Ecotoxicology and Environmental Safety 238 (2022) 113590

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Modelling human health risks from pesticide use in innovative legume-cereal intercropping systems in Mediterranean conditions

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A R T I C L E   I N F O

Edited by Paul Sibley

Keywords:
Human health
Risk assessment
BROWSE model
Long-term effects
Innovative systems
Legume-cereal intercropping
Tillage
Pesticides

A B S T R A C T

Background: The adoption of innovative cropping systems with low pesticide inputs would reduce environmental degradation and dependency on the use of plant protection products. Evaluating the pesticide risk to human health is a growing concern in the assessment of the sustainability of cropping practices. The assessment of human health risks linked to pesticide use in either conventional or innovative cropping systems is poorly documented in the literature.
Objectives: This study focused on the assessment of pesticide exposure and human health risks from pesticide use in arable cropping systems (two monoculture and one intercropping system) associated with the use of various tillage practices (conventional tillage, reduced tillage, and no tillage).
Methods: Human exposure (operators and residents) and health risks from pesticide use were assessed and compared between three conventional and six innovative cropping systems. We used the previously published BROWSE (Bystanders, Residents, Operators, and WorkerS Exposure) model based on data collected from interviews with the farmers and expert knowledge to compare the human health risk from pesticide use in the Setif area. Environmental conditions and the physical characteristics of the farmers were collected on three different farms from 2019 to 2021.
Results: The modelling results demonstrate that human exposure to pesticides was systematically high under conservation tillage (no or reduced tillage) and monoculture cropping (pea and barley) conditions. It was also confirmed that operators experienced the highest cumulated exposure to pesticides (56 mg kg^{-1} bw day^{-1}), followed by resident children seven days after pesticide application (0.66 mg kg^{-1} bw day^{-1}). BROWSE simulations showed that dermal absorption was the most dominant route and represented more than 98% of the total amount of pesticides applied in all cropping × tillage system combinations. Regarding the overall results of the simulated human health risk, barley-pea intercropping was the most interesting system to reduce the risks for both operators and residents for all tillage practices. In addition, intercropping combined with conventional tillage was the most sustainable cropping system in terms of both agronomic performance (crop yield, Land Equivalent Ratio) and human health risk. Furthermore, the availability of advanced crop protection equipment was associated with a significant decrease in exposure and human health risk for both operators and residents.
Conclusions: The prediction of human health risks using BROWSE could help farmers to make the decision to adopt conventional barley-pea intercropping as a good alternative to barley monocultures and pea monocultures under conservation tillage.

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https://doi.org/10.1016/j.ecoenv.2022.113590
Received 30 November 2021; Received in revised form 26 April 2022; Accepted 28 April 2022
Available online 5 May 2022

1. Introduction

Pesticides are chemical substances used in agriculture worldwide to increase crop quality and quantity. According to FAO, Food and Agriculture Organisation of United Nations (2020), the production of major crops has significantly increased since 1960, largely due to pesticides. Worldwide, several pesticides are used to protect crops from weeds and pest damage and, therefore, increase crop productivity (Pan et al., 2021). However, the increased use of pesticides may have a harmful impact on the environmental and socio-economic performance of agricultural systems. The toxic chemical substances in pesticides could contaminate the soil, crops, and surface water through leaching, which may lead to the destruction and loss of plant and animal biodiversity (Mahmood et al., 2016). Furthermore, the overuse of pesticides under conventional crop management (e.g. monoculture) could decrease profitability because of the high cost of chemical pesticides (Giuliano et al., 2016).

In addition to their environmental risks, some pesticides used by farmers can be hazardous to human health. The exposure of humans (farmer, operator, worker, or resident) to pesticides through spraying and the accumulation of pesticide residues in food products is generally associated with various health risks, ranging from short-term to long-term toxic effects (Pan et al., 2020; Grewal1 et al., 2017). In several countries, increasing awareness of the potential health and environmental hazards of pesticides has led to pesticide action plans aimed at restricting the use of pesticides or reducing their impacts. Among those action plans, the adoption of innovative crop and soil management practices is considered one of the most sustainable strategies to improve the environmental, agronomical, technical, and economic performances of cropping systems (Peoples et al., 2019). For example, reduced tillage, crop rotation, and mixed cropping are described as effective practices to improve crop yield and the efficiency of nutrient and water use, in particular under low input conditions (Latati et al., 2017; Bargaz et al., 2017; Plaza-Bonilla et al., 2016). Recent research has shown that the adoption of innovative low-input cropping systems could reduce environmental degradation and the dependency of agriculture on plant protection products (Tamms et al., 2021; Wang et al., 2020). However, there are not enough references available investigating the risks and direct impacts of pesticide use on human health, especially with conservation agriculture practices. The assessment of human exposure to overall pesticide use is indeed limited by (i) the availability or lack of field experimental data that are useful to derive some parameters such as the characteristics of the exposed population, the agro-pedo-climatic conditions, and the pesticide properties (Lammoglia et al., 2017c); (ii) the lack of expert knowledge that is poorly documented in the literature; and (iii) the complexity of assessing the human health risk because of the wide range of exposure routes (inhalation, dermal, and ingestion) (Damalas and Koutroubas, 2016).

Some modelling frameworks such as the Pesticide Handlers Exposure Database, the UK Predictive Operator Exposure Model (UK POEM), the German Operator Exposure Model (German model), have been developed over the past decade to improve understanding and assess the exposure of human health to plant protection products (PPPs) (Butler Ellis et al., 2010). They contain generic exposure data (submitted on a voluntary basis) describing workers mixing/loading and/or applying pesticides in the field. However, they used deterministic calculations to derive exposure values from actual exposure studies where the same formulation types, equipment and methods were employed. Among these frameworks, only a limited number is able to deal with innovative cropping systems (crop residue management, mixed cropping, rotation, and tillage practices) (Lammoglia et al., 2018). In this context, through the EU seventh framework to improve regulatory drift exposure and risk assessment in the European Union, some recent European projects have developed and published new modelling approaches for human exposure to pesticide risks (Kennedy and Butler Ellis, 2017). These include the European Predictive Operator Exposure Model (EUROPOEM), the Bystander and Resident Exposure Assessment Model (BREAM), and the BROWSE model; the latter is a probabilistic model of pesticide exposure (Butler Ellis et al., 2010, 2017a, 2017b). EUROPOEM and BREAM were developed to assess both resident and bystander exposure to pesticides used in agriculture (Van Hemmen, 2001; Anon, 2011). However, their parameterisation was based on general, incomplete regulatory databases in which real cropping practices and exposure routes are poorly incorporated (Butler Ellis et al., 2017a). The BROWSE model is one of the most mechanistic models developed according to the probability distributions of exposure to pesticides (Butler Ellis et al., 2017b). Compared with the previous models, BROWSE aims to improve and describe more realistic scenarios for analysing a wide range of current cropping practices. It has been evaluated and led to more realistic exposure simulations than the models developed previously, thanks to the incorporation of various exposure routes (dermal, ingestion, and inhalation).

Using the probabilistic European model, BROWSE, Lammoglia et al. (2017a, 2017b, 2017c) assessed the risks for human health of various pesticide uses in conventional (monoculture and tillage) and innovative (intercropping and conservation tillage) cropping systems tested in France. The results showed that innovative low-input cropping systems could reduce the human health hazard in comparison with the corresponding conventional cropping systems. They also predicted a higher probability that conservation tillage systems would lead to unacceptable risks for human health because of the high number of pesticide applications.

The main objective of this research paper was to use the BROWSE model to calculate the human health risk of legume-cereal intercropping systems and compare the overall health risk of conventional and innovative cropping systems. Two innovative tillage practices (reduced tillage and no tillage) were compared with conventional tillage practices for three cropping patterns (pea monoculture, barley monoculture, barley-pea intercropping). To refine the simulation and compute more realistic assessments of the human health risk, the characteristics of the exposed population, as well as the pesticide handling techniques, were monitored on three different farms instead of using the BROWSE default values.

2. Material and methods

2.1. BROWSE model

BROWSE is a mechanistic model developed through the European BROWSE project (2011–2014) to predict human exposure to different liquid and solid formulations of pesticides (including seed treatments). It is one of the most mechanistic models developed on the basis of pesticide exposure probability distributions considering the requirements from European EC Regulation 1107/2009 and the Sustainable Use Directive. Compared to models developed previously, BROWSE aims to improve and describe more realistic exposure scenarios to analyse the risk of using plant protection products in various farming practices commonly adopted by farmers today. By definition, an exposure scenario is a set of conditions including operational conditions and risk management measures, that describe how the substance is used and how the manufacturer or importer controls, or recommends downstream users to control exposures of humans and the environment (ECHA, 2008). There are different types of scenarios through which exposure to chemicals of operators, workers, residents and bystanders may occur. This is why, in the BROWSE model, many realistic scenarios have been developed instead of the usual strategy for regulatory approaches, which is to use a limited number of worst-case scenarios for which exposure is assessed. The exposure scenarios were identified and prioritised based on the scenarios covered by the current exposure models, the frequency of each scenario (common case assumption), the extent of the exposure that is foreseen (worst-case assumption), and availability of exposure data (Butler Ellis et al., 2010). For each scenario, BROWSE modelling procedure included the collation of available evidence from the literature, statistical (regression and correlation) analyses of the available exposure data, identifying exposure determinants and allocating effect sizes and distributions of model inputs, adopting experimental data as input for the models (where available and/or appropriate) and finally developing mechanistic algorithms. Depending on the available evidence of exposure determinants of a given scenario, available experimental data and existing models, the most useful type of model was selected. The orchard scenarios for example are modelled empirically while the arable scenarios are evaluated using process-based simulation models (Kennedy and Butler, 2017). To calibrate and validate the BROWSE model, its outputs were compared with estimates generated by other models (e.g. EUROPOEM).

Each scenario is represented in the BROWSE modelling software with default input parameters which can be changed as required. Before every model run, some inputs may be fixed while others are assigned probability distributions. The Browse model includes some predefined constants and distributions taken from EFSA guidance, US Environmental Protection Agency (EPA) or other sources. The probability distributions can be used to represent uncertainty of the input values. Some input distributions are always used because they represent natural variation in conditions around a nominal selected value, or a uniform distance from the source between the minimum and maximum values. Others input distributions are optional and can be selected by the user or replaced with a constant value (Kennedy and Butler, 2017). The scenario refinement options offered by the BROWSE model promise assessments of a wider range of scenarios in comparison to the previous empirical models.

2.1.1. *Population exposed to pesticide risk*

Operators, bystanders, residents, and workers are considered as the main populations for which exposure is assessed. The European Food Safety Authority (EFSA) considers that the definitions of both residents and bystanders should be correlated with the duration of exposure in the area where plant protection products are applied (EFSA, 2014). BROWSE defines bystander exposure as acute exposure, whereas resident exposure is defined in the short or longer term. Regardless of the purposes of the BROWSE model, residents and bystanders are considered as a single population living on-site (near the farm) or temporarily visiting: (i) people who might be in or immediately adjacent to the treated area, (ii) their presence is entirely fortuitous and not related to the work involving PPPs, and (iii) the position of the person could result in them potentially being exposed to the risks (Butler Ellis et al., 2017a). Operators are defined as people who participate in activities related to the application of plant protection products from the handling of the products to applying the pesticides. Pesticide application activities also include the preparation of the spray mixture and cleaning of the machinery after use. The operator can be a professional farmer or a worker employed for this purpose (Lammoglia et al., 2017c). A worker is defined as any person who, as part of their employment, enters an area that has previously been treated with a pesticide or who handles a crop that has been treated with a pesticide. In the BROWSE model, there is currently no scenario for worker exposure to arable crops because these crops are harvested with machines and, therefore, workers are not considered. The crops we studied were harvested mechanically, so this work only presents operator and resident exposure to pesticides.

2.1.2. *Human health risk calculation*

Operators, residents, and bystanders may be exposed to pesticides either directly through contact with spray drift (dermal or inhalation) or indirectly through contact with drift deposits (dermal or ingestion). Exposure could also be induced by vapour drifts resulting from the volatilisation of deposits. Exposure is expected to decline over time from the initial value at the time of application (EFSA, 2014). For the operator, BROWSE assesses three exposure routes: inhalation, ingestion, and dermal. The total cumulated exposure is defined as the sum of these three routes (Butler Ellis et al., 2017b). Dermal exposure occurs through deposition from the air or contact of the hand or other parts of the body with surfaces. It can also be induced through direct contact by dripping or impaction (Damalas and Eleftherohorinos, 2011).

The use of personal protective equipment can affect and reduce pesticide migration. However, for residents, the BROWSE model currently includes exposure to spray mist during pulverisation, as well as exposure to vapour following pulverisation. For people exposed after spraying, the exposure routes are inhalation of the vapour emitted from the treated crop and hand contact with contaminated ground (Lammoglia et al., 2017c). Furthermore, the assessment of exposure and the human health risk is generally determined one day after pesticide application (short term) and seven days after pesticide application (long term), for both operators and residents. In the case of exposure to spray drift, the minimal distance between the location of residents and bystanders and the treated area was also defined by the BROWSE model as being between 2 m and 20 m (Van den Berg et al., 2016). In this study, exposure and the relative human health risk among all the cropping systems assessed were set at the 50th percentile to avoid both over and under simulated data.

Firstly, the BROWSE model simulates the cumulated human exposure to pesticides followed by the quantity of pesticides absorbed. It then calculates the human health risk index (HR index) by dividing the amounts of each pesticide absorbed by the corresponding pesticide AOEL (Acceptable Operator Exposure Level). The AOEL is the maximum amount of an active substance to which a human may be exposed, through any exposure route, without any adverse health effects (mg of active substance per kilogram of body weight). According to the methodology developed by Lammoglia et al. (2017c), when the HR index is over 100%, the risk to human health is considered unacceptable. Conversely, when the HR index is below 100%, the risk is considered acceptable.

2.2. *Study sites*

Three different farm(er)s located in the Setif region in north-eastern Algeria were studied for three successive growing seasons (2019, 2020, and 2021). The first farm (farm 1) was located in Mezloug (36°06′ N, 5°20′ E) in the centre of Setif, whereas the other two farms (farms 2 and 3) were located in Beni fouda (36°15′ N, 5°30′ E) in the north of Setif. Meteorological data for each farm were obtained from the National Office of Meteorology in Setif (https://www.infoclimat.fr/observations-meteo/archives/1er/janvier/2019/setif/60445.html) (Table 1). The climate is typically Mediterranean: a warm temperate climate and dry summer. In terms of the repartition of both mean precipitation and minimal temperatures, there is a significant difference between Mezloug and Beni fouda. From 1981 to 2020, Mezloug and Beni fouda received on average 401 mm and 440 mm of annual rainfall, respectively. The mean annual temperatures were 14.93 °C and 15.11 °C (Table 1), respectively. However, no significant difference was observed in terms of maximum and mean annual temperatures between the two sites.

The soil characteristics of each field were measured. For each field, a composite sample of topsoil (0–30 cm) was obtained from four subsamples. The sand, loam, and clay fractions of the soils were determined using the Particle Size Analysis method (Bowmann and Hutka, 2002). The total N content of the soil was determined using the Kjeldahl method (Lynch and Barbano, 1999), and the total Phosphorus content was determined using the Malachite green method after mixed digestion with nitric and perchloric acids (Valizadeh et al., 2003). The organic carbon content was determined using the Anne method (McBratney et al., 2000). The soil was suspended in deionized water (soil:water ratio = 1:2.5) and the pH measured using a pH-metre (Shen et al., 1996). The soil bulk density and the soil hydraulic parameters were also measured on all the farms. The wilting point and field capacity were determined using the pressure-based method (Richards and Weaver, 1944) and the soil bulk density was measured by weighing the dry soil samples

Table 1
Climate and soil physicochemical and hydraulic characteristics of the fields of the three farmers.

Soil and climate properties (0–30 cm)	Farm 1 Mezloug	Farm 2 Benifouda	Farm 3 Benifouda	Significant level
Mean annual rainfall (1981–2020)	401[b]	440[a]	440[a]	*
Mean annual minimum temperature (1981–2020)	9.25[a]	8.81[b]	8.81[b]	*
Mean annual maximum temperature (1981–2020)	20.64[a]	21.42[a]	21.42[a]	n.s
Annual mean temperature (1981–2020)	14.93[a]	15.11[a]	15.11[a]	n.s
Clay (%)	43[a]	39[a]	41[a]	n.s
Loam (%)	35[a]	32[a]	34[a]	n.s
Sand (%)	22[b]	29[a]	25[c]	**
$CaCO_3$ (%)	22[b]	26[a]	21[b]	*
Soil Organic Matter (%)	1.25[b]	1.92[a]	2.34[a]	**
Total Nitrogen (g kg^{-1})	1.33[b]	1.63[a]	1.76[a]	*
Total Phosphorus (mg kg^{-1})	264[c]	325[ab]	359[a]	***
Available N (mg kg^{-1})	22[b]	33[a]	37[a]	*
Available P (mg kg^{-1})	9.24[a]	12.35[ab]	19.21[b]	*
pH	8.41[a]	8.53[a]	8.12[b]	*
Bulk density (g cm^{-3})	1.39[a]	1.20[b]	1.15[b]	*
Soil water content at wilting point (m^3 m^{-3})	0.15[a]	0.13[b]	0.13[b]	*
Soil water content at field capacity (m^3 m^{-3})	0.24[a]	0.18[b]	0.21[ab]	**

Meteorological data were collected from the website of the National Office of Meteorology in Setif (Algeria) (https://www.infoclimat.fr/observations-meteo/archives/1er/janvier/2019/setif/60445.html).
Means with different letters in the same row are significantly different.
*, ** and *** denote significant differences at $p < 0.05$, $p < 0.01$ and $p < 0.001$, respectively
n.s. Not Significant.

collected using a metal ring pressed into the soil (McKenzie et al., 2004) (Table 1).

All three soils had a loamy-clayey texture but the proportion of sand was higher in Beni fouda (Farms 2 and 3) than in Mezloug (Farm 1). The soil on the three farms was calcareous vertisol (more than 21% calcium carbonate) with an alkaline pH (ranging from 8.12 to 8.53). The soil was relatively poor in organic matter (1.25%) in Mezloug compared to Beni fouda (1.92% and 2.34%), and the overall nitrogen and phosphorus availability was lower in the soil on farm 1 (located in Mezloug) compared to farms 2 and 3 (located in Beni fouda). The soil hydraulic parameters - soil water content at both wilting point and field capacity - were significantly greater in Mezloug than in Beni fouda (Table 1).

2.3. Model set-up

2.3.1. Cropping systems

Three conventional and six innovative cropping systems were studied. The three (3) conventional systems were pea monoculture with conventional tillage (Pea_CT), barley monoculture with conventional tillage (Bar_CT), and barley-pea intercropping with conventional tillage (Interc_CT). The conventional systems were compared with six (6) innovative systems: pea monoculture with reduced tillage (Pea_RT), barley monoculture with reduced tillage (Bar_RT), barley-pea intercropping with reduced tillage (Interc_RT), pea monoculture with no tillage (Pea_NT), barley monoculture with no tillage (Bar_NT), and barley-pea intercropping with no tillage (Interc_NT).

The adoption of barley-pea intercropping by farmers in the Setif region was motivated by the need to reduce N-fertiliser use and pesticide applications with respect to conventional monoculture systems. To reach and ensure such objectives, these intercropping systems were also combined with innovative tillage practices (no tillage and reduced tillage).

With conventional tillage practices, the soil was ploughed using spring mouldboard inversion ploughing (25–40 cm in depth), rotary harrowing, and mechanical weeding. However, for reduced tillage, farmers only used a chisel plough with a packer roller, followed by light disking and drilling. For no tillage practices, soil management was based on direct sowing of the crop (with direct drill planters) into soil that had not been tilled since the previous growing season. The crop sowing density in both monoculture and intercropped systems was chosen according to local standard cropping practices. The seed rate was 115 kg per hectare (ha) for barley monocultures, 130 kg per ha for pea monocultures, and 40 kg and 80 kg per ha for barley and pea intercropping. Barley and pea were sown in a mixture on intercropped plots. All the cropping systems were set up in the 2019, 2020, and 2021 growing seasons with an inter-row distance of 17 cm and were rain-fed with a low fertilisation rate of 30 kg ha^{-1} of nitrogen-phosphorus-potassium fertiliser (NPK).

2.3.2. Pesticide usage and handling

We collected the primary data required to run the Browse model (Table 2, Table S1, and Table S2) from all the cropping systems studied. The name of the pesticide used, the corresponding active substances, the number of applications in each system, and the corresponding dose were collected through a survey (Table 2). Pesticide management was identical from one cropping season to the next. In total, eleven (11) different plant protection products containing nine (9) different active substances were used each year in the nine cropping systems studied (Table 2). The treatment frequency indices calculated (TFI: number of doses applied per hectare in each growing season) are also presented in Table 2 for each cropping system. No pesticides were applied for barley-pea intercropping during crop development (regardless of tillage practices), except for two fungicides (Acil 060FS and Celest extra) used for seed treatment before sowing (Table 2). Glyphosate was only applied in no-till cropping systems. This could be explained by the key role of glyphosate in controlling weed infestation, especially in conservation tillage systems (Table 2).

Some of the BROWSE default input variables originate from European guidance and regulatory databases. To avoid relying too much on those probabilistic default values, the pesticide handling practices and handling equipment were recorded for each farmer, as well as the physical characteristics of the residents or bystanders (adults and children) living within a distance of 2–20 m from the farm plots (Table S1). Table S1 summarises the input parameters related to the scenario, operator personal protective equipment, mixing and loading methods, and resident characteristics for the three farms studied. The farmers were asked which types of safety equipment they used when mixing, loading, and spraying pesticides. The most common safety equipment used included rubber gloves and full-face masks (Type P1), long sleeves, and trousers, but no hood or visor. The information regarding the protective equipment used by farmers was used in the Browse model to select the appropriate protective factors (Table S1). Instead of using the Browse default probabilistic distribution of body weight, the actual body weight of the farmers, as well as the average body weight of the children living near the farms, was obtained from the farmer surveys (Table S1). The required data relating to the physicochemical, toxicological, and environmental properties of the pesticides sprayed were collected from four online databases: Pesticide Properties Database (http://sitem.herts.ac.uk/aeru/), Sage pesticides (https://www.sagepesticides.qc.ca/), PubChem (https://pubchem.ncbi.nlm.nih), and Bayer Crop Science

Table 2
Description of the cropping and tillage systems based on pea and barley monocultures and barley-pea intercropping, and the corresponding pesticide, applied dose, treatment frequency index (TFI), and the different active substances in each cropping system combination.

Crop	Cropping system	Soil management	Products name	Application dose (L ha^{-1})	TFI	TFI Total	Active substances
Pea	Monoculture	Conventional tillage	GESAGARD FW	3	1.1	3.8	Fludioxonil, Prometryn and Fluasifop-P-butyl
			FUSILADE MAX	1.5	1.4		
			CELEST EXTRA	2.6	1.3		
		Reduced tillage	GESAGARD FW	3	1.1	3.8	Fludioxonil, Prometryn and Fluasifop-P-butyl
			FUSILADE MAX	1.5	1.4		
			CELEST EXTRA	2.6	1.3		
		No tillage	FUSILADE MAX	1	1	3.8	Fludioxonil, Fluasifop-P-butyl, Glyphosate, Thiamethoxam and lambda-cyhalothrin
			CELEST EXTRA	2.6	1.3		
			Glyphosate	2	1.3		
			ENGEO	0.1	0.2		
Barley	Monoculture	Conventional tillage	RAPID 750 DF	200	1.3	1.8	Tebuconazole and Tribuneron-methyl
			ACIL 060FS	0.5	0.5		
		Reduced tillage	SEKATOR ® OD	0.15	1	3.1	Tebuconazole, Iodo-sulfuron-methyl-sodium, Amidosulfuron, Mefenpyr-diethyl, Fenoxaprop-P-ethyl and Diclofop-methyl
			ACIL 060FS	0.5	0.5		
			DOPLER PLUS	2.5	1.6		
		No tillage	Glyphosate 480	3	1.9	4.95	Tebuconazole, Glyphosate, Iodo-sulfuron-methyl-sodium, Amidosulfuron, Mefenpyr-diethyl, Fenoxaprop-P-ethyl and Diclofop-methyl
			ACIL 060FS		0.5		
			SEKATOR ® OD	0.15	0.95		
			DOPLER PLUS	2.5	1.6		
Barley + Pea	Intercrops	Conventional tillage	ACIL 060FS	0.5	0.5	1.8	Tebuconazole and Fludioxonil
			CELEST EXTRA	2.6	1.3		
		Reduced tillage	ACIL 060FS	0.5	0.5	1.8	Tebuconazole and Fludioxonil
			CELEST EXTRA	2.6	1.3		
		No tillage	ACIL 060FS	0.5	0.5	3.1	Tebuconazole, Fludioxonil and Glyphosate
			CELEST EXTRA	2.6	1.3		
			Glyphosate 480	3	1.3		

TFI: Treatment Frequency Index.

(https://www.cropscience.bayer.us/products/insecticides) (Table S2). For input data not obtained from the farmer surveys, the values were set at their initial default values as defined in the BROWSE software interface (Kennedy et al., 2017).

2.4. Data analysis

Based on the surveys conducted with each of the farmers in Mezloug and Beni fouda, the names of the pesticides used, including insecticides, herbicides, and fungicides, the number of applications, and the dose applied to the fields were used to calculate the Treatment Frequency Index (TFI). This corresponds to the number of registered doses of pesticides applied to one hectare. The TFI was calculated using Eq. (1). The TFI is equal to 0 when no chemical pesticides are applied. High TFI values highlight both high frequencies of application and the application of excessive doses compared with the official approved rates.

$$TFI = (Applied\ Dose/Recommended\ dose) \times (Treated\ area\ size/Field\ size) \quad (1)$$

The interaction effect of barley and pea in a mixture was calculated using a competition index called Land Equivalent Ratio (LER). This corresponds to the land area required for single crops to produce the same grain yield as intercropping (Jensen et al., 2015) (Eq. (2)). LER was computed as described in Eq. (2).

$$LER_{ab} = Y_{ab}/Y_{aa} + Y_{ba}/Y_{bb} \quad (2)$$

Y_{aa} and Y_{bb} are the barley monoculture and pea monoculture yields, respectively, and Y_{ab} and Y_{ba} are the barley and pea intercropping yields, respectively. LER is used to assess the effectiveness of intercropping. The latter is considered more advantageous when the LER is greater than one.

Using STATISTICA software, the statistical effects of the tillage practices and crop management systems on human exposure, grain yield, and LER were evaluated by performing one-way analyses of variance (ANOVA) for which a significance level of 95% was considered (equivalent to a p-value < 0.05).

3. Results

3.1. Population exposure to pesticide risks

The cumulated human exposure was significantly different among cropping systems (pea monoculture, barley monoculture, barley-pea intercropping) and tillage practices (conventional tillage, reduced tillage, no tillage) (Fig. 1). Furthermore, exposure values were highly different between operators and residents. Operators were the most exposed to pesticides with their cumulated exposure ranging from 14 to 56 mg kg^{-1} bw day^{-1}. However, the highest exposure (0.66 mg kg^{-1} bw day^{-1}) for residents was observed for children, especially exposure one day (D1) after pesticide application (short term exposure). Exposure for children was four and five times higher than adult exposure on D1 (short term exposure) and D7 (long term exposure), respectively. In terms of tillage practices, the highest exposure was observed for barley monocultures with no tillage (Bar_NT), followed by reduced tillage (Bar_RT). This was confirmed for the exposure of all populations on D1 and D7. Cumulated exposure for barley cultivated under conventional tillage (Bar_CT) was nearly three times lower than with no tillage (Bar_NT) for both operators and residents.

Regarding conventional tillage practices, the highest human exposure was observed in pea monocultures (Pea_CT) compared to barley monocultures (Bar_CT) (+ 33%) and barley-pea intercropping (Interc_CT) (+ 99%). In general, the lowest values of human exposure were observed for barley-pea intercropping (Interc_CT) with conventional tillage, with 12.5, 0.03, 0.15, 0.016, and 0.1 mg kg^{-1} bw;day^{-1} for the exposure of operators and adults on D1, children on D1, adults on D7, and children on D7, respectively (Fig. 1).

3.2. Amounts of pesticides absorbed by humans, operators, and residents

Fig. S1 shows the BROWSE results relative to the simulated values of the total amount of pesticides absorbed for each combination of cropping practices (cropping system × tillage system). The amount of pesticides absorbed was largely greater for the operator (from 10 to 50 mg kg^{-1} bw day^{-1}) than for residents (from 0.0025 to 0.38 mg kg^{-1} bw day^{-1}). For residents, children absorbed higher amounts of pesticides than adults, and the quantities absorbed following exposure one day after pesticide application (D1) were higher than seven days after application (D7). The highest total amounts of pesticides absorbed by humans (both operators and residents) were observed for barley monocultures with no tillage (Bar_NT). For barley monocultures, the total amounts of pesticides absorbed were significantly lower with conventional tillage, Bar_CT (65%, 62%, 64%, 66%, and 72% lower for the operator, adults on D1, children on D7, adults on D7, and children on D7, respectively), and reduced tillage, Bar_RT (15%, 7%, 12%, 14%, and 25% lower) compared to no tillage barley monocultures, Bar_NT (Fig. S1).

Regardless of the tillage practices, dermal penetration was the most common exposure route, representing 98% of the total amount of pesticides absorbed by the operators and residents, whereas only 2% were absorbed by inhalation (less than 0.5%) and ingestion (less than 1.5%) for the operators. The rate of ingestion was 0% for exposure of residents to pesticides on D1 and on D7. However, the rate of inhalation was less than 2% (barley monoculture) for the exposure of children on D7 with no tillage, and it was less than 0.5% for the other cropping system combinations (Fig. S1).

3.3. Human health risks after exposure post pesticide application

Figs. 2–4 show the values of the human health risk index (HR) obtained with BROWSE in conventional, reduced tillage and no tillage systems, respectively. For each tillage system, HR values were assessed and compared among cropping systems and farms. A lower health risk was found for the farmer who adopted protective measures compared to their counterparts (Figs. 2–4).

With conventional tillage, HR values varied from 2.7% to 100% of the AOEL indicating an acceptable risk for both operators and residents for all the farm plots (farms 1, 2, and 3). Furthermore, operator HR values (2.7–100%) were largely higher than those observed for the majority of residents except for child at long term exposure (up to 512%). For the exposure of both operators and residents at long term, the highest HR values were calculated on farm 1 and farm 3 with pea monoculture cropping systems (Fig. 2).

For reduced tillage systems, the operator HR was greater than 100% for most of the cropping systems on all three farms. Indeed, the HR values ranged from 270% to 500% of AOEL for most of the cropping systems. However, the highest HR values (up to 3335% of AOEL) were observed more particularly for barley monocultures on farm 1 and farm 3. For both adults and children, human health risk index (HR) was lower than 100% one day after pesticide application, ranging from 14.3% to 15.3% and from 74% to 77.6% respectively. Thus, in this case, the HR values were relatively similar for both cropping systems and farms (Fig. 3). Regarding the exposure of adults and children at long term, the HR index followed the same trend as that observed for operators. The highest HR values were observed more particularly for the barley monoculture systems cultivated by farmer 1 and farmer 3. However, the HR index only exceeded the acceptable exposure level (171%) for exposure of children at long term (Fig. 3).

According to Fig. 4, applying pesticides in no-tillage systems led to an unacceptable risk for operators (HR ranged from 270% to 4000% of the AOEL) in all the cropping systems, except for one application of plant protection products on barley monocultures by farmers 1 and 3 (HR = 71%). Indeed, the highest risk was systematically observed for operators in both barley and pea monocultures (on farm 1 and farm 3), whereas it was lower in intercropping systems (Fig. 4). The same trend was observed for exposure of children on D7; HR was greater than 100% with the application of five plant protection products on barley (HR = 167%) and pea monocultures (HR = 205%) on farm 1 and farm 3. However, all applications of plant protection products for intercropping systems on all three farms led to acceptable risks (HR ranged from 13% to 25%). We also observed an acceptable risk for exposure of children on D7 on farm 2 for all the cropping systems (barley, pea, and intercropping). Regarding exposure one day after application, all HR values varied from 74% to 77.5% indicating no risk for children for all the cropping systems. The lowest HR values for residents were observed for the exposure of adults one and seven days after application. HR was then lower than 16% and 33% for adults after exposure one and seven days after application, respectively (Fig. 4).

3.4. Pesticide use intensity and crop productivity

The plant protection performance estimated with the treatment frequency index (TFI, see Eq. (1)), differed between cropping systems and tillage practices. The use of pesticides in conventional and reduced tillage systems was similar or lower in barley-pea intercrops (total TFI of 1.8 for both tillage practices) than in barley monocrops (total TFI of 1.8 and 3.1, respectively) and pea monocrops (total TFI of 3.8 for both tillage practices). The highest TFI were observed in no tillage systems with total TFI of 3.1, 4.95, and 3.8 for barley-pea intercrops, barley monocrops, and pea monocrops, respectively (Table 2). This index varied significantly for barley-pea intercrops and barley monocrops, whereas the environmental pesticide pressure was identical for all three tillage management practices with pea monocrops (TFI = 3.8).

During all the cropping seasons, grain yield varied significantly in response to the tillage practices in all cropping systems, except in the 2019 season for the barley monoculture (Fig. 5). We observed a consistently greater yield in the barley-pea intercropping system than monocultures resulting in a yield 15–33% and 150–175% greater with intercropped barley-pea than barley monocultures and pea monocultures, respectively. In the intercropping system, the mixed barley and

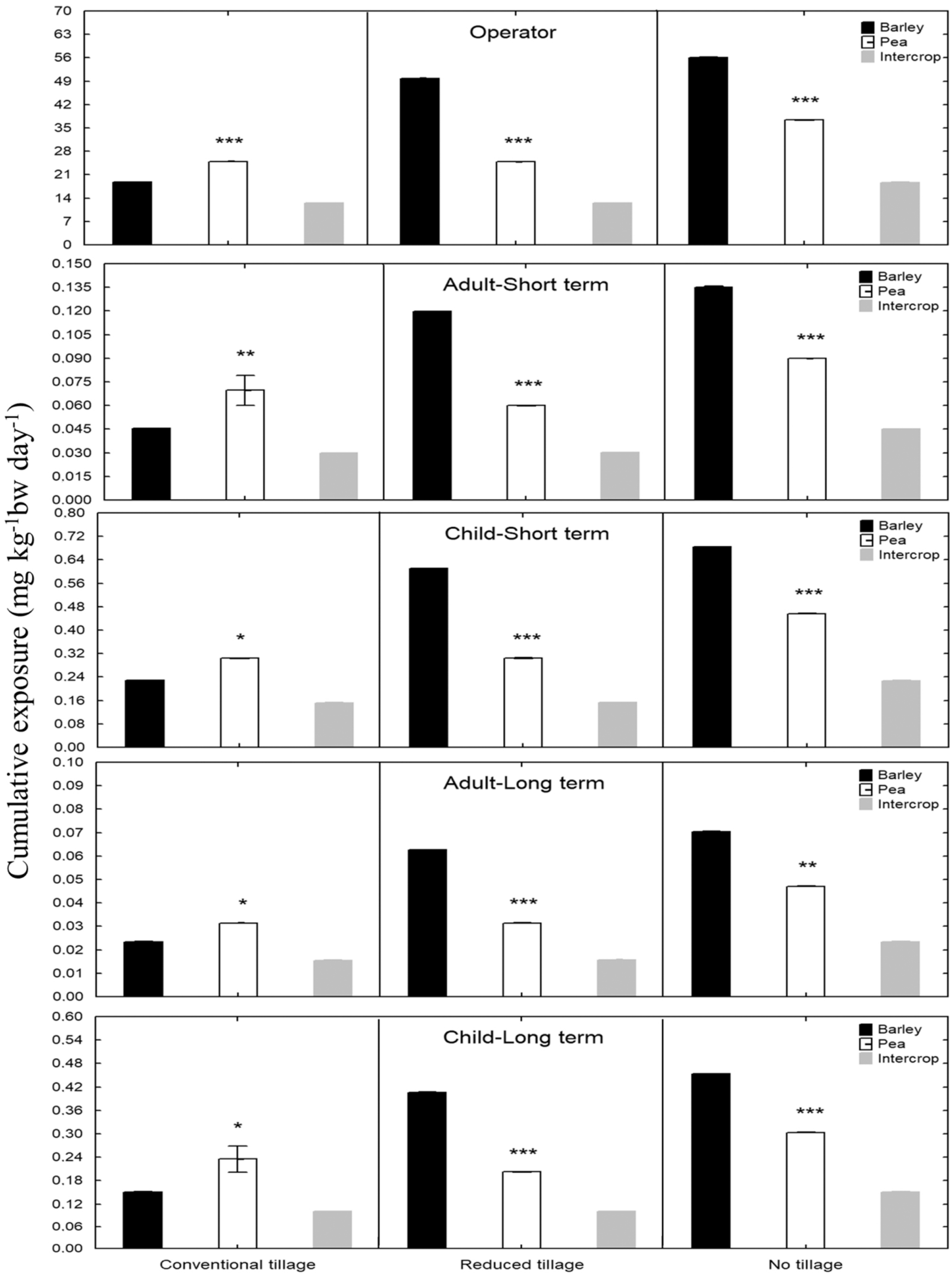

Fig. 1. Cumulative exposure of humans - operator, adult and child on day 1 (short term) and day 7 (long term) after application - to pesticides for barley monocultures, pea monocultures, and barley-pea intercropping in conventional, reduced, and no tillage systems. Data are means and SE of 3 replicates (farmer 1, 2, and 3). Asterisks *, ** and *** denote significant differences at $p < 0.05$, $p < 0.01$, and $p < 0.001$, respectively.

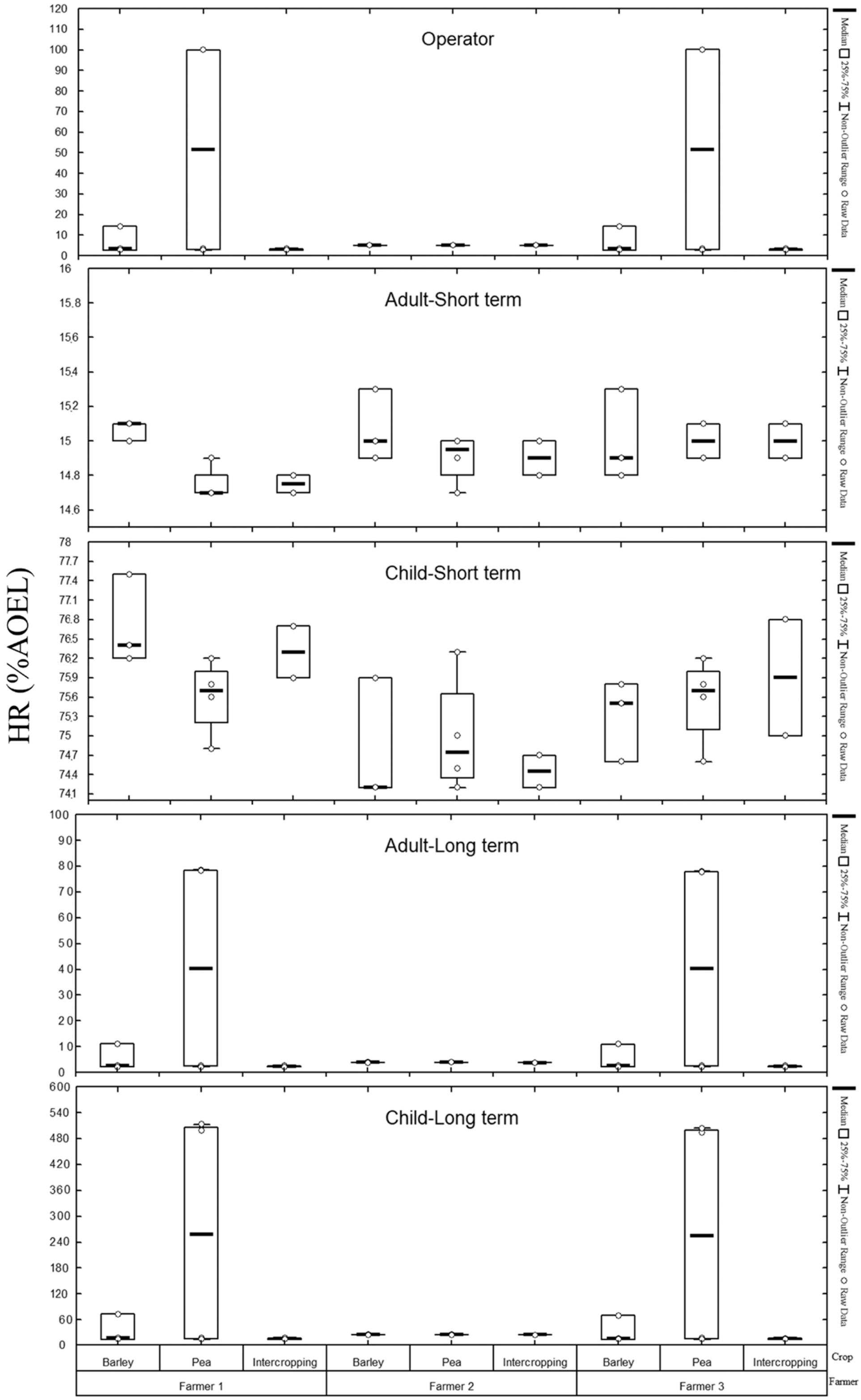

Fig. 2. Distribution of the "Human health risk index" (HR, % of the AOEL), calculated as the ratio of the amount absorbed to the AOEL for each pesticide applied to barley monocultures, pea monocultures, and barley-pea intercropping in conventional tillage systems.

pea yields were greatest on plots using conventional tillage (Inter_CT) (Fig. 5.C). The same pattern was observed for the barley monocultures where the yield with conventional tillage (Bar_CT) was + 39% higher than the yield with no tillage (Bar_NT) in 2021 (Fig. 5.A). Conversely, the pea monoculture yields observed were significantly greater with no tillage (Pea_NT) (+ 26% in 2019 and + 62% in 2020, respectively) and reduced tillage (Pea_RT) (+ 23% in 2021) compared to conventional tillage (Pea_CT) (Fig. 5.B).

Land equivalent ratio (LER) values in each tillage system and cropping season were greater than 1. This led to more yield productivity in the barley-pea intercropping systems than the monocultures, regardless of the tillage practices (Fig. 5.D). The highest LER values were recorded

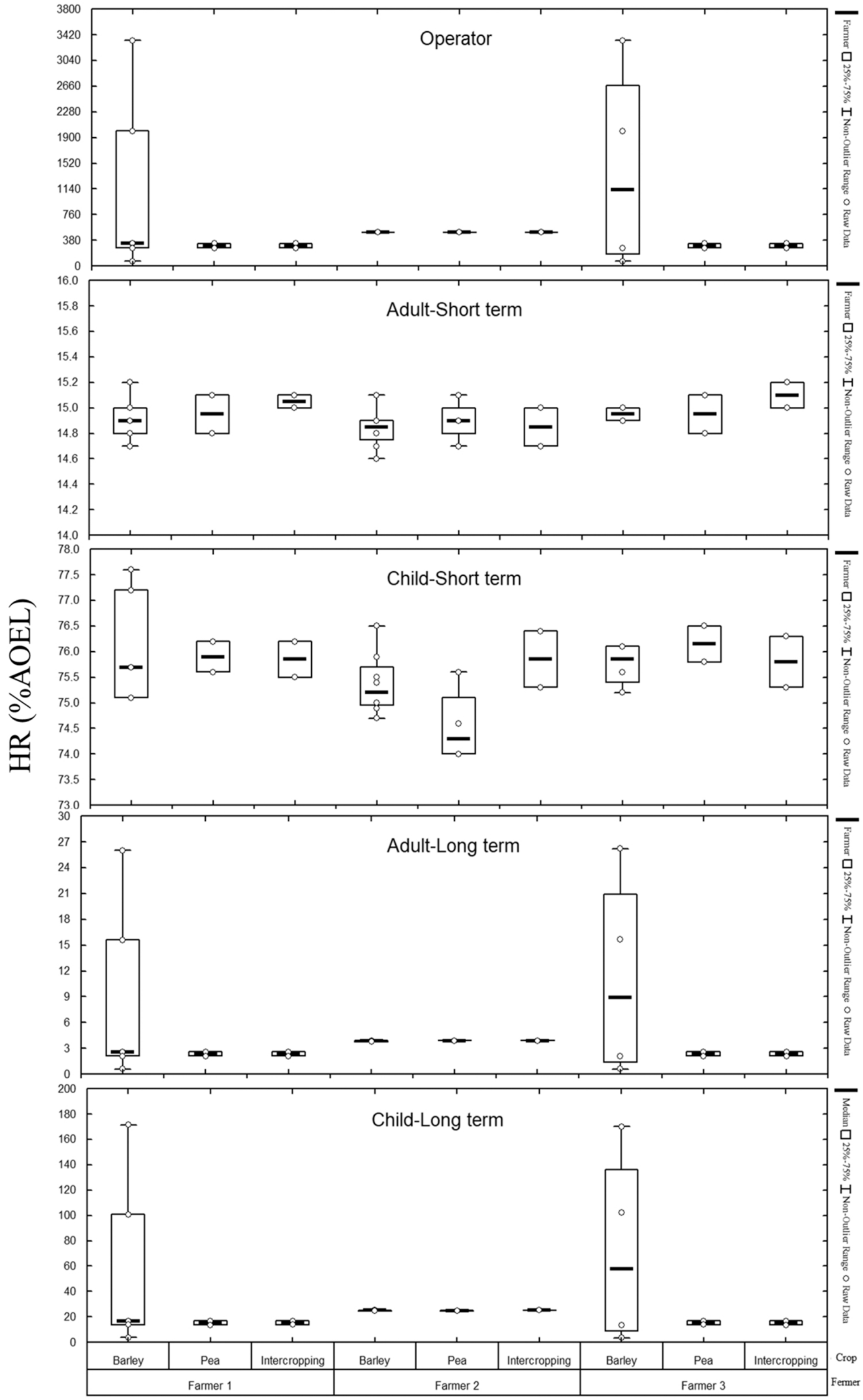

Fig. 3. Distribution of the "Human health risk index" (HR, % of the AOEL), calculated as the ratio of the amount absorbed to the AOEL for each pesticide applied to barley monocultures, pea monocultures, and barley-pea intercropping in reduced tillage systems.

with conventional tillage (1.89, 1.82, 1.67 in 2019, 2020 and 2021 respectively), followed by reduced tillage (1.73, 1.81, 1.49 in 2019, 2020 and 2021 respectively) then no tillage (1.40, 1.19, 1.42 in 2019, 2020 and 2021 respectively). Indeed, intercropping was more advantageous when combined with conventional tillage compared with no tillage with a grain yield 35%, 53%, and 17% higher in 2019, 2020, and 2021, respectively.

4. Discussion

The probabilistic BROWSE model developed within the Browse European project (www.browse-project.eu) were successfully applied to

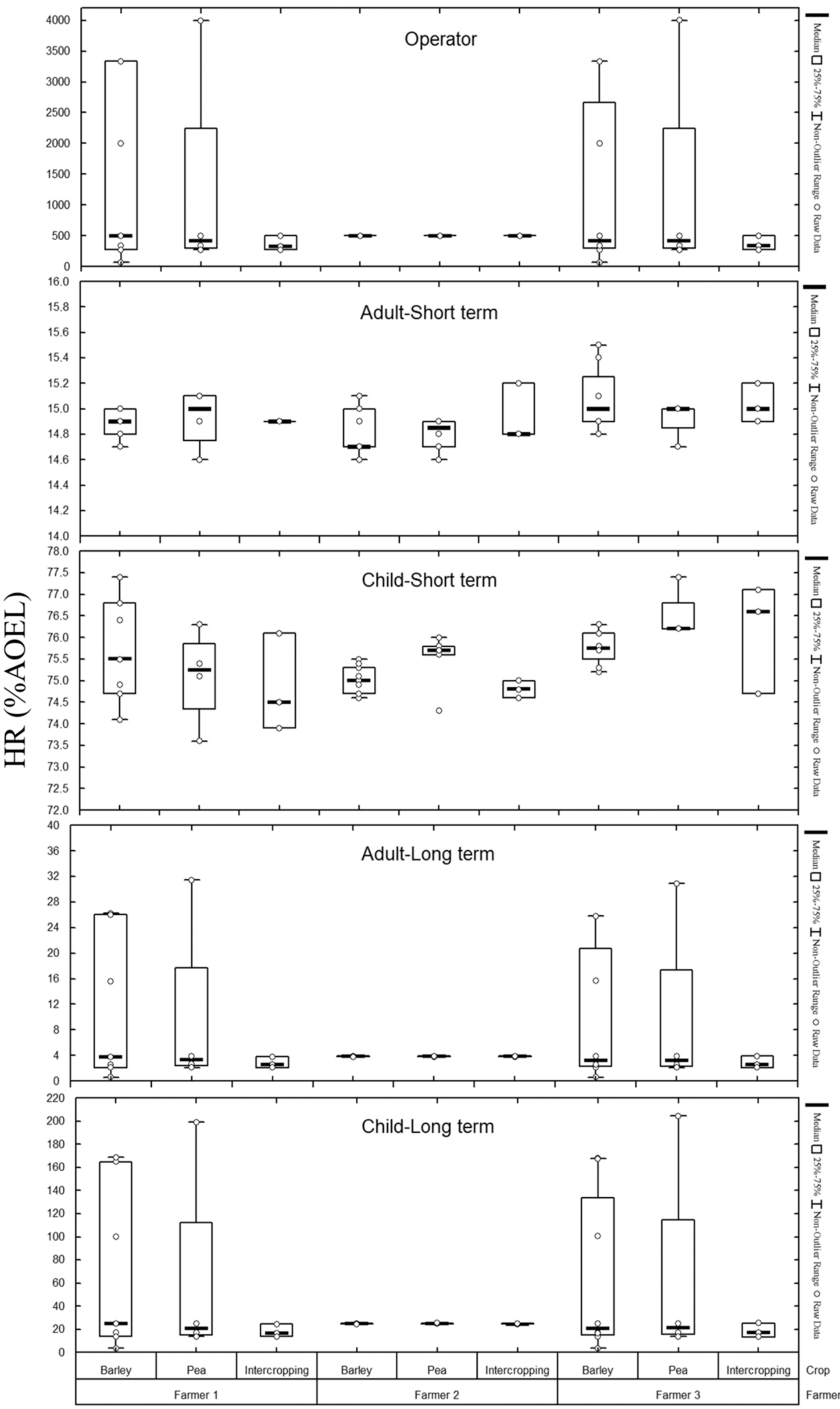

Fig. 4. Distribution of the "Human health risk index" (HR, % of the AOEL) calculated as the ratio of the amount absorbed to the AOEL for each pesticide applied to barley monocultures, pea monocultures, and barley-pea intercropping in no tillage systems.

assess the human health risks from pesticide use in 9 contrasting cropping systems under 3 different farming practices. The values of the parameters used for the BROWSE scenarios were carefully selected to represent farmers' particular real scenario. The main objective of this study was to assess the pesticide exposure and human health risks from pesticide use under different conventional and innovative cropping systems. These cropping practices are commonly adopted on three representative farms in Mediterranean legumes-cereals agro-system.

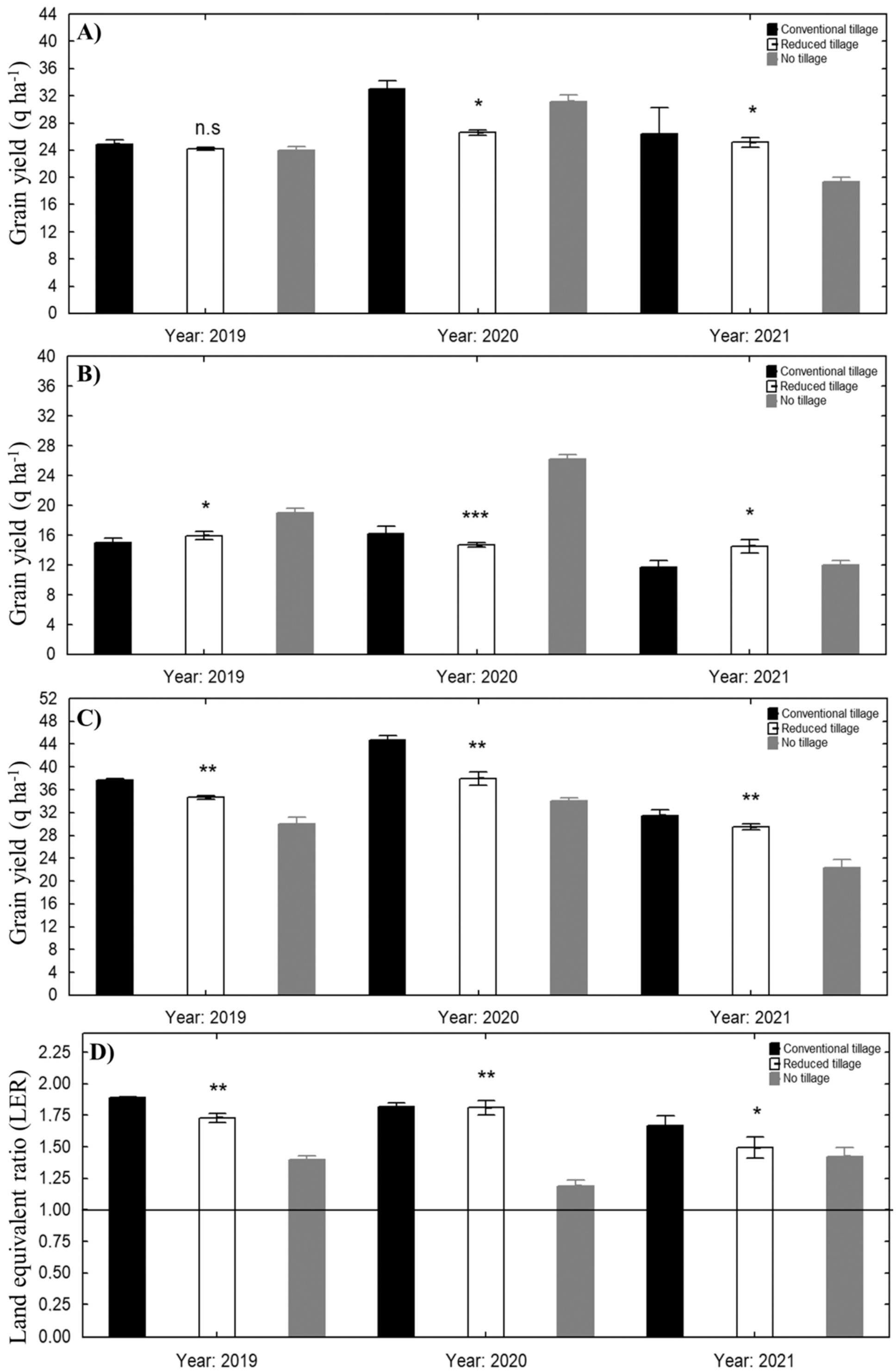

Fig. 5. Grain yield of barley monoculture (A), pea monoculture (B), and barley-pea intercropping system (C), and land equivalent ratio (LER) for grain yield (D) in conventional, reduced, and no tillage systems. Data are means and SE of 3 replicates (farmer 1, 2, and 3). *, ** and *** denote significant differences at $p < 0.05$, $p < 0.01$ and $p < 0.001$, respectively.

Our main results demonstrate that the BROWSE model could accurately reproduce a wide range of arable cropping systems (i.e. monoculture and intercropping system) that are associated with the use of various tillage practices (conventional tillage, reduced tillage, and no tillage).

In the current study, the fields under no-till or reduced tillage practices exhibit higher pesticides use, as they are needed to control weeds, which are otherwise removed with tillage (Alletto et al., 2010). On the conventional tillage systems, the farmers in the Setif area always applied less than three plant protection products containing the following active substances: Tebuconazole and Fludioxonil (fungicides) and Tribuneron-methyl, Prometryne, and Fluasifop-P-butyl (herbicides) (Supplementary Table S1). Conversely, to better control weed development, the farmers in the Setif area applied a wider range of pesticides on untilled plots including the following active substances: Tebuconazole, Fludioxonil, Glyphosate, Iodo-sulfuron-methyl-sodium, Amidosulfuron, Fenoxaprop-P-ethyl, Diclofop-methyl. All of these active substances have been approved since 2009 by the European Commission (European Commission, 2015). In addition, the following active substances are used in pea monoculture - Lambda-cyhalothrin, Prometryn, Fluasifop-P-butyl, and Thiamethoxam - and barley monoculture - Mefenpyr-diethyl - even if they have not yet been approved by the EFSA commission (Cabera and Pastor, 2021). Since human exposure to pesticides is closely linked to the number and frequency of pesticides used (Lammoglia et al., 2017c), human exposure to pesticides in the area of study systematically increased with conservation tillage (no tillage and reduced tillage) (Fig. 1). Human exposure to pesticides in the area of study also increased with monocultures while human exposure was significantly lower in barley-pea intercropping systems. This was probably due to the limited number of pesticides applied in barley-pea intercropping systems and the low concentration of the active substances. Indeed, cereal-legume intercropping is considered one of the most effective cropping systems to reduce weed pressure (Glaze-Corcoran et al., 2020). However, in the barley-pea intercropping systems, exposure observed under no tillage was higher than exposure observed with the other tillage systems due to the application of glyphosate. Previous literature reported a potential health hazard of no-tillage farming systems because of the greater use of pesticides with active substances such as glyphosate and lambda-cyhalothrin (Da Silva et al., 2021; Giuliano et al., 2016). These results are in line with those of Lammoglia et al. (2017c), who confirmed an acceptable human health risk with conventional cereals cultivated in either monoculture or rotation with other crops (i.e., legumes and cereals) under very low input cropping systems. For all the cropping systems investigated in this study, the application of plant protection products was achieved under low input conditions without irrigation and with low N-fertilisation (Table 1).

In terms of agronomic performances, the advantage of barley-pea intercropping was typically confirmed under all tillage systems (Fig. 5). Conventional intercropping was the most efficient system in terms of both yield and LER compared to untilled monocropping systems. Legumes are known to improve yield stability, N acquisition, grain yield, and water use when they are grown in intercropping, more particularly with cereals (Kherif et al., 2020; Kherif et al., 2021; Latati et al., 2019). Cowden et al. (2020) reported a significant advantage of barley-pea intercropping due to the complementarity of the component species under low N soil conditions.

The present results also revealed that human exposure to pesticides and the corresponding risks for human health are generally linked to the TFI in the case of the cropping systems studied. The highest TFI were observed in monocultures with no tillage compared with conventional intercropping systems (Table 2). Furthermore, operators and children were the most exposed populations to pesticide across all the cropping systems. For operators, the highest exposure observed could be explained by their direct exposure to concentrated pesticides during preparation (including mixing and loading) and application (Damalas and Koutroubas, 2016). The highest exposure of children is primarily due to the difference in their body weight compared to adults (Lammoglia et al., 2017c).

Regardless of the tillage practices, dermal absorption was the most dominant pesticide exposure route. It represented more than 98% of the total amount of pesticides applied in all the cropping × tillage system combinations (Fig. S1). Our findings are consistent with other modelling or field studies. Wang et al. (2020) evaluates the handler's exposure to pesticides from stretcher-type power sprayers in orchards. They found that inhalation exposure was negligible compared with dermal exposure, and that hands were the most exposed body part. Using exposure data from the Agricultural Handler Exposure Database (AHED) and the Pesticide Handler Exposure Database (PHED), Pouzou et al. (2018) created probabilistic estimates of exposures. Even if the dermal route of exposure contributes to most of the total doses most of the time, in some of their simulated cases, the amount of pesticides absorbed via dermal route was exceeded by the amount of pesticides absorbed via inhalation (Pouzou et al., 2018). While empirical statistical models consider a linear relationship between the amount of pesticide applied and the amount of exposure, the probabilistic estimations highlighted the variability of the relative contribution to the dose between dermal and inhalation exposure. There are very few models able to produce probabilistic estimates of human health risk and exposure to plant protection products, since traditional models/databases used to generate summary statistics for occupational risk assessments of pesticide handlers in official calculations (Van den Berg et al., 2016). The BROWSE simulation also showed that exposure by ingestion was low (only 2%) and only detected for operators; it was not observed in the resident populations. Dermal exposure on the hands may result in exposure by ingestion through hand-to-mouth contact, especially for farmers (operators) that are routinely exposed to pesticides (Butler Ellis et al., 2017b). However, it should be noted that the BROWSE model is a more conservative model than other risk assessment models, and the predicted amounts of pesticides absorbed might be overestimated. Indeed, BROWSE usually predicts higher potential bystander and resident exposure than other existing regulatory models (Butler Ellis et al., 2017b).

Additionally, the availability of advanced crop protection equipment (cabin with filtered ventilation) was associated with a significant decrease in exposure and human health risk for both operators and residents (see farmer 2, Table S1). In assessing human health risk related to pesticide use, this study highlights the importance of considering the working patterns typical for the region where the pesticide is to be used. Farm operators' exposure to pesticides is influenced not only by the properties of the compound but also by a range of factors, including agricultural and environmental factors, protection measures (e.g. application equipment and personal protective equipment) and physical characteristics of the farmers (e.g. body weight) (Figs. 2–4 and Table S1).

According to the recent literature, only one study has focused on risk assessment of overall pesticide use on human health in innovative (no tillage, rotation, cover crops, and intercropping) and traditional (conventional tillage and monoculture) cropping systems (Lammoglia et al., 2017c). However, in the aforementioned study, the different conventional and innovative cropping systems used to assess the human health risk (HR) varied considerably in terms of the number and composition of the crop species. This could influence pesticide application in terms of treatment frequency, type, and number of pesticides applied in each cropping system. Consequently, the human health risk will automatically be different between the cropping systems studied. A strength of our study is that it assesses and compares human health risks from pesticides applied in different cropping systems with (i) the same crop composition, (i) the same climate, and (iii) relatively similar soil conditions (Table 1). This enables a better, more effective comparison of these systems. Furthermore, in this study, we estimated the agronomic performance (grain yield and LER) of all the cropping systems studied on each farm during three successive growing seasons (2019, 2020, and 2021). This helped us determine whether the performances of these cropping systems in terms of reducing human health risk agree with those of agronomic issues.

The limitations of the present modelling research include the reliance on the inherent equations and underlying data deployed in the BROWSE model. The predefined constants and predictive models currently included in the BROWSE model are based on data generated in Northern Europe and North America and might not reflect the South Mediterranean conditions. Therefore, there is a need for validation of BROWSE exposure estimations against biological monitoring and field measurement in Algeria. Studies to measure human exposure have been conducted using a range of methodologies to determine potential exposure of humans to pesticides in Algeria (Slimani et al., 2011; Moussaoui et al., 2012). It might be useful to upload additional exposure data in the BROWSE model to allow an update of the mechanistic model prediction.

5. Conclusion

The main objective of this modelling study was to evaluate and compare pesticide exposure and human health risks for operators and residents in nine contrasting cereal-legume cropping systems under three different farming practices using the BROWSE probabilistic risk assessment model. This study is the first to demonstrate the performance of innovative cereal-legume intercropping in terms of the risks to human health of overall pesticide use. The human health risk assessment based on the BROWSE model was considered recently as a valuable approach to identify innovative cropping systems with an acceptable human health risk from pesticide use. The choice of the BROWSE model instead of other empirical models was also appropriate in order to distinguish the exposure and health risks of adults and children at short and long term. This modelling approach was also adopted to propose some improvements to either conventional or innovative cropping systems by identifying the pesticides that induce potential risks. The model predicted clear differences in exposure across farmers driven by variations in pesticide application equipment and personal protective equipment. Dermal exposure was the dominant route for both operators and residents, whereas exposure by inhalation and ingestion represented less than 2%. In general, the total amounts of pesticides absorbed were typically correlated with human exposure across most of the cropping systems studied. The conventional tillage was the only system that led to an acceptable human health risk compared with the corresponding conservation tillage. However, the highest and unacceptable human health risks were systematically found in reduced and no tillage systems, especially with barley and pea monocultures. The results also showed that conventional intercropping systems were the most sustainable cropping systems in terms of both agronomic performances (i.e. grain yield and yield advantage) and human health risk. This system can then be recommended as a good alternative to forage-species-based monoculture systems under conservation tillage. The methodological approach adopted in this study could be extended to a wide range of grain-legume-based intercropping systems, as well as to other innovative cropping systems (i.e., cover crops, rotation, agroforestry-based cropping systems).

Funding information

The authors received financial support from the Directorate General of Scientific Research and Technological Development (DGRSDT-MESRS_Algiers) within the framework of the PRIMA Foundation, call 2019-Section 2—BIODIVERSIFY "Boost ecosystem services through high Biodiversity-based Mediterranean Farming systems"-project. This work was partially funded by the PRIMA Foundation, call 2019-Section 1—GA n.X "Research-based participatory approaches for adopting Conservation Agriculture in the Mediterranean Area—CAMA". This paper has been supported by the RUDN University Strategic Academic Leadership Program (Rebouh Yacer and Latati Mourad: Soil analysis).

CRediT authorship contribution statement

B. Zemmouri: data collection, field experiment management, data analysis and writing original manuscript. **K. Lammoglia:** supervising of modelling work and manuscript revision. **Y.N. Rebouh** and **F.Z. Bouras:** Data collection, manuscript English revision and contributed in the revision of manuscript. **M. Latati:** Research supervisor, participated in manuscript writing, data collection and management, as well, the hypothesis formulation and organisation of modelling work. All authors provided critical feedback on the manuscript, at last gave final approval for publication.

Declaration of Competing Interest

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

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2022.113590.

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