSF) for Pb was indicated as 0.004 mg kgbw$^{-1}$ day$^{-1}$, 0.0085 (mg kgbw$^{-1}$ day$^{-1}$)$^{-1}$, respectively, in a different study (Sarwar et al., 2019).

$$RfD = \frac{NOAEL}{UF \times MF} \quad (2)$$

Six risk measures (HQ, DDI, DIM, HRI, THQ, CR) were adopted for this study as each of these methods includes endpoint analysis (allowable value for human ingestion), and the methods were documented in Table 2. The DDI and DIM can be compared with Recommended Daily Allowances (RDA), which is the average daily dietary intake level required to meet the nutrient requirements of the majority (97–98%) of healthy people of a specific sex, age, life stage, or physiological condition (such as pregnancy or lactation) (Zohoori and Duckworth, 2019). There is no requirement for Pb ($RDA_{Pb}$) as it does not contribute to body functions (Luis et al., 2014). The allowable limits collated in Jin et al. (2014) suggests that the allowable limit of Pb intake varies globally (for example, in France (0.20 μg kgbw$^{-1}$ day$^{-1}$), Canada (0.13 μg kgbw$^{-1}$ day$^{-1}$), Australia (0.12–0.13 μg kgbw$^{-1}$ day$^{-1}$), Lebanon (0.14 μg kgbw$^{-1}$ day$^{-1}$), Germany (0.26 μg kgbw$^{-1}$ day$^{-1}$), Korea (0.41 μg kgbw$^{-1}$ day$^{-1}$), Spain (1.21 μg kgbw$^{-1}$ day$^{-1}$) and Italy (0.92 μg kgbw$^{-1}$ day$^{-1}$)). A distribution was fitted to the weekly equivalent (above) limits to capture the variability around the allowable limit (Fig. A3). The mean allowable weekly intake (AWI) limit was calculated as 2.7 μg kgbw$^{-1}$ week$^{-1}$. In the absence of a recommended limit, this limit can compare the simulated human exposure to Pb. Hence, an assumption can be made for RDA based on allowable weekly intake (AWI), 0.0004 mg kgbw$^{-1}$ day$^{-1}$ (=0.0027/7). However, it must be noted that AWI is introduced for a comparison purpose as this is an average of the global limit and multiple regions, and absolute risk can be evaluated by widely accepted risk measures such as HQ, DDI, DIM, HRI, THQ, CR. According to Qu et al. (2018), cited in (Nguyen et al., 2019), carcinogenic risks (CR) acceptance by the USEPA ranged from $1 \times 10^{-6}$ to $1 \times 10^{-4}$, in which risks exceeding $1 \times 10^{-4}$ are considered as unacceptable while the risks below $1 \times 10^{-6}$ are not likely to pose significant health risks. A moderate CR value $1 \times 10^{-3}$ was proposed by (Pipoyan et al., 2019), and also the threshold level ($CR > 10^{-4}$) was suggested. The adopted risk classification for CR is documented in Table 2.

### 2.7. Scenario analysis

Along with the baseline scenario, three other scenarios were performed (Table 3). Scenario S1 looked at Pb levels' and anthropogenic Pb's addition to the baseline concentration (BC) Pb in soil. This anthropogenic level of Pb was received from the study area. Scenario S2 was set to look at the influence of the variability of the level of Pb in the biosolids as distribution was fitted to the data presented in Table A1. Finally, S3 represents a hypothetical scenario where it is assumed that no one consumes potatoes; the objective was to check the influence of potato consumption which appeared as the most commonly eaten solid food product among selected vegetables in this study (IUNA, 2011).

## 3. Results

### 3.1. Data analysis on Tellus data release

The spatial distribution of Pb concentration is presented in Fig. A4. The concentration of Pb in soil (A and S samples) ranged from 7 to 264 and 4–258 mg kg$^{-1}$, respectively, while Pb concentration in surface water (W samples) ranged 0.01–0.564 μg l$^{-1}$ which is less than the lowest

**Table 2**
Risk assessment measures used in this study.

| Name of method | Equation | Parameters and unit | Classification of risk | Reference |
|---|---|---|---|---|
| Hazard quotient (HQ) | $HQ = DI \times \frac{C_{Mveg}}{RfD_o \times BM}$ (3) | DI = daily intake of vegetable (kg day$^{-1}$)<br>$C_{Mveg}$ = concentration of metal in vegetable (mg kg$^{-1}$)<br>BW = the average body weight, adult (78.1 kg[a])<br>$RfD_o$ = oral reference dose for the metal; varies (1.4 × 10$^{-3}$, 4 × 10$^{-3}$ mg kg$^{-1}$ day$^{-1}$) | HQ > 1 risk (non-cancer)<br>HQ < 1 no adverse health effects | (Singh and Kumar, 2020)<br>$RfD_o$ (USEPA, 1987), (Nguyen et al., 2019), (Sarwar et al., 2019) |
| Daily Dietary Index (DDI) | $DDI = A \times B \times \frac{C}{BW}$ (4) | A = metal content in vegetable (mg kg$^{-1}$)<br>B = dry weight of the vegetable consumed (kg)<br>C = approximate daily intake of vegetable (kg day$^{-1}$)<br>BW = average human body mass (78.1 kg[a]) | Can be compared to RDA, but for Pb, there is no RDA. Assumption can be made for RDA based on AWI, **0.0303 mg day$^{-1}$** (=0.0027 × 78.1/7) | (Gupta et al., 2019) |
| Daily Intake of Metal (DIM) | $DIM = A \times C \times \frac{D}{BW}$ (5) | A = Metal(loid) concentration in plants (mg kg$^{-1}$)<br>C = conversion factor (0.085 is to convert fresh vegetable weight to dry weight)<br>D = daily intake of vegetables (kg day$^{-1}$)<br>BW = the average body weight, adult (78.1 kg[a]) | Can be compared to RDA, but for Pb, there is no RDA. Assumption can be made for RDA based on AWI, **0.0004 mg kg$^{-1}$ day$^{-1}$** (=0.0027/7) | (Tsafe et al., 2012) |
| Health Risk Index (HRI) | $HRI = \frac{DIM}{RfD}$ (6) | As above | HRI < 1 safe | (Gupta et al., 2019) |
| Target Hazard Quotient (THQ) | $THQ = \frac{EF_r \times ED \times FI \times MC}{RfD_o \times BW \times AT} \times 10^{-3}$ (7) | $EF_r$ = exposure frequency (365 days year$^{-1}$)<br>ED = exposure duration (70 years)<br>FI = food ingestion (g person$^{-1}$ day$^{-1}$)<br>MC = the metal concentration in food (μg g$^{-1}$, on a fresh weight basis)<br>$RfD_o$ = oral reference dose for the metal; varies (1.4 × 10$^{-3}$, 4 × 10$^{-3}$ mg kg$^{-1}$ day$^{-1}$)<br>BW = the average body weight, adult (78.1 kg[a])<br>AT = averaging time for noncarcinogens (365 days year$^{-1}$ × number of exposure years, assuming 70 years assumed) | THQ < 1 unlikely to experience adverse effects | (Zhuang et al., 2009)<br>$RfD_o$ (USEPA, 1987) |
| Cancer Risk (CR) | $CR = ADD \times CSF$ (8)<br>Where $ADD = \frac{C \times IR \times EF \times ED}{BW \times AT}$ (9) | ADD = average daily dose (mg kg$^{-1}$ day$^{-1}$)<br>CSF = cancer slope factor (0.0085 mg kg$^{-1}$ day$^{-1}$)$^{-1}$<br>ADD can be computed from Eq. (9).<br>C = concentration of pollutants (mg kg$^{-1}$ or mg l$^{-1}$)<br>IR = ingestion rate (kg day$^{-1}$ or l day$^{-1}$)<br>EF = exposure frequency (days year$^{-1}$)<br>ED = exposure duration (years)<br>BW = the average body weight, adult (78.1 kg[a])<br>AT = average time (day) | CR (for a single contaminant) > 1 × 10$^{-6}$ carcinogenic risk<br>Adopted classification for the model based on Pipoyan et al. (2019)<br>High risk ≥ 0.001;<br>Moderate risk ≥ 0.0001 but <0.001;<br>Low risk ≥ 0.000001 but <0.0001;<br>Very low risk < 0.000001 | (Mehta et al., 2018; Nain and Kumar, 2019)<br>CSF (Sarwar et al., 2019) |

[a] The average weight of 78.1 kg was considered based on 86.9 kg (18 to 64 y older male) and 70.375 kg (18 to 64 y older female). Beyond 65 y age, it is 82.4 kg and 68.1 kg for male and female, respectively.

limit of Pb (5 μg l$^{-1}$) as set by the U.S. Code of Federal Regulations (2019). The level in sediment was found to be between 13 and 108 mg kg$^{-1}$. A histogram of A soil samples collected for Co. Louth is presented in Fig. A5, which suggests that 76% of the observations were below 50 mg kg$^{-1}$ (EU lower threshold), and in 94% cases, the concentration of Pb in the soil was below 80 mg kg$^{-1}$ ((Department of the Environment and Local Government, 2009)); the Pb concentration in the soil (max 264 mg kg$^{-1}$, Figs. A4 and A5) was observed to be below the permissible upper threshold of 300 mg kg$^{-1}$ set by the EU.

### 3.2. Spatial data interpolation

The result of two interpolation methods (IDW and Kriging) is illustrated in Fig. A6a and SF6b, respectively. With resampling (at the same sampling locations) Pearson correlation coefficient between IDW and Kriging was calculated as +0.557, which means a moderate uphill (positive) relationship could be established. With a comparison between histograms of IDW and Kriging (Fig. A7), it was found that the IDW raster was full of contrast; however, the Kriging raster uses a weighted

**Table 3**
Scenario analysis: deviation from the baseline scenario.

| Abbreviation | Scenario name | Deviation from the baseline scenario |
|---|---|---|
| BS | Baseline scenario | NA: Note – Lognorm (41.1,21.5) used for baseline concentration; range 14–188 mg kg$^{-1}$ |
| S1 | With biosolids: Pb level received from Louth County Council: 7.9 mg kg$^{-1}$ | BS: No anthropogenic concentration of Pb is added to Pb levels in background soil<br>S1: Concentration of Pb in soil = Baselineconcentration of Pb + monitored level of Pb in biosolids |
| S2 | With biosolids: fitted distribution | S2: Concentration of Pb in soil = Baseline concentration of Pb + uniform(0, monitored level of Pb in biosolids with fitted distribution on published literature values documented in Table A1) |
| S3 | No potato consumed | A hypothetical scenario where it is assumed that no potato is ingested |

average. Therefore, IDW was chosen for better representation of the sampled data points, and the result of IDW was used further in the analysis.

*3.3. Correlation between A, S, W, and C sample values*

An analysis for correlation coefficient between 4 samplings (A, S, W, and C) revealed that there is a strong (positive) linear relationship (coeff. + 0.958) between A and S sampling (Table A3) which would seem obvious in that the topsoil layer is a derivative of the soil layer beneath. Furthermore, a moderate uphill relationship was established between Pb levels in soil samples (coeff. + 0.530, +0.521) and sediment samples; conversely, a weak uphill (positive) linear relationship (coeff. + 0.313) was found between Pb levels in water and sediment points.

*3.4. Estimation of Pb concentration in topsoil and surface water*

The clipped raster images (Fig. 3) of arable land and pastures states that the variability of Pb concentration in topsoil ranges from 14.0 to 188.0 (mean 39.7) and 7.0 to 248.4 (mean 41.1) mg $kg^{-1}$ in arable lands and pastures, respectively. Therefore, the Pb level in pastures is higher compared to arable lands. The overall Pb concentration in surface water was 0.01–0.563 μg $l^{-1}$; however, the range of Pb levels near the drinking water treatment plants (plants A and B) was found as 0.01–0.034 μg $l^{-1}$.

*3.5. Exposure assessment: daily and weekly human exposure*

The human exposure (HE) and calculated Weekly Intake Per Body Weight (WIPBW) are presented in Fig. 4. The daily HE ranged from 0.013 mg $day^{-1}$ (5th %ile) to 0.211 mg $day^{-1}$ (95th %ile) with a mean of 0.073 mg $day^{-1}$ (Fig. 4a) while a mean WIPBW was calculated as 0.0065 mg kg $bw^{-1}$ $week^{-1}$ (5th %ile 0.001 and 95th %ile 0.018, Fig. 4b) which is greater than the AWI of 0.0027 mg kg $bw^{-1}$ $week^{-1}$ (Fig. A3). The simulated mean WIPBW of 0.0065 mg kg $bw^{-1}$ $week^{-1}$ is equivalent to 0.0009 mg kg $bw^{-1}$ $day^{-1}$, lower than the LOAEL of 0.0012 mg kg $bw^{-1}$ $day^{-1}$.

*3.6. Scenario analysis and goal-seek function*

Fig. 5 demonstrates that drinking water to the overall HE is negligible compared to crops and vegetables. It should be noted that this study

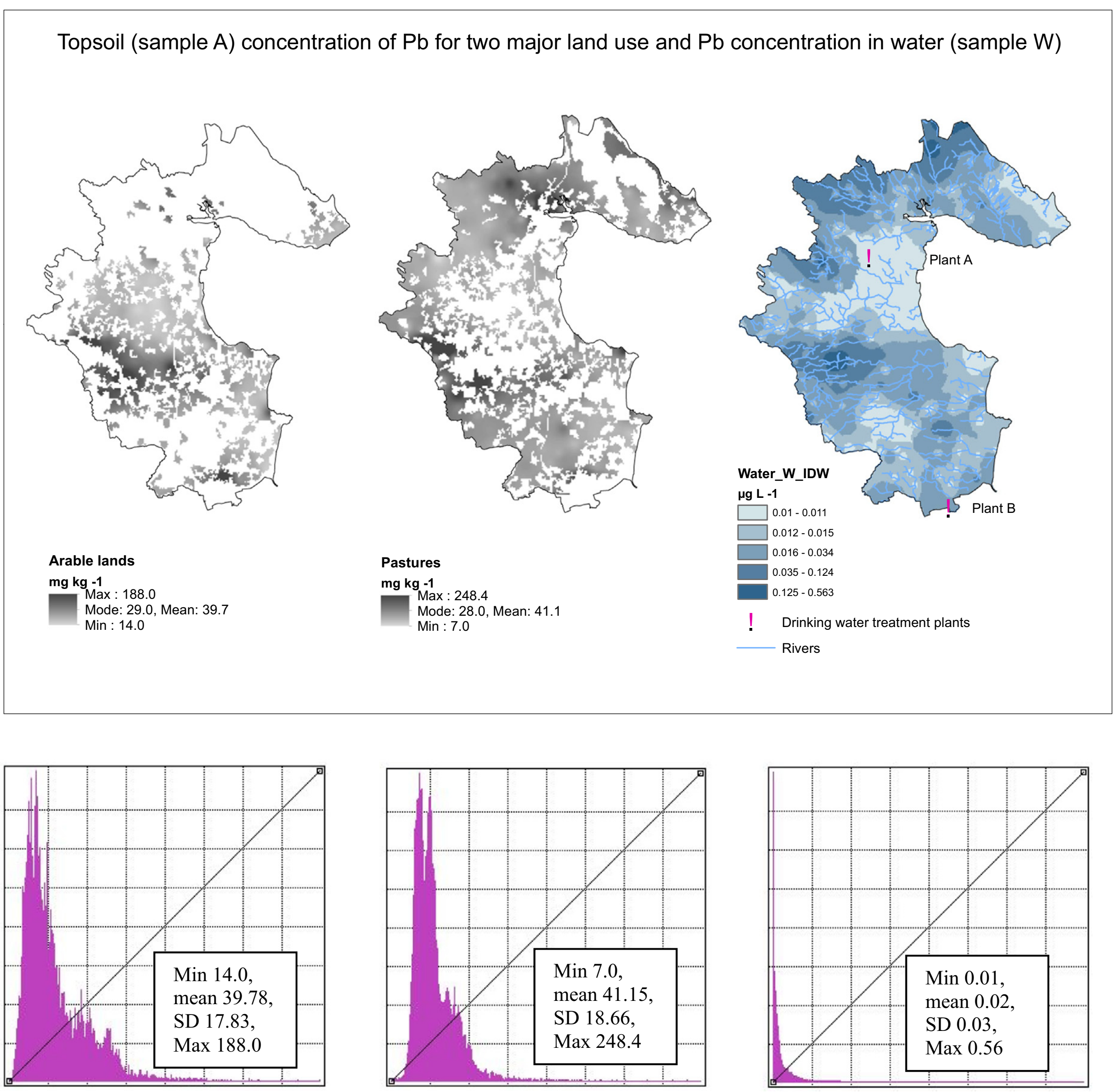
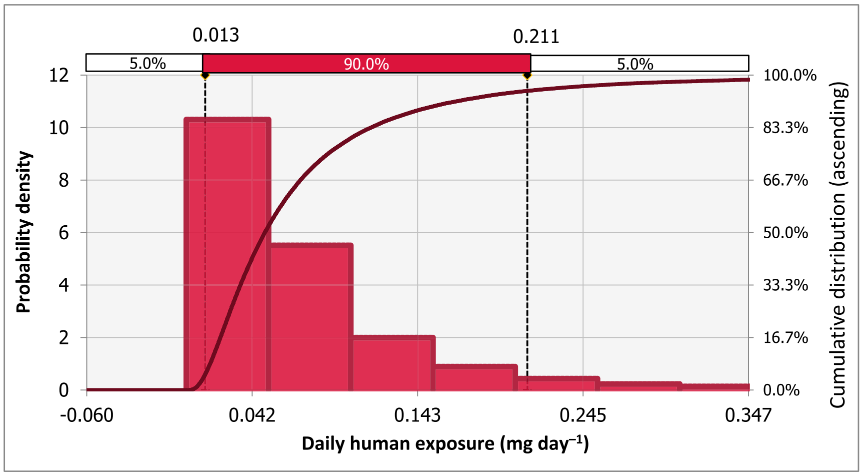
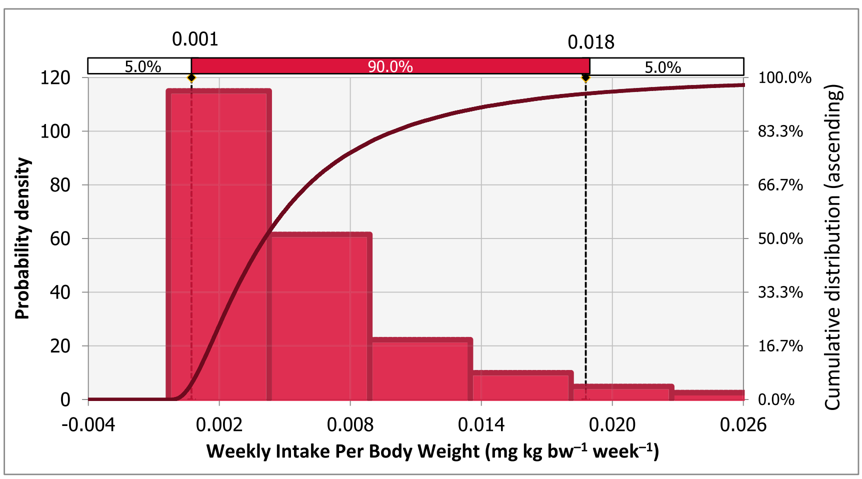



**Fig. 3.** The regional and data variability distribution of Pb concentration in topsoil (Sample A) of arable lands, pastures, and surface water.




**a) Daily Pb exposure**




**b) WIPBW**

**Fig. 4.** Daily (a) and weekly (b) human exposure to Pb.

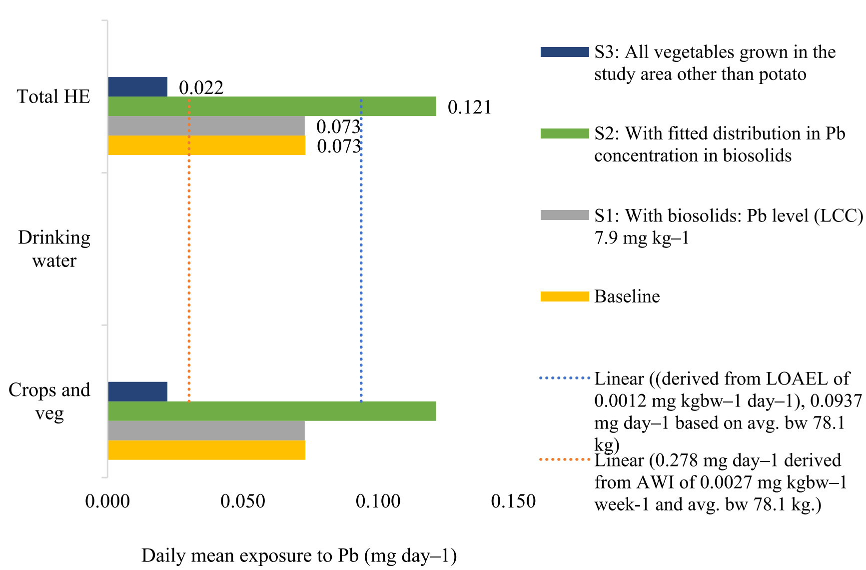



**Fig. 5.** Scenario analysis for daily mean human exposure (HE) to Pb.

focussed on geological sources for surface water contamination and does not consider potential contamination in the distribution network (i.e. viz. lead pipes) (EPA, 2019). The scenario analysis (Fig. 5) indicates that the variability in the Pb levels in biosolids can be a crucial parameter of the model as the daily mean HE to Pb for S2 was the highest (0.121 mg day$^{-1}$) followed by baseline and scenario S1 (0.073 mg day$^{-1}$). The lowest HE value was observed corresponding to scenario S3 (0.022 mg day$^{-1}$).

### 3.7. Evaluated risk

Hazard Quotient (HQ), Daily Dietary Index (DDI), Daily Intake of Metal (DIM), Health Risk Index (HRI), Target Hazard Quotient (THQ) and Cancer Risk (CR) were found as 0.234 to 0.669, 0.002, 0.0002, 0.020 to 0.057, 0.234 to 0.669, and 0.00001, respectively (Table 4) which signifies a low to moderate risk posed by Pb. DDI, DIM, HRI, CR indicated 'safe' or 'no risk' or 'low risk' due to human exposure to Pb. HQ and THQ displayed 'no adverse health effects' and 'no risk', respectively, at higher RfDo of Pb as $4 \times 10^{-3}$ mg kgbw$^{-1}$ day$^{-1}$. This RfD$_o$ value is also considered in recent studies (Aendo et al., 2019; Guo et al., 2018; Pipoyan et al., 2019; Zhuang et al., 2009). However, at lower RfDo of $1.4 \times 10^{-3}$ mg kgbw$^{-1}$ day$^{-1}$ (Nguyen et al., 2019), HQ and THQ pose 'non-cancer-risk' and 'risk', respectively.

## 4. Discussion

This study highlighted the importance of GIS tools to collate data for Pb levels in soil and water. Pb concentration in topsoil varied from 14 to 188 (mean 39.7) and 7.0 to 248.4 (mean 41.1) mg kg$^{-1}$ in arable lands and pastures, respectively. Based on the literature reviewed, the highest potential bioaccumulation was observed in potatoes (5th percentile 0.006, mean 0.019, 95th percentile 0.033), followed by leafy vegetables (5th percentile 0.007, mean 0.011, 95th percentile 0.015) and carrots (5th percentile 0.002, mean 0.005, 95th percentile 0.009). The daily mean HE to Pb for the general population was found to be 0.073 mg day$^{-1}$, where the mean weekly exposure was calculated as 0.0065 mg kg bw$^{-1}$ week$^{-1}$. The fitted distribution to the biosolids (S2) scenario increases the daily HE from 0.073 to 0.121 mg day$^{-1}$ compared to the no-biosolids or baseline scenario. A sensitivity analysis was performed, and the result (Fig. 6) revealed that the consumption of potato, as the largest food group, is the most sensitive parameter of the positively correlated model (coeff. + 0.66) the HE. This variability (or uncertainty) is followed by concentration of Pb in the arable soil (+0.49), bioaccumulation factor of tuber vegetables (+0.37), consumption of salad vegetables (+0.20), and green vegetables consumption (+0.13) which are in topmost sensitive parameters. A goal-seek function was used in @RISK software (Advance Analyses) to estimate

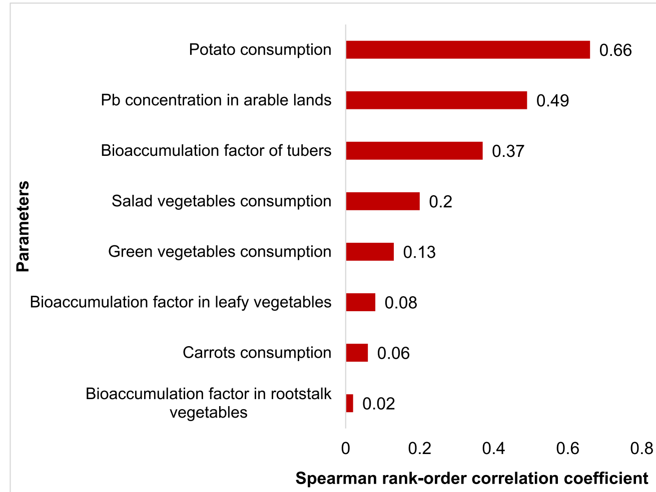



**Fig. 6.** Sensitivity analysis and Spearman rank-order correlation coefficients of parameters showing the influence of the model input variabilities.

the maximum level in soil media, resulting in an exceedance of the LOAEL of 0.0012 mg kgbw$^{-1}$ day$^{-1}$. The back-calculated threshold was found to be 51 mg kg$^{-1}$, which would result in the mean daily human exposure to Pb below 0.0012 mg kgbw$^{-1}$ day$^{-1}$. This observation ensures limited uptake and bioaccumulation by plants, ensuring reduced Pb human exposure through the 'environment: food' pathway. Also, the back-calculated limit Pb in the soil (51 mg kg$^{-1}$) is in line with the permissible range for soils (range 50–300 mg kg$^{-1}$) recommended by the European Commission (The European Commission, 1986) but below the limit of 80 mg kg$^{-1}$ recommended by the Codes of Good Practice for the Use of Biosolids in Agriculture before application of biosolids to agricultural land (Department of the Environment and Local Government, 2009). The quantitative study on Pb exposure considers a worst-case scenario as the overall concentration of Pb is treated as a summation of baseline concentration plus a concentration in biosolids. It should be noted that this is a pessimistic approach as the actual level is likely to be less than this due to the loss of Pb following a combination of land application of biosolids and rainfall events. At lower RfDo of $1.4 \times 10^{-3}$ mg kgbw$^{-1}$ day$^{-1}$ (Nguyen et al., 2019), HQ and THQ pose 'non-cancer-risk' and 'risk', respectively. If HQ is greater than 1, the pollution might result in a potential risk (non-carcinogenic) to human health (Nguyen et al., 2019; Qu et al., 2018). However, Sarwar et al. (2019) raised an important point: it may be overestimated if only the HQ is checked; instead, the hazard index (HI) must be evaluated considering other metals.

**Table 4**
The evaluated risk for baseline and no-biosolid scenario using multiple risk evaluation methods.

| Methods* | Baseline score | Risk evaluation | S1 score | Risk evaluation | S2 score | Risk evaluation | S3 score | Risk evaluation |
|---|---|---|---|---|---|---|---|---|
| HQ | | | | | | | | |
| (RfD$_o$ $4 \times 10^{-3}$) | 0.234 | NoAHE[a] | 0.233 | NoAHE[a] | 0.389 | NoAHE[a] | 0.071 | NoAHE[a] |
| (RfD$_o$ $1.4 \times 10^{-3}$) | 0.669 | NoAHE[a] | 0.666 | NoAHE[a] | 1.111 | Non-cancer-risk | 0.202 | NoAHE[a] |
| DDI | 0.001 | No risk | 0.001 | No risk | 0.002 | No risk | 0.000 | No risk |
| DIM | 0.000 | No risk | 0.000 | No risk | 0.000 | No risk | 0.000 | No risk |
| HRI | | | | | | | | |
| (RfD$_o$ $4 \times 10^{-3}$) | 0.020 | Safe | 0.020 | Safe | 0.033 | Safe | 0.006 | Safe |
| (RfD$_o$ $1.4 \times 10^{-3}$) | 0.057 | Safe | 0.057 | Safe | 0.094 | Safe | 0.017 | Safe |
| THQ | | | | | | | | |
| (RfD$_o$ $4 \times 10^{-3}$) | 0.234 | No risk | 0.233 | No risk | 0.389 | No risk | 0.071 | No risk |
| (RfD$_o$ $1.4 \times 10^{-3}$) | 0.669 | No risk | 0.667 | No risk | 1.111 | Risk | 0.202 | No risk |
| CR | 0.00001 | Low risk | 0.00001 | Low risk | 0.000013 | Low risk | 0.00000 | Low risk |

**Note**: * Units - HQ (unitless), DDI (mg day$^{-1}$), DIM (mg day$^{-1}$ body weight kg$^{-1}$), HRI (unitless), THQ ($10^{-3}$ kg$^{-1}$ person$^{-1}$), CR (unitless).
[a] NoAHE = No adverse health effects.

### 4.1. Assumption and limitations

1. This study assumed that the population consumes the specified food products grown in the designated case study site.
2. This study only focuses on the foods that are not mixed with other ingredients; however, secondary food products, such as processed and homemade potato products, chipped, fried, and roasted potatoes, which are not considered, may give rise to further Pb-exposure. Given this, cumulative exposure routes should be investigated.
3. Human exposure to Pb was calculated for only immune-competent Irish adults.

### 4.2. Comparison with similar studies

Recently, many studies looked at the risk assessment of Pb through air/dust. Moghtaderi et al. (2019) reported that the pollution load index (PLI) showed that the anthropogenic activities (traffic and combustion of fossil fuels as well as industrial activities) are primary sources of Pb contamination in school dust. The non-cancer risk of individual metals for both children and adults followed the decreasing trend of Pb > Cr > As>Ni > Cd > Cu > Zn > Co and Cr > Pb > As>Cd > Ni > Cu > Zn > Co, respectively. Therefore Pb was highlighted as a concern. The hazard index (HI) of Pb for children is very close to the safety limit. In terms of carcinogen risk, Pb levels were found to be within the cancer threshold limit. Also, Goudarzi et al. (2018) revealed that Zn and Pb were the most abundant elements among the studied $PM_{10}$-bound heavy metals, followed by Cr and Ni. The carcinogenic risks and the integral hazard quotient (HQ) of Pb in $PM_{10}$ for children and adults via inhalation and dermal exposures exceeded $1 \times 10^{-4}$ in a few areas. Megido et al. (2017) also suggest that cancer and non-cancer risk values were in the acceptable range for adults, with some exceptions; however, a greater health risk was estimated in the case of children through toxic elements (As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V, Zn) in $PM_{10}$. Four major contributing factors to heavy metal pollution has been identified, viz. traffic-related exhaust (34.47%); coal combustion (25%), the manufacture and use of metal components (25%); and the use of pesticides, fertilizers, and medical devices (14.88%) (Men et al., 2018).

The human health risk from Pb in drinking water is comparatively reported as low based on the heavy metal pollution index (HPI), heavy metal evaluation index (HEI), and water quality index (WQI) (Dippong et al., 2019). Mohammadi et al. (2018) reported that the Pb levels in Qom's Sohan (a kind of traditional Persian saffron brittle toffee) in 2015 varies from 30 to 1750 μg $kg^{-1}$. Dahmardeh Behrooz et al. (2021) found that a higher non-carcinogenic and carcinogenic risks (integrated for Cd, Cr, Co, Cu, Mn, Ni, Pb, Zn, As) were associated with the inhalation pathway in adults and children, except carcinogenic risk for children, where the ingestion route remains the most important. Also, Liu et al. (2014) reported that ingestion is the major route of exposure for road dust for both adults and children, followed by dermal exposure. The non-carcinogenic health risk resulting from exposure to the potentially toxic metals in urban trunk road dust was within the safe level based on the Hazard Index (HI), except in pollution hotspots where exposure to Pb, Cr, and Cu may be hazardous to children. In general, children are more vulnerable to this heavy metal exposure (Wang et al., 2020); therefore, these findings trigger a great future concern.

Sharafi et al. (2019a) showed that the rinsed cooking has higher efficiency for removal of Pb (42.9%) than the Kateh method (Persian stove-top rice) (26.9%). Additionally, by increasing rice soaking time from 1 to 12 h, Pb removal was increased up to 42.6%. Sharafi et al. (2019a) suggested that Tehran households cook rice by rinse method after washing and soaking the rice for 5 h. Therefore, the cooking method has also a significant impact on the final concentration of metals in food. The geographical factor also plays a vital role. Sharafi et al. (2019b) reported that the risks from As and Pb in Indian rice and Cd in Iranian rice were considerably higher than others among grown or imported rice in Iran. Pirsaheb et al. (2021) surveyed seven types of high-consumption cereals, including lentil, peas, corn, split peas, bean, rice, and wheat (totally 48 brands) from the market of Kermanshah city (Iran). They found that the total non-carcinogenic total target hazard quotient (TTHQ) of heavy metals for almost all cereals is less than acceptable level (=1), while the corresponding total carcinogenic risks (TCR) for almost all cereals is higher than the allowable limit ($=10^{-4}$). Various practical measures can be taken to reduce the amount of heavy metals in cereals, including avoiding excessive consumption of fertilizers and pesticides for cereals production, cultivating cereals in high-quality soils, and continuous monitoring of cereals in the market (Pirsaheb et al., 2021).

Huang et al. (2020) revealed that the consumption of sweet potato flesh is a lower health risk, while shoots pose a greater health risk to local people, and Cd is the main cause of the risk based on the hazard index (HI). It must be noted that the level of total Pb in the soil was 124.81 ± 5.10 mg $kg^{-1}$. Furthermore, the median, arithmetic mean, and the geometric mean of Pb level in potato was 0.21, 0.26, and 0.23 mg $kg^{-1}$, respectively (SD 13 mg $kg^{-1}$). This study evaluated the simulated mean levels of Pb in potato as 0.800 (5th percentile 0.185 and 95th percentile 1.886) mg $kg^{-1}$ where the mean level of Pb in the soil was 41.1 (SD 21.5) mg $kg^{-1}$. Therefore, as indicated in the sensitivity analysis, the bioaccumulation factor played a vital role in the elevated levels of Pb in potatoes. (Musilova et al., 2015) stated that especially mobile forms of Pb in soil (0.100–0.295 mg $kg^{-1}$), higher than the critical value (0.1 mg $kg^{-1}$ of fresh matter) based on the Commission Regulation (EC) No 1881/2006 (The European Union, 2006), represent a risk resulting in the high content of Pb in potatoes (0.244–0.855 mg $kg^{-1}$ of fresh matter). In another study (Sanaei et al., 2021), the sensitivity analysis revealed that daily ingestion rate, exposure duration, and metal concentration are the key sensitive parameters of a probabilistic model evaluating hazard index (HI). Ćwieląg-Drabek et al. (2020) also showed that the average content of Pb (0.57 mg $kg^{-1}$ fresh weight) in vegetables exceeded maximum permissible concentrations according to the European quality standards. These findings are in line with the key findings of the current research.

### 4.3. Recommendation and future work

1. Field-based tests are necessary to estimate the actual bioaccumulation factor to predict the potential metal(loid)s level in the edible portion of the crops taken up from the soil. Therefore, a periodic soil test is required where root and tubers crops are grown.
2. This study highlights the significance of limiting the Pb concentration in the agricultural soil media (51 mg $kg^{-1}$ for the conditions studied in this report) to ensure limited uptake and bioaccumulation by plants, ensuring reduced Pb human exposure through the 'food' pathway.
3. According to GSI (2020), ~90% of the soil samples in the studied region had a soil concentration of Pb below 51 mg $kg^{-1}$ (northern counties), highlighting production areas suitability for selected crops.
4. Based on this study's findings, it is recommended to conduct a similar quantitative human health risk assessment on Cd and As exposure for future work. In addition, the concentration of As in both surface and groundwater needs to be evaluated as international evidence suggests groundwater abstraction for drinking water is one of the predominant pathways for As.
5. It is crucial to identify the most suitable areas to grow certain crops that could minimise the overall bioaccumulation of metal(loid)s by crops.
6. Future studies should assess the bio-transfer of metal(loid)s through animal products where Pb levels in pastures soil (through the grass to milk or meat products) and water (through fish products) can be included.
7. Experiment-based case studies for $Bio_{acc}$ specific to Irish conditions are recommended.

8. Validation at the point of food consumption on Pb levels in food products may improve predictive modeling confidence.
9. This study proposes expanding the current study for future work looking at different toxic heavy metal(loid)s, including the health impacts on adults, the elderly, and children.
10. Levels of Pb in human blood are the ultimate criteria to evaluate Pb exposure; therefore, a health survey would improve understanding in this area.

## 5. Conclusion

Lead (Pb) concentration in topsoil varied from 14.0 to 188.0 (mean 39.7) and 7.0 to 248.4 (mean 41.1) mg $kg^{-1}$ in arable lands and pastures, respectively. The quantitative analysis revealed that the bioaccumulation factor's highest value was observed for root and tuber crops, followed by leafy vegetables. The daily mean human exposure (HE) to Pb for the general population was found to be 0.073 mg $day^{-1}$, where mean weekly exposure was calculated as 0.0065 mg kg $bw^{-1}$ $week^{-1}$. The use of biosolids (scenario S2) increases the daily HE from 0.073 to 0.121 mg $day^{-1}$ compared to the no-biosolids or baseline scenario. A sensitivity analysis highlighted that the intake of potatoes (as a highly consumed food product) is a critical input and is positively correlated with the HE followed by concentration of Pb in the arable soil, bioaccumulation factor of tubers, consumption of salad vegetables, and consumption of green vegetables. Based on six risk assessment methodologies (HQ, DDI, DIM, HRI, THQ, CR), there is a low to moderate risk posed by Pb within the system boundary of the probabilistic model. The significance of limiting Pb concentrations in the soil to 51 mg $kg^{-1}$ is highlighted, and this is in agreement with the lower permissible limit of Pb in the soil (range 50–300 mg $kg^{-1}$) recommended by the European Commission and a more refined limit of 80 mg $kg^{-1}$ recommended by the Codes of Good Practice for the Use of Biosolids in Agriculture. It is recognised that this limit may be location, crop and metal-specific, and related to baseline levels in the environment and the crops propensity to bioaccumulate the metal into edible tissue components.

## CRediT authorship contribution statement

**Rajat Nag**: Conceptualisation, Methodology, Data curation, Visualisation, Investigation, Writing - original draft. **Enda Cummins**: Conceptualisation, Supervision, Writing - review & editing.

## Declaration of competing interest

The authors declare that they have no known competing for financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.151168.

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