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Pesticide pollution associations with riverine invertebrate communities in England

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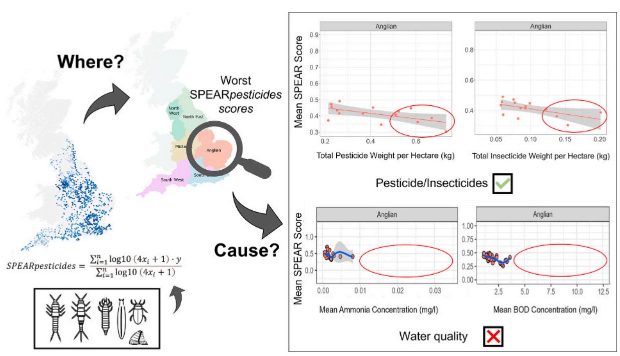
HIGHLIGHTS

- Pesticide use and water quality explained regional SPEAR$_{pesticides}$ scores.
- Regional SPEAR$_{pesticides}$ scores predict pesticide threat to riverine invertebrates.
- Pesticide impact to English riverine invertebrates differs regionally.
- Pesticide threat to riverine invertebrates is greatest in the Anglian region.

GRAPHICAL ABSTRACT



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ABSTRACT

Globally freshwater biodiversity has experienced major decline and chemical pollutants are believed to have played a significant role in this decline, but this has not been well quantified for most riverine invertebrate populations. Here we applied a biogeographically independent trait-based bioindicator, *SPEAR$_{pesticides}$* across sites across five regions (Northern, Midlands and Western, Anglian, Southeast, and Southwest) in England to investigate for associations specifically between pesticide use/pollution and riverine invertebrate communities over a 55-year period (1965–2019). Both spatially and temporally post-1990, the Anglian and Thames regions consistently showed the lowest *SPEAR$_{pesticides}$* scores, illustrating the presence of fewer pesticide sensitive species. The Anglian region had the highest pesticide use compared to all other regions from 1990 to 2018 and there were negative relationships between the level of pesticide/insecticide use and the regional *SPEAR$_{pesticides}$* score. Biochemical Oxygen Demand and ammonia, as measures of general water quality, were also negatively correlated with the *SPEAR$_{pesticides}$* scores across the regions, but these factors were not the driver for the lower *SPEAR$_{pesticides}$* scores seen in the Anglian region. Based on *SPEAR$_{pesticides}$* scores, riverine invertebrate communities

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in England have been most impacted in the Anglian region and we evidence chronic insecticide exposure is likely a significant factor in shaping the status of those invertebrate communities.

1. Introduction

Globally, since the 1970s, freshwater systems' have experienced the greatest biodiversity decline compared to other planetary environments (Living Planet Report, 2020). The UK is no exception with major declines especially in freshwater biodiversity, most notably prior to the 1990s (Outhwaite et al., 2020). Since the 1990s with more rigorous water quality regulations, there has been a general increase (recovery) in freshwater invertebrate biodiversity in areas of the UK, however some taxa, particularly freshwater molluscs appear still to be in general decline (Outhwaite et al., 2020; van Klink et al., 2020). Furthermore with only 14 % of English rivers are classified as having 'good ecological status' as defined by criteria according to the EU Water Framework Directive, our freshwater taxa are still facing pressures which are likely hindering population recovery (The Rivers Trust, 2021; EU WFD). Against this, the UK governments 25 year Environment Plan is committed to achieving 'waters richer in plants and wildlife' and substantially reducing the levels of harmful chemicals entering the environment (including those from agriculture) (Defra, 2021a).

Factors associated with declines in freshwater invertebrate biodiversity include habitat alteration, climate change, invasive species and pollution (Aldridge et al., 2004; Maynard and Lane, 2012; Jourdan et al., 2018). Chemical pollution sources into UK rivers are wide ranging and include wastewaters, mining waters, industry discharge, agricultural run-off, and urban and transport run-off. Different UK regions are dominated more heavily by certain pollution sources, for example, the Anglian region by agriculture, whereas the Midlands and Northern England by mine-waters (Environment Agency, 2008a; Bernick et al., 2017; Smith et al., 2018). Identifying associations between specific chemical pollutants/classes of pollutants with changes in populations and communities of specific freshwater invertebrate taxa however is challenging, not least because of the very wide range of chemicals and their mixtures discharged into surface freshwaters (Dowson et al., 1996; Beasley and Kneale, 2002; Hirst et al., 2002; Smith et al., 2010). Of these chemicals, pesticides, including insecticides, molluscicides, and acaricides, used for decades, are designed to target and kill terrestrial invertebrate pests and their use has been increasing by geographical area (24 % increase in hectares sprayed between 2000 and 2016 in the UK) (Friends of the Earth Policy, 2019; Whelan et al., 2022). Since the 1990s many of the pesticides considered as 'high-risk' substances to biota have been replaced, following the introduction of the EU's more restrictive registration process (EC 91/414 and subsequently EU Regulation 1107/2009), and attempts have been made to design pesticides with more specific biological target sites (e.g. nicotinic acetylcholine receptors, nAChRs) (Umetsu and Shirai, 2020). Nevertheless, most of these compounds have still been shown to have unintended effects on non-target terrestrial invertebrates, such as honeybees (Pettis et al., 2013; Di Prisco et al., 2013; Thompson et al., 2014; Woodcock et al., 2016, 2017) and bumblebees (Feltham et al., 2014; Rundlöf et al., 2015; Baron et al., 2017; Muth and Leonard, 2019; Siviter et al., 2021). Information regarding pesticides impact on freshwater invertebrates has been less forthcoming than for terrestrial invertebrates. Nevertheless, across mainland Europe there are cases showing adverse impacts of acute spills and run-off events of pesticides on freshwater invertebrate populations (Schulz and Liess, 1999; Werner et al., 2000; Mugni et al., 2011; Wurzel et al., 2020). Evidence for chronic exposure effects of pesticide on freshwater invertebrate communities, however, is relatively sparse (Courtemanch and Gibbs, 1980; Van Dijk et al., 2013).

Across parts of Europe, including in Germany, France, Finland, Iberia, and in other countries globally, including in Kenya, Australia, Brazil and Argentine, a bioindicator known as $SPEAR_{pesticides}$ has been applied to assess the impact of pesticides on freshwater invertebrate communities (Beketov et al., 2009; Schäfer et al., 2011; Kuzmanović et al., 2016; Hunt et al., 2017a; Hunt et al., 2017b; Ganatra et al., 2021; Liess et al., 2021). This index determines the proportion of pesticide sensitive invertebrate species in a community through a trait-based approach (Liess and Von Der Ohe, 2005) and provides an indication of the relative pesticide pressure. There has been little application of $SPEAR_{pesticides}$ in the UK, despite the fact that the majority of UK freshwater invertebrate species have been added to the trait database needed to perform the $SPEAR_{pesticides}$ calculation (Environment Agency, 2008b).

Here, the primary focus was to better establish relationships between pesticides and impact on riverine macroinvertebrate populations across regions of England. We employed the $SPEAR_{pesticides}$ bioindicator to investigate where in England freshwater invertebrates have been most affected by pesticide pollution. Eastern England, particularly the Anglian region, has the greatest quantity of cropped land (tilled land) and crop farming (particularly arable) that uses a greater tonnage of pesticides compared with pastoral farming (Robinson and Sutherland, 2002; Smith et al., 2018). Thus, we hypothesised that the Eastern regions (particularly Anglian) would have lower $SPEAR_{pesticides}$ scores both spatially and temporally compared with the other regions studied (Northern, Midlands and Western, Southeast and Southwest). We also sought to assess for possible relationships between regional $SPEAR_{pesticides}$ scores and pesticide/insecticide use. Further, given that $SPEAR_{pesticides}$ scores can be influenced by general organic pollution, we used two of the most routine measures of general water quality (ammonia and biochemical oxygen demand (BOD)), to assess this influence across the study regions. A further purpose of our work was to identify areas of England where the riverine invertebrate populations are most strongly influenced by pesticide and insecticide use and in turn enable future field-based studies to be more directed for understanding relationships between chronic exposure to specific classes of pesticide and population level associations with freshwater invertebrates.

2. Material and methods

2.1. Pesticide usage data

Pesticide usage data for all crop types (arable; bulb and flower; fodder, forage and grassland; protected; hardy nursery stock; hops; mushrooms; orchard; soft fruit; outdoor vegetable) in England was collected from The Food and Environment Research Agency (FERA) Pesticide Usage Surveys (PUS, the most thorough regional data for crop pesticide use in England) for the years spanning 1990–2018. The PUS dataset provided the predicted quantity of pesticides used in the five study regions, Anglian, Southeast, Southwest, Midlands and Northern (see Fig. 1). The methods used to predict total pesticide tonnage (kg) are detailed briefly in the SI and reported by Thomas (2002). It is accepted that values obtained from the PUS data could over-or under-estimate pesticide usage as individual farmers may choose which pesticides to apply and may not adhere to recommended application rates (for a discussion on this, see FERA, 2021).

The pesticide usages are included for arable, orchard, soft fruit and hop crops (see table S1 for details). Total yearly pesticide usage data were available every two years only (e.g. 1990, 1992, 1994 etc.,) as these were the only years which included arable farming (cereals) which account for 86–90 % of crops grown in the UK and thus the mass proportion of pesticide (insecticide use) (Thomas, 2002).

From the total predicted pesticide usage data, chemicals belonging to the class insecticides were filtered to also determine the total predicted tonnage of insecticides used each year per region (for the insecticide list, see table S2). Most studies have demonstrated that insecticides are more harmful to invertebrate communities compared with other major pesticide groups (e.g. herbicides, fungicides and biocides) (Münze et al., 2015; Ganatra et al., 2021).

2.2. The $SPEAR_{pesticides}$ bioindicator

$SPEAR_{pesticides}$ is a trait based bioindicator used to assess the potential impact of pesticide pollution on riverine invertebrate communities. The specific details of $SPEAR_{pesticides}$ metric are detailed in the SI and in Liess and Von Der Ohe (2005).

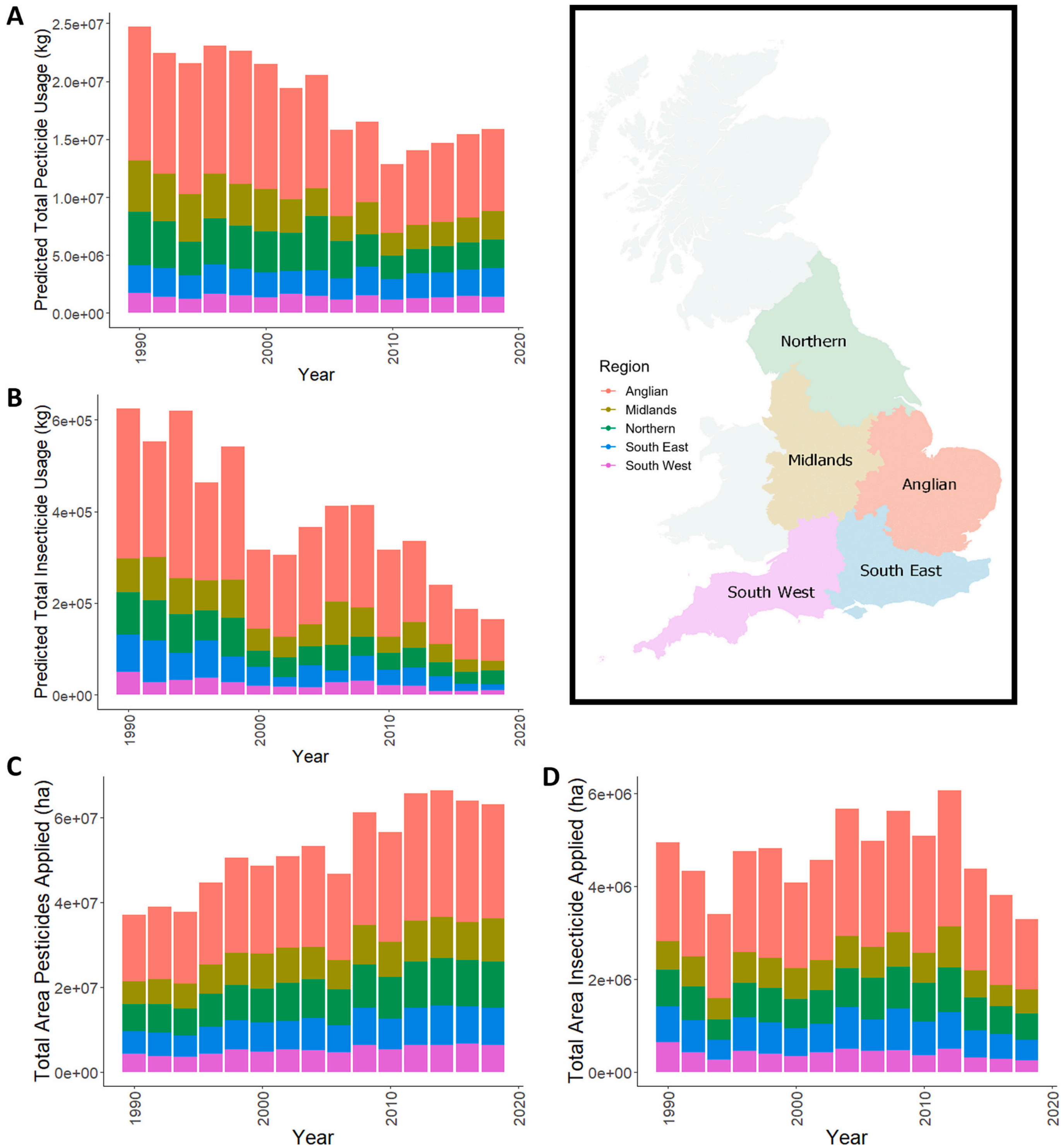

Fig. 1. Bar plots showing the predicted amount (kg) of pesticide (A), Insecticide (B) used and total areas of application for pesticides (C) and insecticides (D) per year/region. We emphasise here that some of the more recently developed pesticides have become more innately (and selectively) toxic to certain target invertebrates, in turn requiring lower application rates to achieve the same effect as achieved with historical pesticides, and therefore the tonnage of pesticides applied as a whole over time (years) is not necessarily directly representative of overall toxic pressure (Umetsu and Shirai, 2020).

The $SPEAR_{pesticides}$ metric is the proportion of species which are defined as *at risk* or *not at risk* in a riverine sample and is calculated as follows:

$$SPEARpesticides = \frac{\sum_{i=1}^{n} log10\ (4x_i + 1) \bullet y}{\sum_{i=1}^{n} log10\ (4x_i + 1)}$$

where: N, is the number of taxa, *i* is the taxon, χi is the abundance of the taxon *(i)* and if taxon *i* is classed as SPEAR (a species at risk) the *y* is given a numerical score of one, if not SPEAR, *y* is given as zero (Environment Agency, 2008b). Species abundance data are log-transformed ($\log(4\times + 1)$) (Knillmann et al., 2018) so that abundant taxa are not disproportionally represented and meaning the $SPEAR_{pesticides}$ score more equally represents both the population density and incidence of vulnerable taxa (Knillmann et al., 2018; Ganatra et al., 2021).

The outcome of $SPEAR_{pesticides}$ metric is a number based on the proportion of pesticide sensitive and pesticide insensitive species present in a community sampled (Knillmann et al., 2018) with five ecological status classes associated with a $SPEAR_{pesticides}$ score: (1) $\geq$0 - <2.0 is classed as very poor, very few pesticide sensitive species and likely a major impact of pesticides on the invertebrate community; (2) $\geq$0.2 - <0.4 is classed as a poor status of the macroinvertebrate community; (3) $\geq$0.4 - <0.6 is classed as moderate status; (4) $\geq$0.6 - <0.8 is classed as good and finally (5) $\geq$0.8 is classified as high status, where there is an abundance of pesticide invertebrate sensitive species and likely negligible pesticide pressure on the invertebrate community (Knillmann et al., 2018).

The SPEAR database was initially designed for studies on German rivers (Environment Agency, 2008b; Liess et al., 2008) but contains UK species for its use on UK rivers. Furthermore, the $SPEAR_{pesticides}$ is biogeographically independent allowing for comparisons across geographical regions which may differ in altitude, temperature, rainfall, topology, geology (i.e. is not limited by differences in species that occur across different regions; Schäfer et al., 2007).

2.3. Data collection and application of the $SPEAR_{pesticides}$ bioindicator

The freshwater macroinvertebrate data collected from English rivers by the UK Environment Agency is found in the BIOSYS database (https://environment.data.gov.uk/ecology/explorer/). The ChemPop database contains data for English rivers sites from the nearest chemical measuring site on the same river (coming from the Environment Agency (WIMIS database), sourced from the Centre for Ecology and Hydrology's ChemPop. ChemPop macroinvertebrate data and site abiotic factors were available for approximately 200 sites for each geographical region included in this study (Northern, Midlands and Western, Anglian, Southeast, and Southwest) for the period between 1965 and 2019. The sampling sites in ChemPop had been pre-selected from the UK Environment Agency WIMIS database based on their frequency of sampling over the monitoring period and excluded sites only sporadically sampled (fewer than 29 samples) over the 55-year period. The defined geographical regional boundaries in ChemPop are dominated by different farming practices and thus pesticide use across regions. Crop farming, particularly arable, uses a greater quantity of pesticide than livestock farming, and for crops pesticides are applied directly to the field (Robinson and Sutherland, 2002). The majority of regions include arable farming, particularly the Anglian and Midlands regions which are dominated by cereal and general (arable and horticulture crops) cropping (Smith et al., 2018). The Northeast region is also dominated by cereal and general cropping, but pastoral farming is also prominent (Smith et al., 2018). The Southeast contains a mixture of farming types with cropping practices including cereal, general horticulture (e.g. fruit, vegetables, flowers, bulbs, mushrooms and hardy nursery stock) in addition to pastoral farming (Smith et al., 2018). The Southwest and Northwest regions of England are dominated by pastoral farming, and in some areas arable farming is absent (Smith et al., 2018).

The macroinvertebrate data were used to calculate a $SPEAR_{pesticides}$ score for each sample collected at each site using the software INDICATE version 2021 (https://www.systemecology.de/indicate) (Liess and Von Der Ohe, 2005). Macroinvertebrate species level information was used to calculate $SPEAR_{pesticides}$ scores for a total of 1519 sites across England. All riverine sites were included in the analyses, even for cases where there were fewer than 2 samples per year or fewer than 10 species per sample. This is because the standardised sampling methods employed by the UK Environment Agency means that even small samples of a few macroinvertebrates species are likely to be representative of the sites' invertebrate community (i.e. fewer than 10 species in a sample demonstrates a poor macroinvertebrate community; Schäfer et al., 2011). As the $SPEAR_{pesticides}$ bioindicator is designed for use on small to medium-sized rivers and has not been validated for use on large rivers (Liess and Von Der Ohe, 2005) rivers >40 m in width were removed from our analyses (Münze et al., 2015). Across all regions macroinvertebrate samples containing species that were not linked to traits in the available database made up a small portion (less than ~15 %) of the macroinvertebrate species sampled (see fig. S1).

A $SPEAR_{pesticides}$ score was calculated first for every sample of invertebrates collected by the Environment Agency and these scores were then aggregated (per site), to produce an average $SPEAR_{pesticides}$ score for every site monitored across England over the 55-year period. The purpose of this approach was to identify hotspots across England, where there were lower average $SPEAR_{pesticides}$ scores.

Next, all $SPEAR_{pesticides}$ scores for all samples were aggregated by site and year (avoiding pseudoreplication), to produce the mean $SPEAR_{pesticides}$ score for site per year. The mean $SPEAR_{pesticides}$ scores for all sites were then aggregated by region, to compare the regional difference in mean annual site $SPEAR_{pesticides}$ scores. To identify the region/s with the lowest $SPEAR_{pesticides}$ scores over time, the aggregated regional $SPEAR_{pesticides}$ scores were used to produce a mean SPEARpesticide score for each region for each year. This process was then repeated using $SPEAR_{pesticides}$ scores of samples collected during May–July only.

2.4. Pesticide use and $SPEAR_{pesticides}$

We investigated the possible influence of regional crop pesticide/insecticide use on $SPEAR_{pesticides}$ scores, using annual regional pesticide/insecticide use (kg/ha) and mean annual $SPEAR_{pesticides}$ scores. PUS data does not include usage statistics for livestock rearing and thus was outside of the scope of this analysis. We also assessed the possible influence of regional crop pesticide/insecticide use on $SPEAR_{pesticides}$ scores for invertebrate samples collected for the period between May–July. This is because this is the time when most pesticides are applied and the strongest impact of pesticides on freshwater macroinvertebrate communities are most likely to occur (Environment Agency, 2008b). Regional $SPEAR_{pesticides}$ scores were filtered to include only years for where matching pesticide/insecticide usage data were available (2016, 2018, etc.). A complication in these analysis was that the regional boundaries defined by the datasets (ChemPop, $SPEAR_{pesticides}$ and PUS, pesticide/insecticide data) were not identical to those for the $SPEAR_{pesticides}$ score and pesticide usage (see maps in Figs. 1 and 2, see also SI S1 for more information). Thus, association analyses between pesticide/insecticide use (kg/ha) and $SPEAR_{pesticides}$ data could be performed for the Anglian, Southeast and Southwest regions only. Whilst pesticide/insecticide toxicity may have changed over time as some chemicals developed and applied have become more effective (innately more toxic to targeted pests species) and in turn requiring lower doses/field application rates (Umetsu and Shirai, 2020), pesticide/insecticide usage still provides a proxy for pesticide/insecticide pressure in a river system albeit with the assumption that higher levels of pesticide application equate with a greater chance of pesticide run-off into rivers.

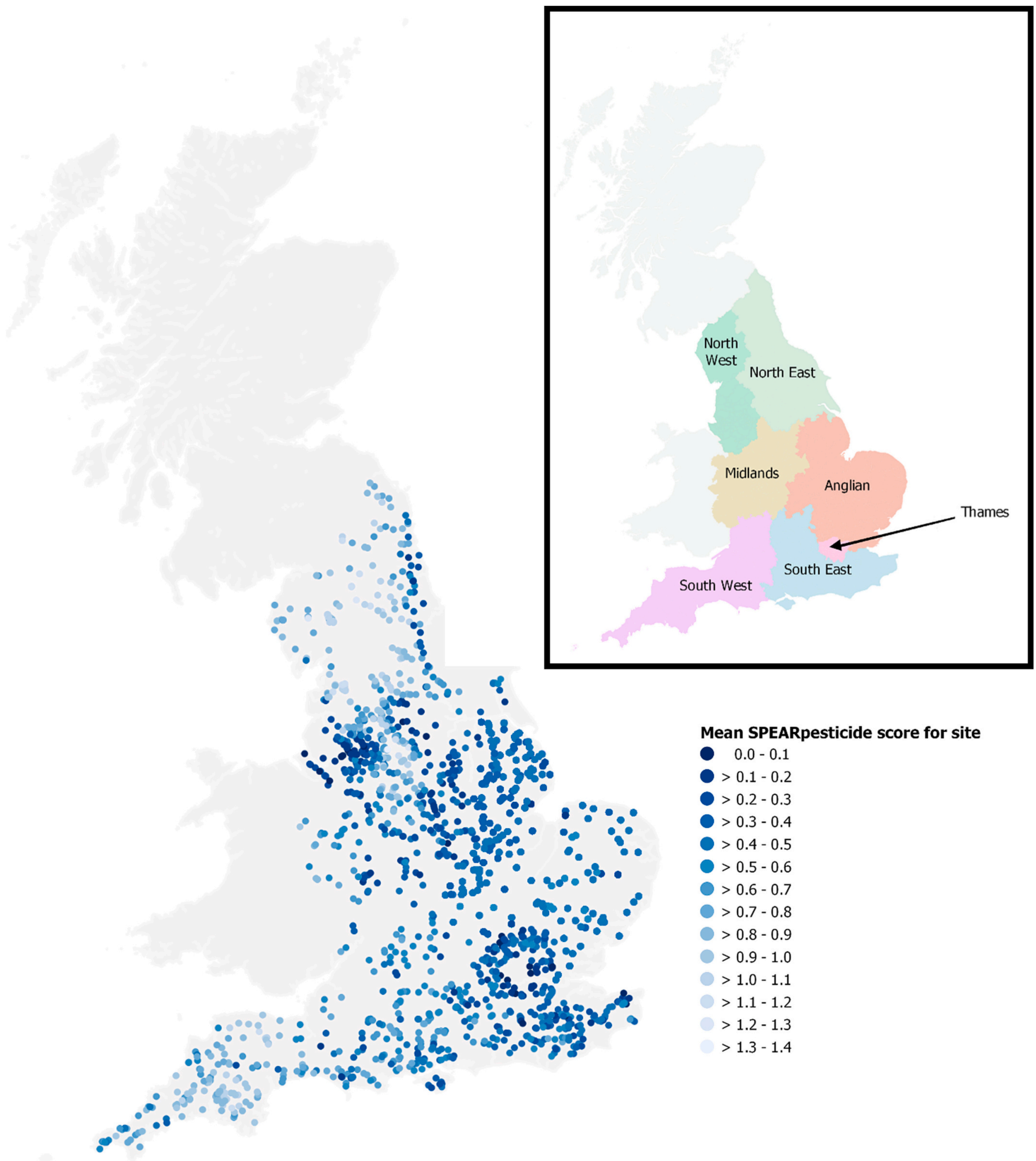

Fig. 2. Geographical map illustrating the mean yearly $SPEAR_{pesticides}$ score per site across all years monitored. Increasing blue colour spot intensity indicates a decreasing $SPEAR_{pesticides}$.

2.5. Associations between water quality parameters and $SPEAR_{pesticides}$ and pesticide use

$SPEAR_{pesticides}$ can also be affected by organic pollution as well as pesticides (Malherbe et al., 2018). To assess whether organic pollution was driving $SPEAR_{pesticides}$ scores across the study regions we assessed these scores against two of the most widely used measures of general water quality, Biochemical Oxygen demand (BOD) and ammonia (which largely derives from fertiliser use in England's arable landscapes) (Natural England, 2022). High BOD concentrations can indicate reduced oxygen availability and this negatively impacts riverine invertebrate communities and causes declines in taxa richness (Vigiak et al., 2019).

Similarly high ammonia concentrations often coincide with reduced taxa richness (Reif, 2002).

To investigate for possible associations between selected water quality measures and $SPEAR_{pesticides}$ scores regionally, BOD and ammonia (mg/l) data in the ChemPop dataset collected from sites between May – July were aggregated for each year (Pickwell et al., 2022). The mean BOD and ammonia concentrations were then calculated across all sites for each year, to provide a regional yearly average. Only the sites with $SPEAR_{pesticides}$ data for May–July which had BOD and ammonia concentrations information were included in this analysis. In cases where several water quality samples were taken for any given day the average of these samples was used to avoid pseudoreplication. Where BOD and ammonia measurements were below the limit of detection, the limit of detection was used as the measured value in these cases. To assess if poor water quality (as defined by BOD and ammonia measurements) correlated with pesticide/insecticide use (kg/ha), the average regional ammonia and BOD concentrations (mg/l) were compared (separately) with pesticide and insecticide (kg/ha) per year for samples collected during May – July.

2.6. Statistical analysis

We used R Core Team (2020) version 4.2.1 to conduct all statistical analyses, and QGIS (2022) version 3.10.13 for regional mapping. We used the *R* packages *lme4* (Bates et al., 2015) to fit mixed effects models, *arm* (Gelman et al., 2022) to generate posterior intervals of effect sizes/parameter values, *DHARMa* (Hartig, 2022) to check model fit and for spatial autocorrelation, *glmmTMB* (Brooks, 2023) to address spatial autocorrelation and ggplot2 (Wickham, 2016) to produce graphs of raw data and model estimates.

We performed model selection using an information theoretic approach to rank models based on their support in the data using AICc. We considered all models within Δ6 AICc unit of the top model to have equal support. All code and analyses required to reproduce these analyses are provided online at (https://github.com/ImogenPW/SPEARpesticide-code.git). To examine variation in $SPEAR_{pesticides}$ score at the landscape scale, we fitted a general linear mixed effects model (GLMM) with $SPEAR_{pesticides}$ score as a response, and random intercepts of sampling site nested within region, and a Gaussian error structure. This allows us to estimate the relative amount of variation partitioned i) among sites with regions and ii) among regions. This dataset contained 14,976 observations from 7 regions and 1481 sites. To examine the relationship between $SPEAR_{pesticides}$ score and pesticide use, we fitted a GLMM with $SPEAR_{pesticides}$ as the response, the interaction between region and pesticide use (kg/ha) as fixed effects and a random intercept for year. For number of observations per year see fig. S5b. To examine the relationship between $SPEAR_{pesticides}$ score and insecticide use, we fitted a GLMM with $SPEAR_{pesticides}$ as the response, and region and insecticide use (kg/ha) as fixed effects with a random intercept for year. For number of observations per year see fig. S5b. To examine the relationship between $SPEAR_{pesticides}$ score and ammonia, we fitted a GLMM with $SPEAR_{pesticides}$ as the response, the interaction between region and ammonia (mg/l) as fixed effects and a random intercept for year. For number of observations per year see fig. S6. To examine the relationship between $SPEAR_{pesticides}$ score and BOD, we fitted a GLMM with $SPEAR_{pesticides}$ as the response, the interaction between region and BOD (mg/l) as fixed effects and a random intercept for year. For number of observations per year see fig. S7. Spatial autocorrelation assessed using Moran's Index test and accounted by included spatial random effect to the models.

Finally, Kendall's rank correlation test was used to test for collinearity between yearly pesticide/insecticide use and mean of water quality parameters (BOD and ammonia) per region.

3. Results

3.1. Pesticide and insecticide application across the study regions

Between 1990 and 2018, the total crop area (ha) treated with pesticides increased by 71 % in the Anglian region, by 97 % in the Midlands, 66 % in the Northern region, 66 % in the Southeast, and 50 % in the Southwest. Pesticides use on crops in England has, however, changed significantly over this time, with various formulations banned, new (generally more targeted) pesticide introductions and changes in the number of pesticide applications (Hillocks, 2012). Contrasting with an increase in the areas of land treated with pesticides, across all the regions studied there has been a general decline in overall quantity of pesticides used from the 1990s; in Anglian a 38 % decline, in the Midlands a 44 % decline, in the Northern region a 46 % decline, in the Southwest a 18 % decline (Fig. 1). In the Southeast there has been little change in overall pesticide use over this time (an apparent 0.5 % reduction). There has also been a general reduction in the use of insecticide between 1990 and 2018, in Anglian by 72 %, in the Midlands by 71 %, in the Northern region by 66 %, in the Southeast by 85 %, and in the Southwest by 80 %. From 1990 to 2018 there were the following reductions in area of the area of land treated with insecticides; Anglian −29 %, Midlands −17 %, Northern −29 %, Southeast −43 %, Southwest −17 %. Overall, for all study years the Anglian region consistently had higher levels of pesticide and insecticide use and greater areas of land application compared with the other study regions (Fig. 1). In turn the Anglian regions' riverine systems are likely to have received the greatest pressure from pesticides applied to the land through crop application.

3.2. Regional areas with invertebrate communities most (likely to be) impacted by pesticide pollution

Across the riverine sites monitored, lower mean $SPEAR_{pesticides}$ scores ($SPEAR_{pesticides}$ score < 0.6), are predominately found in eastern regions of England with few hotspots also in the Midlands and the Northeast (Fig. 2). Whilst variation in $SPEAR_{pesticides}$ scores is largely explained by site, region also show a considerable influence, shown in table S12.

The mean $SPEAR_{pesticides}$ score for each region varied over the study years, with a general increase in $SPEAR_{pesticides}$ scores over time, indicating the increasing presence of more pesticide sensitive species (Fig. 3). The lowest mean $SPEAR_{pesticides}$ scores occurred in the Northwest region of England in the pre-1990s (see table S5). Post-1990 the Thames and Anglian region had the lowest mean $SPEAR_{pesticides}$ scores, and this was independent of the time of year the samples were taken (whole year versus just May–July). These analyses suggests that, based on the available monitoring data, the Thames and Anglian regions contained less pesticide sensitive taxa (post-1990) compared to the other geographical regions, particularly the South West.

There was a negative relationship between annual pesticide/insecticide use (kg/ha) and annual mean $SPEAR_{pesticides}$ score across all regions (see Fig. 4 and table S13); i.e. the more pesticide/insecticide used, the lower the regional $SPEAR_{pesticides}$ score). Interestingly, the impact of pesticide/insecticide use on $SPEAR_{pesticides}$ score differs across regions, with the greatest influence in the Southeast and Southwest and the weakest influence seen in the Anglian region (see estimates in table S13). However, the Anglian region has experienced the greatest pesticide and insecticide usage (kg/ha) which is far above (>1.5-fold) those found in other regions. Furthermore, for similar levels of usage (kg/ha) of both pesticide and insecticide across regions, the Anglian region consistently showed lower $SPEAR_{pesticides}$ scores, illustrating invertebrate populations in the Anglian region are more degraded by pesticides.

3.3. Water quality and $SPEAR_{pesticides}$ scores

The relationship between measures of the selected water quality parameters (ammonia and BOD) and $SPEAR_{pesticides}$ scores differed

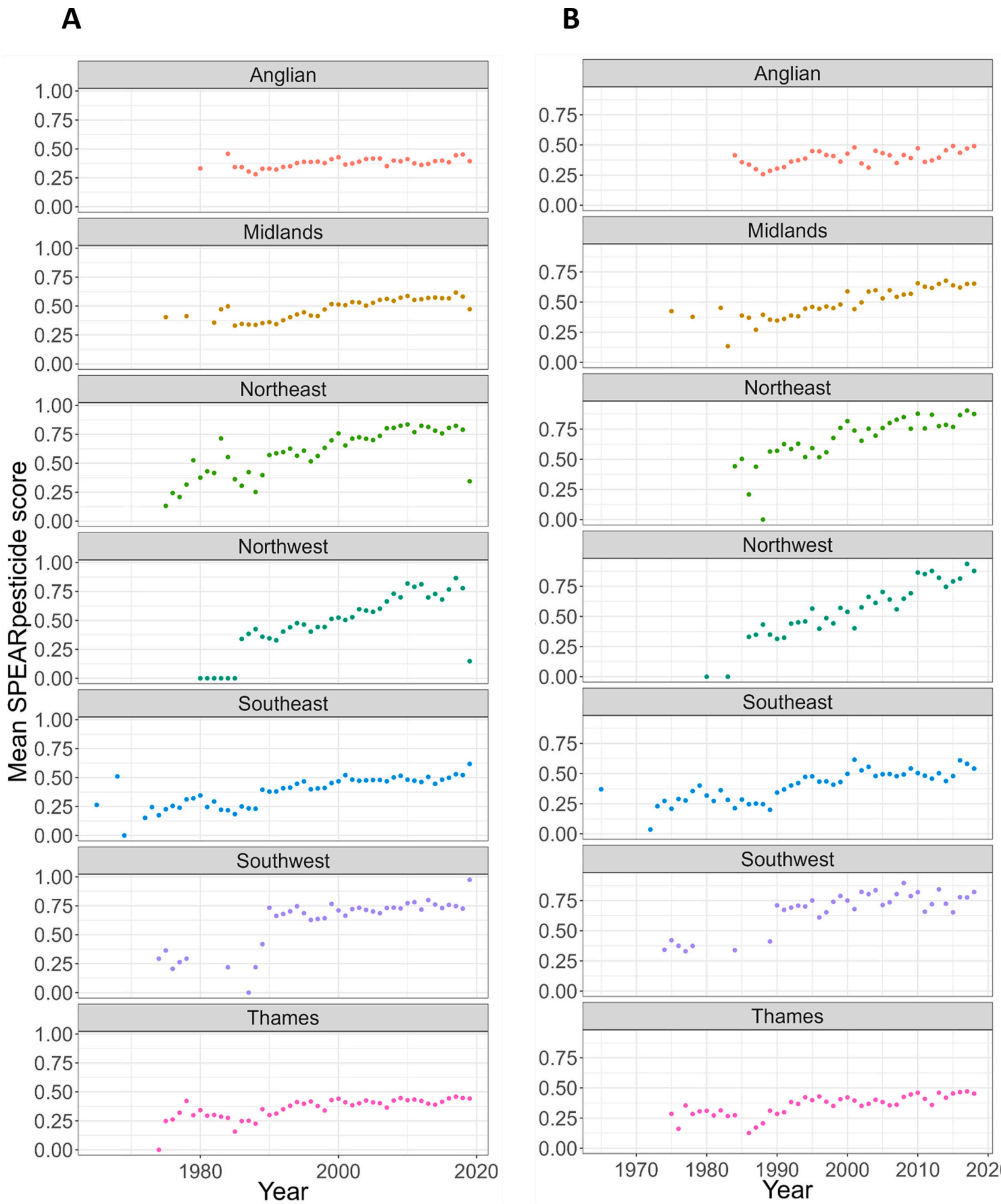

Fig. 3. Scatterplots showing the change in mean site $SPEAR_{pesticides}$ scores per region over 55 years (A) data from all months and (B) data from May – July (see Fig. 2 for regional boundaries).

between regions. For ammonia all regions except the Southwest there was a negative relationship between ammonia concentration (mg/l) and $SPEAR_{pesticides}$ score (i.e higher ammonia concentration associated with lower $SPEAR_{pesticides}$ scores; see table S14, see Fig. 5). For the Southwest on the other hand, there was a positive association between ammonia concentration and $SPEAR_{pesticides}$ (see table S14 and Fig. 5). Excluding the Southwest, the greatest effect of ammonia on $SPEAR_{pesticides}$ occurred for the Northeast, followed by Thames, then the Southeast, Northwest, Midlands, and lastly, the Anglian region (table S14).

All regions except for the Northeast showed a negative relationship between BOD (mg/l) and $SPEAR_{pesticides}$ scores (see Fig. 5 and table S14). In the Northeast region $SPEAR_{pesticides}$ scores were low at both low and high BOD concentrations (see table S14 and Fig. 5). The strongest effect of BOD on $SPEAR_{pesticides}$ score was found for the Southwest region, followed by the Thames, then Southeast, Midlands, Northwest, Northeast and finally the Anglian (table S14).

3.4. Pesticide use and water quality

Higher pesticide and insecticide use (kg/ha) were generally associated with higher levels of ammonia and BOD (see Fig. 6). There was a significant positive correlation between yearly mean ammonia

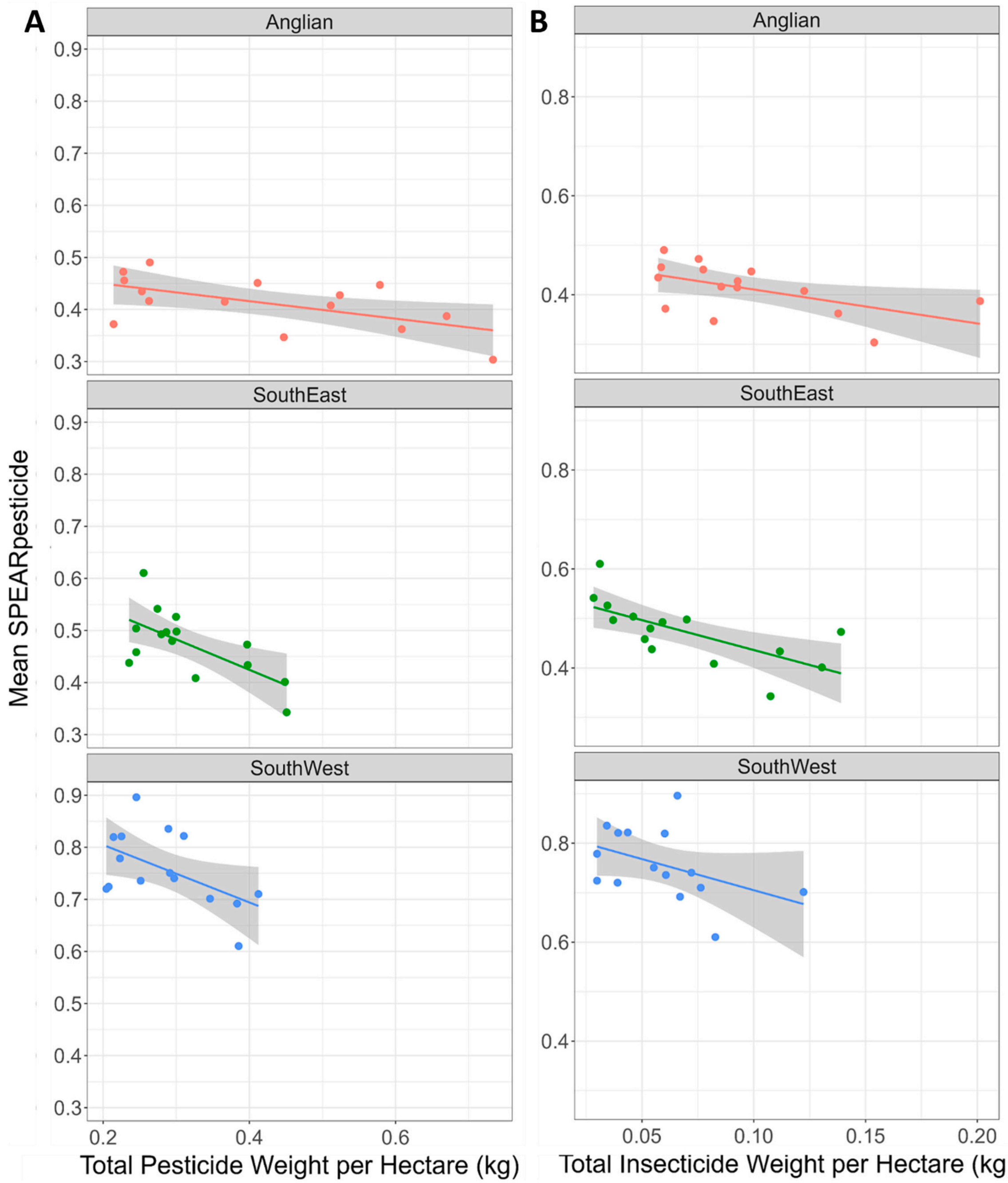

Fig. 4. Mean annual $SPEAR_{pesticides}$ score for all sites sampled (between May – July for regions Anglian, South East and South West) compared to the predicted pesticide usage for the same year (1990 to 2018) for the study regions; A – Total pesticide usage; B – Insecticide usage. Grey shaded area shows 95 % confidence intervals along with linear regression lines (See table S13 for model estimates).

concentration (mg/l) and pesticide use (kg/ha) for the Anglian and Southeast regions, but this was not statistically significant for the Southwest region (table S15). There was a significant positive relationship between mean annual BOD and pesticide use for the Anglian and Southwest regions, but not for the Southeast. There were significant positive correlations also between yearly mean ammonia concentration (mg/l) and total insecticide use, for the Anglian and Southeast regions, but not for the Southwest region (table S15). There were also significant positive correlations between yearly mean BOD and total insecticide use for the Southwest and Anglian regions, but not for the Southeast.

4. Discussion

Overall, we show there has been a general improvement in $SPEAR_{pesticides}$ scores across all the regions studied since 1990 albeit these were starting from a relatively low baseline due to major historical pollution. Further, we show that pesticide, BOD and ammonia show associations with $SPEAR_{pesticides}$ scores across the different regions. We show that high rates of pesticide/insecticide application post-1990 are associated with the lowest annual average $SPEAR_{pesticides}$ scores (i.e., fewer pesticide sensitive invertebrate species) for all regions studied except Thames, with the Anglian region most heavily impacted. Lower $SPEAR_{pesticides}$ scores in the Thames region are likely a consequence of factors that include high organic pollution and general poor water quality as a

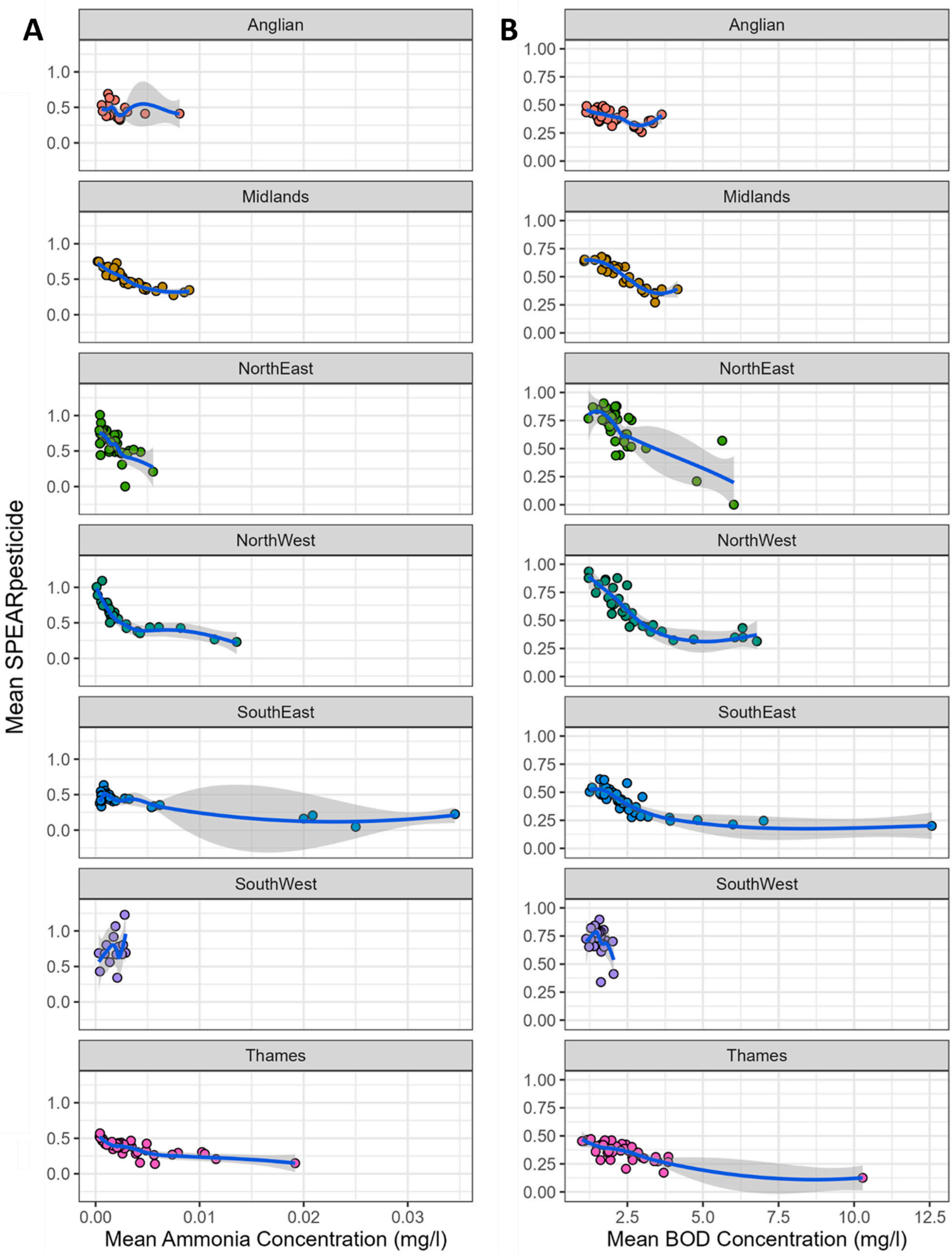

Fig. 5. Relationships between mean $SPEAR_{pesticides}$ and water quality parameters for the study regions; A, ammonia; B, BOD. Smoothing lines shown in blue along with 95 % confidence intervals shown by grey shaded area. The lines of best fit are shown for visual aid and were not part of the models applied (see Section 2.6). See table S14 for model estimates.

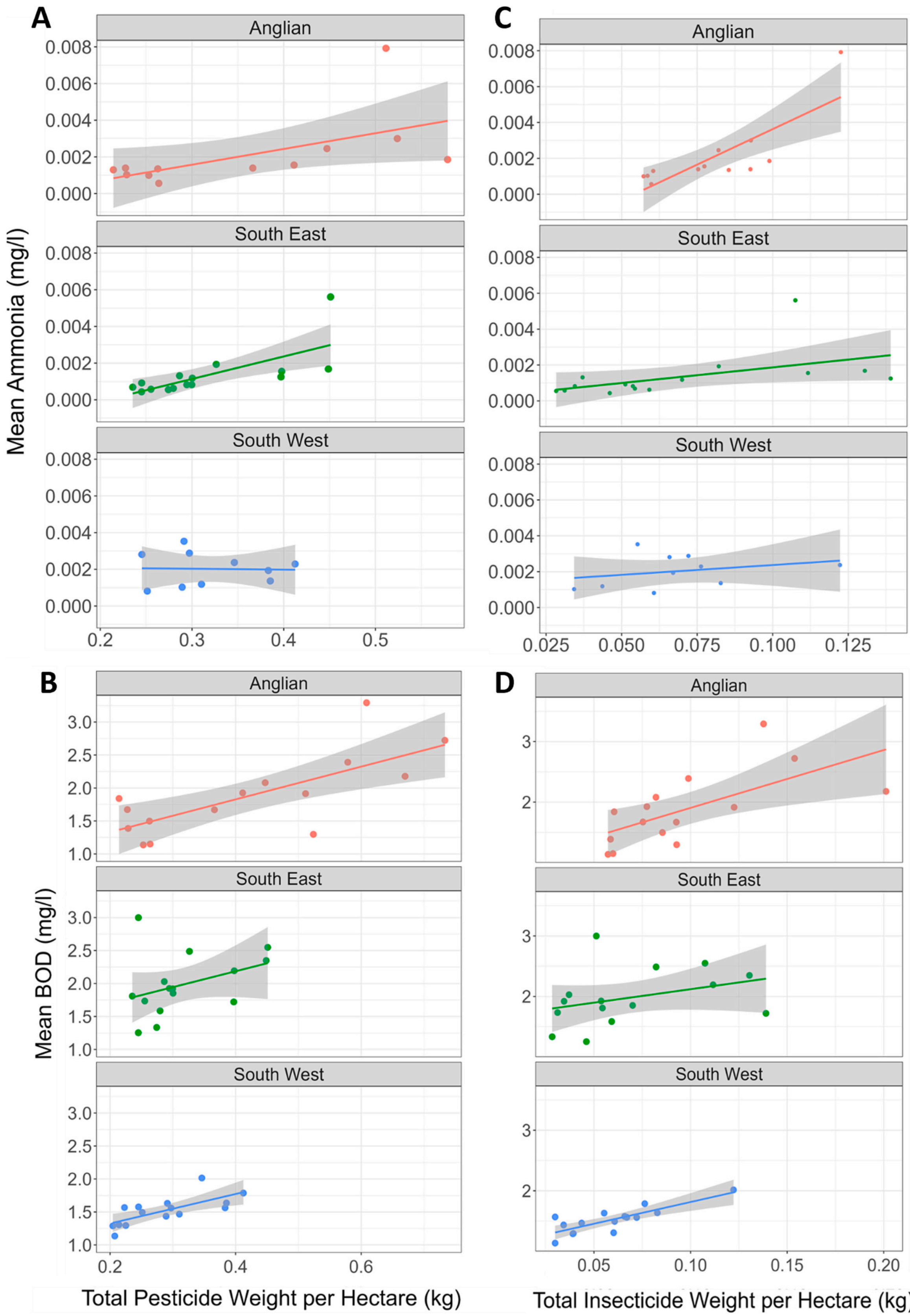

Fig. 6. Relationships between total pesticide and insecticide use and measures of water quality for the different study regions; A, ammonia vs pesticide use; B, BOD vs pesticide use; C, ammonia vs insecticide use, D, BOD vs insecticide use. Grey shaded area shows 95 % confidence intervals along with linear regression lines. See table S15 for model estimates.

consequence of sewer infrastructure problems and road run-off and temperature from urbanisation, rather than high pesticide use (Greater London Authority, 2018).

Between 1990 and 2018, the greatest quantity of pesticides and insecticides (kg) was applied to crops across the largest regional land (ha) area in the Anglian region and this was associated with lower $SPEAR_{pesticides}$ scores compared with other regions with similar applications rates (kg/ha) of pesticide/insecticides. This finding support previous studies across regions in mainland Europe reporting a negative relationship between pesticide/insecticide pollution and $SPEAR_{pesticides}$ score (Schulz and Liess, 1999; Liess and Von Der Ohe, 2005; Hunt et al., 2017b; Ganatra et al., 2021). Our analyses identifies the Anglian region as of greatest concern for potential crop pesticide (insecticide) pollution impact on riverine invertebrate communities in England.

The lower $SPEAR_{pesticides}$ scores in the Anglian region at the outset of this study period likely reflects degraded invertebrate communities from the beginning of the assessment period as a result of past chemical pressures in this region (Langford et al., 2009). The intensity of historical agricultural farming in Anglia dating back to 1985 particularly supports the likely high historical chemical (pesticide) pressure when compared to other regions (Baker et al., 2019). Furthermore, we observed that in the Southeast, a region with poorer general water quality (as assessed by higher ammonia/BOD concentrations) compared with Anglian, there were higher annual $SPEAR_{pesticides}$ scores (for the period 1990–2018; Fig. 3).

4.1. Regional pesticide and insecticide use

The Anglian region has consistently received the highest applications of pesticide and insecticides compared with other regions (see Fig. 1) in accordance with this region is dominated by arable farming and the greater associated use of pesticides compared to other cropping types (FERA, 2015, 2020a, 2020b). It is likely therefore that there is a greater threat to biota from pesticide use run off in the river systems in this region (Kattwinkel et al., 2011). The decline in both pesticide and insecticide use from 1990 to 2018 across all regions (with the exception is the Southeast) where there has been a concomitant increase in the spatial area of pesticides applications may be explained by the use of more effective chemicals with greater specificity and toxicity, in turn requiring lower application rates (Lawson, 1994; Umetsu and Shirai, 2020). Further factors driving reductions in pesticide application rates include increasingly lower profit margins in farming (driving farmers to be more cost effective), shifts towards more organic farming, and a greater societal awareness of the potential harmful impacts of pesticides. Furthermore, the ban of certain pesticides and/or their restricted emergency authorisation use and only in certain geographical locations and for certain crops types, may also part explain some of the reductions in pesticide/insecticide use (e.g. neonicotinoids, Defra, 2021b).

4.2. Regional status of invertebrate populations in English rivers

Across the sites monitored in England by the Environment Agency between 1965 and 2018, $SPEAR_{pesticides}$ scores <0.6 (classified as moderate to poor with regards to proportional presence of pesticide sensitive species), were commonplace, prominently in the Eastern regions, with hotspots also in the Midlands and Northeast (Fig. 2). The hotspots for low $SPEAR_{pesticides}$ scores in Northeast and Midlands are in areas with high urbanisation and industrial manufacturing (Vaughan and Ormerod, 2012; Bernick et al., 2017; Cole et al., 2018) and likely relate to general aspects of poor water quality. Indeed, studies of urbanised areas with high levels of industrial activity in North Lincolnshire, Staffordshire, Derbyshire and Yorkshire in the England have previously shown highly degraded riverine invertebrates populations are associated with general measures of poor water quality, such as low dissolved oxygen (Beasley and Kneale, 2002; Langford et al., 2009).

Analysing the $SPEAR_{pesticides}$ scores from a temporal (inter-annual) perspective, post-1990, the Anglian and Thames regions consistently had lower average annual $SPEAR_{pesticides}$ scores (see Fig. 3 and Table S5). This finding for the Anglian and Thames regions was independent of whether macroinvertebrate samples were taken during the period May–July or throughout the year. The lower $SPEAR_{pesticides}$ scores observed in the Thames region (Greater London), where only ~7 % of land is used for agriculture (London Assembly, 2010; Cole et al., 2018; Greater London Authority, 2018; Smith et al., 2018), are likely a consequence of urban pollution, urban runoff and wastewater effluent; recently, only one of 47 river waterbodies surveyed in London was classed as 'good' as a result of urban pollution from road run-off, poor river maintenance and problems with sewage infrastructure (Greater London Authority, 2018). Pre-1990, the Northwest region of England had the lowest yearly average $SPEAR_{pesticides}$ score, however, at that time there was a low spatial resolution with relatively few sites monitored for all regions (see fig. S5) adding a cautionary note as to whether these $SPEAR_{pesticides}$ scores were truly representative of the region as a whole. However, post-1990, and with a considerably increased sampling regime the Northwest region consistently had the third highest average $SPEAR_{pesticides}$ scores (see Fig. 3). Post-1990, the Southwest region demonstrated consistently high $SPEAR_{pesticides}$, suggesting macroinvertebrate communities were largely unimpacted by pesticide pollution which is supported by a consistently lower pesticide usage and more dominant pastoral farming occurring across the region, especially compared with the Anglian region (see Figs. 1 and 3; Smith et al., 2018). It is perhaps worth emphasising that the sites selected from the WIMIS database for the ChemPop database (by CEH) were based on there being a 'sufficient' number of samples taken and not on e.g. poor water quality or other parameters, such that the sites studied are likely representative of the region's sites generally (especially as sampling sites are spread across the whole region, see Fig. 2).

Thus, based on the monitoring data, and accepting its limitations, the Anglian and Thames regions had the greatest number of sites recorded consistently with the fewest pesticide sensitive species, compared with the other regions. This highlights both the Anglian and Thames regions are of concern with regards to their riverine invertebrate biodiversity in comparison to other regions, with a substantial absence of pesticide sensitive species in these regions.

4.3. Pesticides effects on the riverine invertebrate communities

We found that the Anglian region's riverine invertebrate communities are likely the most heavily impacted by pesticide and insecticide use (kg/ha; i.e. had the lowest average annual $SPEAR_{pesticides}$ scores).

The influence of pesticide and insecticides on regional riverine invertebrate communities (mean $SPEAR_{pesticides}$ scores) assumes the same proportion (%) of pesticide and insecticide applied to land reaches rivers independent of region. Run-off potential of pesticides (specifically insecticides) have been reported generally similar across regions in England with the exception of the Northwest and Southwest where factors including those relating soil texture, soil organic carbon content, and mean land slope, result in significant areas of land with low/very low run-off potential (Kattwinkel et al., 2011). Overall, therefore major differences in pesticide/insecticide pressure on regional riverine systems should largely relate to the land application quantity (kg/ha). In support of this we found a negative relationship between annual mean $SPEAR_{pesticides}$ scores and the amount of pesticide/insecticide use (kg/ha) across the regions of Anglian, Southeast and Southwest (see Fig. 4, table S13). Interestingly, the greatest influence of pesticide usage on $SPEAR_{pesticides}$ score was in the Southeast and Southwest region with the weakest influence in the Anglian (see table S13). When applying a model to compare the influence of insecticides on regional $SPEAR_{pesticides}$ score based on the fitted line slopes, there was no difference in this effect based on region (see table S13). The differing influence of pesticides on $SPEAR_{pesticides}$ scores across the different regions could be due to different type of pesticides applied and/or their proportional mix. For instance, if pesticide use was made up of a large proportion of insecticides, we would expect this to have a stronger influence on $SPEAR_{pesticides}$ scores compared with a mix dominated by herbicides. This would be supported by the current literature showing insecticide pollution is the main determinant of $SPEAR_{pesticides}$ (Schulz and Liess, 1999; Liess and Von Der Ohe, 2005; Hunt et al., 2017; Ganatra et al., 2021).

Despite similar pesticide and insecticide use (kg/ha) across the Southeast and Anglian regions, the $SPEAR_{pesticides}$ scores were consistently higher in the Southeast. The latter may arise as a consequence of higher historical uses in the Anglian region resulting in more heavily impacted (degraded) riverine communities or even greater use of restricted pesticides through emergency authorisation and a slow recovery from this (Defra, 2021b). The higher $SPEAR_{pesticides}$ scores for the Southwest where there were similar levels of pesticide application to the Southeast and Anglian may in part relate to differences in run-off potential across regions. High risk of pesticide (insecticide) surface run-off from agricultural fields has been shown predictive of lower $SPEAR_{pesticides}$ scores (Schulz and Liess, 1999; Hunt et al., 2017a) and as indicated above the Southwest region is predicted to have lower run-off potential of insecticides, compared to that in the Southeast/Anglian region that is predicted to be median/high (Kattwinkel et al., 2011).

4.4. General water quality effects on riverine invertebrate communities

The $SPEAR_{pesticides}$ indicator is designed to identify macroinvertebrate communities impacted by pesticide (insecticide) pollution, however, other factors including riverbed degradation, organic pollutants and various water quality parameters related to more general pollution (e.g. nitrates, phosphates and dissolved oxygen content etc.,) can influence the $SPEAR_{pesticides}$ score. In turn potentially this can result in overestimates of pesticide (insecticide) pressure at a given site (Bunzel et al., 2013; Malherbe et al., 2018; van der Lee et al., 2020). It is well established that some pesticide sensitive species include species that are also sensitive to general pollution, and other factors including temperature, pH, turbidity etc., notably species belonging to the Ephemeroptera, Plecoptera and Trichoptera (Lydy et al., 2000; Reif, 2002; Weijters et al., 2009; Ippolito et al., 2012; Suhaila and Che Salmah, 2017). Thus, it was no surprise that ammonia and BOD concentrations a negative relationship with $SPEAR_{pesticides}$ scores (see Fig. 5 and table S14). The high BOD likely explains the high density of low $SPEAR_{pesticides}$ scores surrounding industrialised and heavily urbanised areas of England and the high ammonia contributing to the low $SPEAR_{pesticides}$ scores in areas of farming (particularly arable dominated) (Bernick et al., 2017; Cole et al., 2018). Industry, urbanisation and farming (livestock rearing, use of inorganic fertilisers, manure spreading) all increase levels of ammonia and BOD in rivers (Whelan et al., 2022). Over the past century improvements in wastewater treatment from industry and domestic use have resulted in reduced BOD and ammonia concentrations and particularly since 1991, after the implementation of European Urban Waste Water Treatment Directive (UWWTD: 91/271/ED). This is likely to continue with transposed legislation from the EU (specifically the Water Framework Directive, UK Statutory Instrument 2017 No.407) in England and Wales for protecting urban and non-urban areas (Environment Agency, 2022; Whelan et al., 2022). Importantly, wastewater treatment improvements have coincided with reported increases in macroinvertebrate richness at some study sites, such as the River Ray, Swindon and more generally in chalk-streams across the Southwest of England (Durance and Ormerod, 2009; Johnson et al., 2019; Whelan et al., 2022).

General water quality is unlikely, however, to be a major determinant of lower $SPEAR_{pesticides}$ scores seen in the Anglian region. If this were the case, we would expect higher BOD and ammonia concentrations and a much stronger influence of both parameters on $SPEAR_{pesticides}$ than that for the other regions (Bunzel et al., 2013; Malherbe et al., 2018; van der Lee et al., 2020). The Southeast region, which has higher $SPEAR_{pesticides}$ scores (see Fig. 3), had both higher recorded concentrations of ammonia and BOD and showed a stronger influence of ammonia and BOD on $SPEAR_{pesticides}$ score (see Fig. 5; all regions show stronger relationship between BOD/ammonia and $SPEAR_{pesticides}$ than the Anglian region). We thus conclude that the consistently lower average annual $SPEAR_{pesticides}$ scores in the Anglian region are most related to the application quantity of pesticides/insecticides in this region, rather than water quality more generally, albeit other (non-measured) variables may also be a contributing factor. Our analysis illustrates that Anglian is the region of most concern in England with regards to impact of pesticides on riverine macroinvertebrate populations and where there is a considerably lack of pesticide sensitive invertebrate taxa. We suggest therefore that study sites in the Anglian region in England are targeted for studies to more precisely define interrelationships between the effects of specific pesticide/insecticides (or classes thereof) versus other environmental water quality influences, including fluvial geomorphology, temperature, pH, salinity, and nutrients, on riverine invertebrate populations.

5. Conclusions

Here we provide evidence that high pesticide usage is negatively associated with invertebrate populations in regions of England. Pesticide and insecticide usage was negatively associated with lower $SPEAR_{pesticides}$ scores for regions across England, with freshwater invertebrate populations in the Anglian region shown to be the most impacted by crop pesticide/insecticide use. Other more general features of poor water quality were also negatively associated with invertebrate populations, with low $SPEAR_{pesticides}$ scores associated with BOD and ammonia across all regions, and therefore compounding the analyses for associations between pesticide exposures and the status of riverine invertebrate communities.

CRediT authorship contribution statement

Imogen Poyntz-Wright: Conceptualization (lead), methodology (lead), formal analysis (lead), investigation (lead), data curation (lead), resources (supporting), writing—original draft preparation (lead), writing—review and editing (equal), visualization (lead) and project administration (lead). **Charles Tyler**: Conceptualization (supporting), methodology (supporting), writing—review and editing (equal) and supervision (lead). **Xavier Harrison**: methodology (supporting), formal analysis (supporting), writing—review and editing (supporting) and supervision (supporting). **Andrew Johnson**: resources (lead), writing—review and editing (supporting). ***Susan Zappala:*** *writing—review and editing (*supporting*)*. All authors have read and agreed to the published version of the manuscript.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Imogen Poyntz-Wright reports financial support was provided by Natural Environment Research Council. Imogen Poyntz-Wright reports financial support was provided by United Kingdom Department for Environment Food and Rural Affairs. Susan Zappala reports a relationship with JNCC Support Co that includes: employment.

Data availability

I have shared a link to my code and data sources in manuscript.

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Appendix A. Supplementary data

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