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Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol

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Abstract

Background: In a previous study, we found that 45% of public playgrounds near intensively managed agricultural areas were contaminated with mainly endocrine active pesticide residues in spring. Here, we investigated potential contamination over the course of a year.

Methods: Residue data were analyzed from 96 grass samples collected in spring, summer, autumn, and winter by the South Tyrolean Medical Service in 19 public playgrounds, four schoolyards, and one marketplace located within intensively managed agricultural landscapes. Samples were analyzed for 281 substances using gas-chromatography and mass-spectrometry.

Results: A total of 32 pesticide residues and one preservative agent were found. Almost all of the sites (96%) were contaminated with at least one residue during the year; in 79% of the sites, more than one residue was found. Among the detected residues, 76% are classified as endocrine active substances, with the highest concentrations of the insecticide chlorpyrifos-methyl (0.71 mg kg^{-1}), the herbicide oxadiazon (0.64 mg kg^{-1}), and the fungicides captan (0.46 mg kg^{-1}) and fluazinam (0.23 mg kg^{-1}). The number of residues, their concentrations, and the proportion of contaminated sites varied across seasons ($p < 0.001$). Twenty-five residues were found in 83% of the sites in spring (median concentration 0.240 mg kg^{-1}), nine in 79% of the sites in summer (0.092 mg kg^{-1}), three in 50% of the sites in autumn (0.076 mg kg^{-1}), and four in 17% of the sites in winter (0.155 mg kg^{-1}). Playgrounds already examined in 2017 in the previous study, were more often contaminated with multiple pesticide residues in 2018 ($p = 0.045$).

Conclusion: This study confirms previous findings of widespread pesticide contamination of public sites within intensively managed agricultural areas. Moreover, pesticide residues were also found in periods with little or no pesticide application in the field (autumn and winter). It is worrisome that many of the detected residues are endocrine active substances and that some of them (thiacloprid, bupirimate, captan, folpet) are “suspected human carcinogens”, according to EU authorities. Thus, we call for more effective controls of pesticide applications to minimize pesticide drift into public places.

Keywords: Agrochemicals, Non-target area, Pesticide drift, Exposure, Seasonal variation, Intensive agriculture, Apple production, Vineyards, Endocrine active substances, Maximum residue levels (MRL)

Highlights

- Grass samples from 24 public sites in South Tyrol (Italy) were contaminated with a total of 32 pesticides and one preservative agent.

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- At least one residue was found at 96% of the sites across all seasons of a year; more than one residue was found at 79% of the public sites.
- Fungicides (fluazinam, captan) were most abundant; 76% of the residues were classified as endocrine active, among them the insecticide chlorpyrifos.
- Contamination was highest in spring, with a maximum residue load of 0.96 mg kg⁻¹. Some of the pesticide concentrations measured exceeded European maximum residue levels for food.

Background

Worldwide pesticide use is constantly increasing from 0.49 kg ha⁻¹ cropland in 1961 to 2 kg ha⁻¹ in 2004 [1]. It is expected that in 2020 global use of pesticide active ingredients will reach about 3.5 million tons [2, 3]. This trend results in a continuous increase in the environmental pesticide load and thereby the risk of human exposure via water, air, soil, and food [1, 4–6].

Due to a high proportion of pesticide-intensive fruit and wine production, Italy's pesticide use is about four times higher than the European average [7–9]. The Italian Trentino-Alto Adige Region and the Autonomous Province of Bolzano-South Tyrol report for 2018 an average use of 24.1 kg ha⁻¹ for fungicides, 13.0 kg ha⁻¹ for insecticides/acaricides, and 0.8 kg ha⁻¹ for herbicides, while the country-wide averages for Italy in 2018 were 3.55 kg ha⁻¹, 0.6 kg ha⁻¹ and 0.88 kg ha⁻¹, respectively [10]. Moreover, due to the limited availability of cultivable and habitable land in the mountainous province of Bolzano-South Tyrol, apple orchards and vineyards often directly border on public places such as playgrounds, parks, schools, or private gardens [11]. Hence, the amount of non-target pesticide contamination in public places has been shown to correlate with the distance to apple orchards and their proportion in the landscape [12]. However, drift and atmospheric degradation of pesticides also depend on the physico-chemical properties of the pesticide product, droplet size during spray applications, and meteorological conditions during and after spraying [12–16].

Knowing the pesticide contamination of playgrounds is important as they are often visited by children or pregnant women, who are considered particularly vulnerable [17]. Our previous finding, that most of the pesticides detected at playgrounds in South Tyrol are endocrine active (EDA) or endocrine-disrupting chemicals (EDC) [12] is worrisome because EDAs and EDCs affect the hormone balance of fetuses, children, and teenagers [18–21]. Moreover, residents living near agricultural areas with intensive pesticide use have been shown to develop several health deficiencies, including asthma, confusion,

anxiety, weakness, problems with speaking and orientation, poor memory, frequent urination, and increased salivation [22]. Chronic pesticide exposure can also affect the neurological and behavioural development in children resulting in altered reflexes, attention deficit hyperactivity, and psychomotor and neurological development disorders [5, 23]. Importantly, even low levels of pesticide exposure over years can affect children's development, health, and behavior [23, 24].

In a previous study, we already demonstrated the diffuse low-level exposure of playgrounds during springtime [12], but it is unclear if exposures remain for the rest of the year. The present study thus aimed to assess year-round exposure levels of public sites in the Italian province of Bolzano—South-Tyrol, which is among the leading apple- and wine-producing areas in Europe [25].

We hypothesized that: (i) higher pesticide exposure would mainly occur in spring and summer when pesticides are actually applied, while in autumn and winter less to no pesticide residues would be expected [26]; (ii) public sites closer to intensively managed agricultural fields would be more often and higher contaminated than sites further away, and that (iii) pesticide contamination levels would be similar in spring 2017 and spring 2018.

These hypotheses were tested using data of residue levels of public sites across agricultural landscapes over the course of a year collected and analysed by regional authorities.

Materials and methods

Sampling and site description

After surveys in 2016 [27] and 2017 [12, 28], the Department of Environmental Medicine of the South Tyrolean Medical Service (Departement für Gesundheitsvorsorge – Betriebliche Sektion für Umweltmedizin des Südtiroler Sanitätsbetriebs) conducted in 2018 a further survey on residue contamination in the Autonomous Province Bolzano—South Tyrol (Italy). For the current study, these data were courteously provided by the South Tyrolean Medical Service.

Sites were selected and sampled by trained staff of the regional authority in 17 rural municipalities. The agricultural production areas included mainly apple orchards, vineyards, grasslands, and fields with vegetable crops. The samplings included 19 playgrounds, four schoolyards, and one public market place, and are referred to hereafter as public sites. Sampling was conducted between May 2018 and March 2019 on four sampling dates (spring, summer, autumn, and winter) making up a total of 96 samples. Fourteen of these playgrounds were already investigated in the previous study in spring 2017 [12].

In accordance with our previous results [12], sampled public sites were classified by their distance from the closest apple orchards and vineyards, hereafter called risk sites with < 100 m distance and control sites with ≥ 100 m distance. The distance was measured from the center of the sample site to the first agricultural plants of the agricultural sites. The final site selection included 7 sites in the Vinschgau Valley, 8 in the Etsch Valley, 6 in the Lower Etsch Valley, and 3 in the Eisack Valley, resulting in a total of 20 (83.3%) risk sites and 3 (12.5%) control sites. For one site no distance could be assessed due to unclear measurement. Geographical data obtained from the regional geographic information system Geobrowser (Autonomous Province Bozen/Bolzano—South Tyrol, 2017), included elevation and distance to the nearest orchards. To relate residue levels to precipitation amount within 5 days prior to sampling, data were obtained from the nearest meteorological stations [29].

Further, current results were compared with maximum residue limits (MRL) of lettuce, spinach and strawberries, based on the EU MRL database [30]. This quantitative evaluation was chosen because we assumed that similar numbers and amounts of residues would be detected in vegetables and fruits growing in private gardens and farms located at equal distances from agricultural areas as the investigated public sites [30].

Sampling and laboratory analysis

Access to the 96 laboratory reports including information on sampling and analysis was granted upon official request for "General Citizen Access" at the office for Institutional Affairs of the Autonomous Province of Bolzano-South Tyrol [31] on 14th December 2019.

Concretely, per site, a minimum of five selected grass samples was taken as representative of the sample site and grouped into one composite sample of about 300–400 g fresh mass [12]. Sampling was conducted 4 times between May 2018 and March 2019: first in spring 2018 between May and June, second in July 2018 (pre-harvest time), third in late summer/autumn 2018 between August and October (harvest time), and fourth in winter between January and March 2019, when no pesticide application is assumed [26, 32].

Plant material was analyzed by the official and accredited Laboratory for Food Analysis and Product Safety of the Autonomous Province of Bolzano-South Tyrol (Landesagentur für Umwelt und Klimaschutz – Labor für Lebensmittelanalysen und Produktsicherheit). Briefly, after acetonitrile extraction (701881-acetonitril for LC-MS, ITW Reagents PanReac AppliChem) according to the dispersive SPE—QuEChERS-method [33], residue concentrations of grass samples were analyzed using a triple quadrupole GC/MS series system (Agilent 7000C,

Waldbronn, Germany) and API 4000™ LC-MC/MS series system (AB Sciex, Concord ON, Canada), following the European standards UNI EN 15662:2009 and 15662:2018 [34], to identify up to 281 substances. Measured residue concentrations were based on the wet weight of fresh grass samples. Only active compounds were analyzed; no account was taken of metabolites and adjuvants [12, 35]. Unfortunately, the official list for pesticide monitoring performed by the South Tyrolean authorities did not include the frequently used herbicide glyphosate. However, neonicotinoid insecticides such as thiacloprid and imidacloprid were included.

Statistical analyses

Continuous variables were presented as medians with IQR (interquartile range) and categorical variables as percentages. Descriptive statistics and univariate test statistics were performed using RStudio (Version 1.3.1073) [36] including the packages doBy, psych, jtools, PMCMR, dplyr and tigerstats [37–42]. Chi-squared test and Fisher's exact test were applied for the discrete variable *number of detected pesticides* and independent 2-group Mann–Whitney-*U* test for zero-inflated continuous data on residue concentrations between risk sites and control sites, and between contaminations of spring 2017 and spring 2018. The Kruskal–Wallis test and, if major effects were found, post hoc test for multiple comparisons of mean rank sums (Nemenyi test) were used to compare residue data among the four different sampling dates. Bonferroni correction method for multiple testing was used for post-hoc pairwise tests of continuous data (Nemenyi test) and for multidimensional contingency tables of count data.

The sum of rainfall (mm) was calculated for a 5-day period before the sampling date and correlated with the number and concentration of fungicides. A two-sided *p* value of < 0.05 was considered statistically significant.

The distance categories "near" (< 100 m) and "far" (≥ 100 m) were first computed—in accordance with our previous results—and additionally for the terciles of the distance measurements. Differences in medians of the distance groups were analysed with Wilcoxon rank-sum tests with continuity correction respectively with Kruskal–Wallis rank-sum test for the three distance categories.

Results

In total, across all study sites and seasons, we found 32 pesticides and one preservative agent (2-phenylphenol). The detected pesticides across all four sampling sites included 24 fungicides, seven insecticides, and one herbicide (Table 1). In 23 of the 24 sites, at least one pesticide was detected over the entire year. In particular, over the

Table 1 Characteristics of residues detected in the current study: Pesticide category and active ingredient, substance class, frequency of detection (Freq.) and maximum concentration (Max. conc.) in current samples, categorisations regarding carcinogenicity (Carc.), endocrine disruptor/activity (EDC), volatility and predicted mean dissipation half-life (HL; upper CI for residues with wide HL ranges in brackets)

Pesticide category/ detected residue	Substance class	Freq.	Max. conc. (mg kg ⁻¹)	Carc. [56]	EDC ^a [94, 95]	Volatility ^b [94]	HL ^c [59]
Herbicides							
Oxadiazon	Oxidiazole	1	0.64	–	Cat1	Low	3
Insecticides							
Chlorpyrifos	Organophosphate	2	0.71	–	Cat3	Low	4
Chlorpyrifos-methyl	Organophosphate	4	0.06	–	Cat2	Low	3
Imidacloprid	Neonicotinoid	5	0.03	–	Cat3	Low	4 (29)
Methoxyfenozide	Diacylhydrazine	5	0.03	–	Cat2	Low	7
Phosmet	Organophosphate	8	0.68	–	Cat3	Low	2
tau-Fluvalinate	Pyrethroid	1	0.01	–	Cat3	Low	3
Thiacloprid	Neonicotinoid	2	0.01	2	Cat3	Low	4
Fungicides							
Ametoctradin	Triazolopyrimidine	1	0.01	–	–	Low	–
Boscalid	Carboxamide	1	0.04	–	Cat1	Low	7
Bupirimate	Pyrimidinol	1	0.03	2	Cat3	Low	–
Captan	Phthalimide	22	0.46	2	Cat3	Low	5
Cyflufenamid	Amidoxine	2	0.01	–	–	Low	–
Cyprodinil	Anilinopyrimidine	3	0.02	–	Cat3	Low	6
Difenoconazole	Triazole	10	0.12	–	Cat3	Low	5
Diphenylamine	Amine	2	0.51	–	–	Low	–
Dodine	Guanidine	12	0.23	–	–	Low	3
Fluazinam	Phenylpyridinamine	19	0.24	–	Cat2	Low	4 (69)
Fludioxonil	Phenylpyrrole	9	0.04	–	Cat3	Low	5
Fluxapyroxad	Pyrazolium	4	0.09	–	–	Low	–
Folpet	Phthalimide	5	0.15	2	Cat3	Low	5
Meptyldinocap	Dinitrophenol	5	0.15	–	Cat2	Low	3
Penconazole	Triazole	3	0.04	–	Cat3	Low	8
Penthiopyrad	Carboxamide	2	0.10	–	Cat2	Low	–
Propiconazole	Triazole	1	0.05	–	Cat2	Low	5
Pyraclostrobin	Strobilurin	1	0.01	–	Cat3	Low	4
Pyrimethanil	Anilinopyrimidine	1	0.03	–	Cat3	Low	5
Quinoxifen	Quinoline	2	0.02	–	–	Low	6 (110)
Spiroxamine	Morpholine	1	0.02	–	Cat2	Low	10
Tebuconazole	Triazole	1	0.04	–	Cat1	Low	8
Tetraconazole	Triazole	1	0.01	–	Cat1	Low	5
Zoxamid	Benzamide	1	0.02	–	Cat3	Low	–
Preservative agent							
2-phenylphenol	Phenole	1	0.02	–	–	High	–

No data available/not listed = "–"

^a ED category 1: confirmed in animal studies, ED category 2: in-vitro study confirmed, ED category 3: endocrine active or potential disruptive, according to the structure of chemical substance and biochemical properties

^b Renewal Assessment Reports (RARs) including vapor pressures documented by the Reporting Member State in the process of EU approval for pesticides

^c Mean predicted dissipation HL for residues were rounded to full days. Additionally, very high maximum HLs (>25 days) are listed in brackets

course of the year, 20 sites showed at least one instance of contamination with 2–3 residues, (4 of these sites more than once a year) and 11 sites were contaminated with 4 or more residues. A maximum of 11 residues was detected at a playground in the Lower Etsch Valley in the village of Neumarkt in a 40 m distance from an agricultural field. One playground in Leifers (Marconistrasse) was located 25 m from an agricultural field and was contaminated with at least one pesticide the entire year. Only one playground in the city of Bolzano (playground Firmian) located at 40 m distance from an agricultural field showed no pesticide contamination at any of the sampling events.

The pesticides detected most frequently were the fungicides captan (one site in spring, 12 in summer, and 9 in autumn), followed by fluazinam (7 sites in spring and 12 in summer), dodine (two in spring, 10 in summer), and difeconazole (10 sites in spring). The highest concentrations were found for chlorpyrifos-methyl (0.71 mg kg⁻¹), oxadiazon (0.64 mg kg⁻¹), captan (0.46 mg kg⁻¹) and fluazinam 0.23 mg kg⁻¹) (Table 1).

The majority of the detected residues (76%, 25 out of 33) are classified as endocrine active (EA) substances: in particular, 4 pesticides belong to category 1 (endocrine disruptor ED as confirmed in animal studies [43–45]; 7 belong to category 2 (ED in vitro confirmed); and 15 are endocrine active substances or potential EDs belonging to category 3 (potential EA according to chemical structure and biochemical properties). Only 7 (21%) of the detected residues are without any known endocrine activity [1, 43, 46] (Table 1).

Summarized concentrations of all EA substances on the investigated public sites were up to 1.12 mg kg⁻¹ per grass sample, with a mean of 0.34 mg kg⁻¹ (Table 2).

Seasonal pesticide contamination

Pesticide contamination of sites differed significantly among the four seasons. The percentage of contaminated sites (83%) and the number of detected residues (25 out of 33, Figs. 1, 2 and Additional file 1: Table S1) were significantly higher ($p < 0.001$) in spring compared to the other seasons. Median residue concentration was also higher in spring (0.24 mg kg⁻¹, IQR = 0.28 mg kg⁻¹,

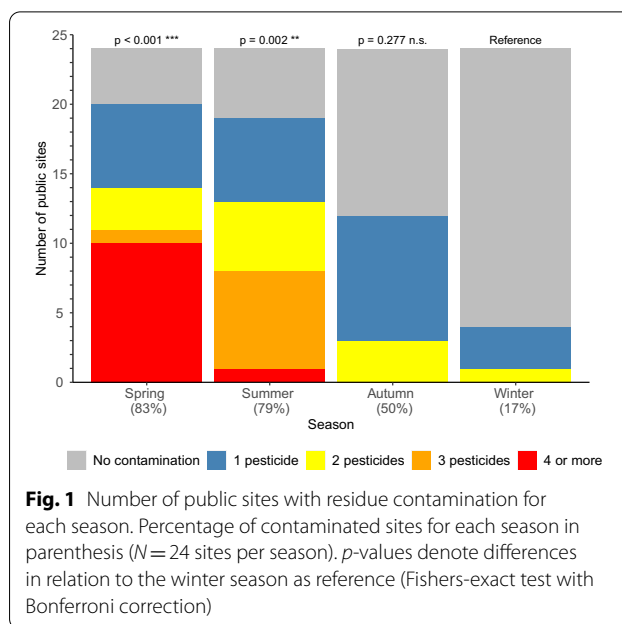


Fig. 1 Number of public sites with residue contamination for each season. Percentage of contaminated sites for each season in parenthesis ($N = 24$ sites per season). p -values denote differences in relation to the winter season as reference (Fishers-exact test with Bonferroni correction)

max = 0.96 mg kg⁻¹), but did not differ significantly from other seasons when including only the contaminated samples ($p = 0.128$, Kruskal–Wallis test, 2). However, including all samples, the seasons spring, summer and autumn differed significantly in pesticide load compared to residue loads detected in winter (Additional file 1: Table S2).

Over the course of a year, repeated contamination was observed for seven sites: On three sites decreasing concentrations of fluazinam were detected from spring to summer; five sites showed rising concentrations of captan from summer to autumn and one site had similar concentrations of dodine in spring and summer. Two sites showed repeated contamination with several residues mainly with captan and fluazinam or fluazinam and dodine (Fig. 3).

Distance from agricultural sites

Total median pesticide concentration was not significantly higher at public sites < 100 m distance from agricultural fields (median 0.02 mg kg⁻¹, IQR 0.1 mg kg⁻¹,

Table 2 Pesticide loads (mg kg⁻¹ grass) on contaminated public sites per season

Parameter	Spring ($N_c = 20$)		Summer ($N_c = 19$)		Autumn ($N_c = 12$)		Winter ($N_c = 4$)	
	Med. (IQR)	Max	Med. (IQR)	Max	Med. (IQR)	Max	Med. (IQR)	Max
Pesticide load	0.24 (0.28)	0.96	0.09 (0.13)	0.52	0.08 (0.07)	0.21	0.01 (0.03)	0.09
Fungicide load	0.18 (0.21)	0.4	0.09 (0.05)	0.52	0.08 (0.07)	0.21	0.01 (0.04)	0.09
EDA load	0.21 (0.25)	0.94	0.08 (0.13)	0.51	0.08 (0.07)	0.21	0.01 (0.03)	0.09

Medians (Med.), Interquartile range (IQR) and maximum concentrations (Max), $N = 24$ sites per season. No differences in concentrations for the contaminated sites across the four seasons were found. Including all sites, the seasons, spring and summer differ significantly from concentrations detected in winter

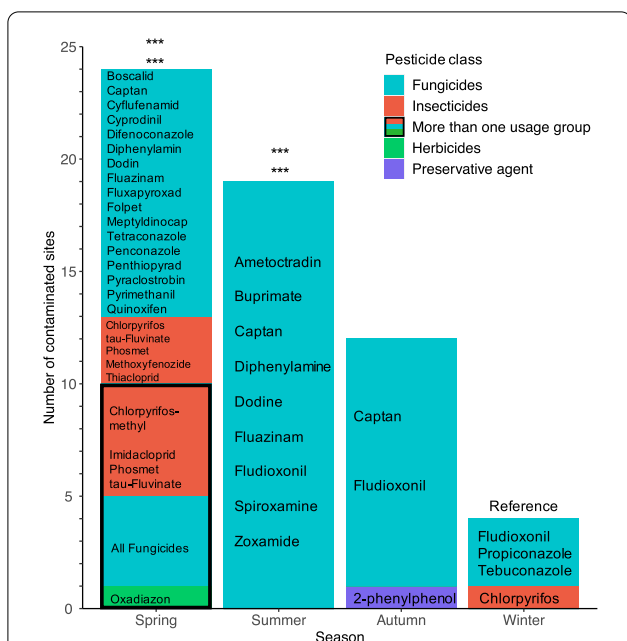


Fig. 2 Number of contaminated sites with different pesticide classes (and preservative agent) in the four seasons. Names of detected residues across all sites within a season are listed for each pesticide class; not all residues were found at each site. Total number of residues = 33. Significance values *** ($p < 0.001$, adj. Fisher’s exact for multiple comparisons across seasons and within seasons) denote the difference between the total number of residues and number of EDA residues for spring and summer compared to winter

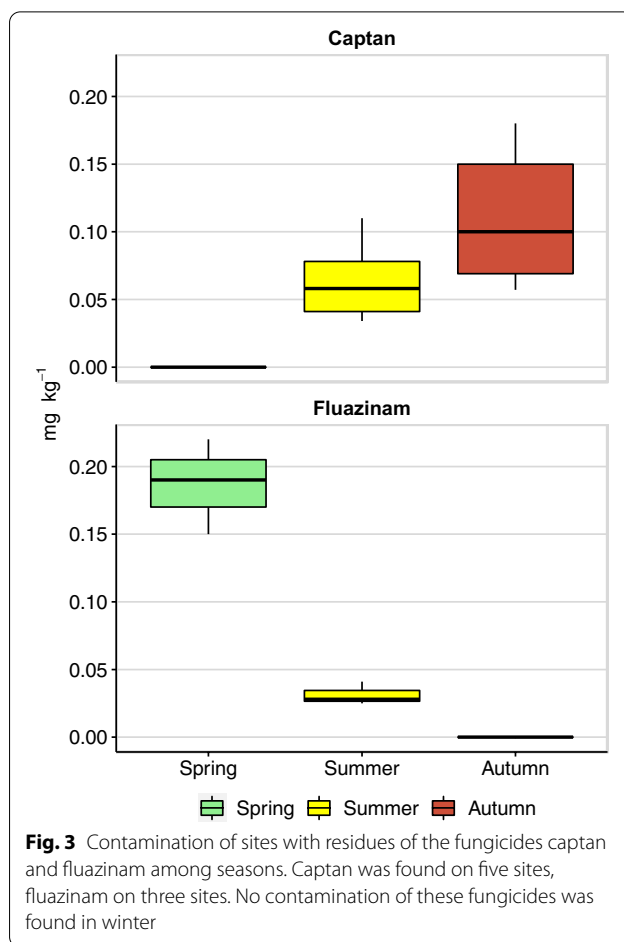


Fig. 3 Contamination of sites with residues of the fungicides captan and fluazinam among seasons. Captan was found on five sites, fluazinam on three sites. No contamination of these fungicides was found in winter

$n=20$) compared to sites >100 m away (median 0.0 mg kg^{-1} , IQR 0.04 mg kg^{-1} , $n=3$, Wilcoxon rank-sum test with continuity correction, $p=0.156$). There was also no difference between medians of pesticide loads for the tercile categorization (15–30 m, 31–40 m, 41–600 m) (Additional file 1: Figures S1 and S2).

Pesticide contamination of playgrounds in spring 2017 versus spring 2018

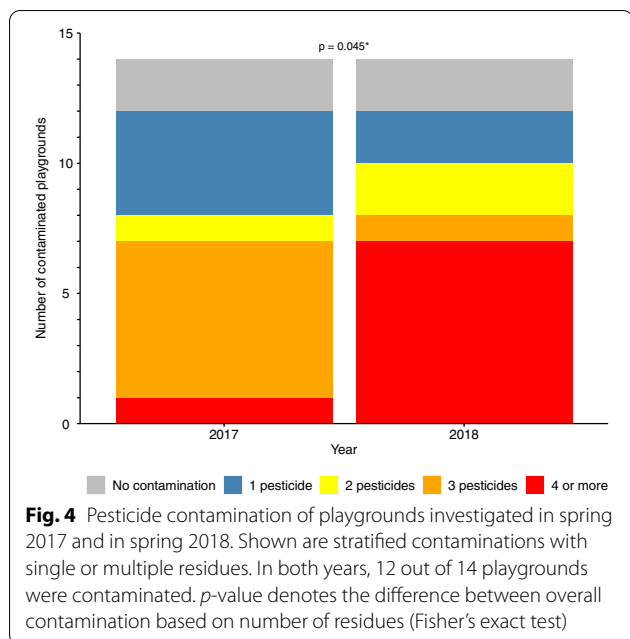
Both, in spring 2017 and spring 2018, 12 out of 14 investigated playgrounds were contaminated. However, in spring 2018 significantly more playgrounds were contaminated with multiple pesticides than in spring 2017 ($p=0.045$, Fig. 4). The median number of contaminated playgrounds was also significantly higher in spring 2018 (median=3.5, IQR=4.8, max=11) as in spring 2017 (median=2, IQR=1.8, max=4, paired Mann–Whitney U , $p=0.01$). Considering particular pesticides, more playgrounds were contaminated with difenoconazole in spring 2018 (7 sites) than in spring 2017 (1 site, Fisher’s exact test, $p=0.033$, Additional file 1: Table S6). Also, the concentration of fluazinam was higher in spring 2018 (med.=0.19, IQR=0.07) than in spring 2017 (med=0.03, IQR=0.04, Mann–Whitney U , $p=0.039$).

Total pesticide load (mg kg^{-1}) did not differ significantly between years.

The amount of rainfall 5 days before sampling in spring 2018 (median=10.6, IQR=10.9) was higher than in spring 2017 (median=5.8, IQR=7.8) but the difference was not significant ($p=0.069$). Pesticide concentration across sites did not significantly correlate with the amount of rainfall (Pearson correlation, $r=0.11$, $p=0.063$).

Comparison of residues in grass to MRLs in vegetables and fruits

Current residue levels were compared with MRLs for food items typical for home gardens—lettuce, spinach, and strawberries [30]. Comparing the residues found in the grass samples with the MRLs for these three food items, the MRLs for chlorpyrifos would have been exceeded up to 71-fold, for fluazinam up to 24-fold, for dodine up to 23-fold, for captan up to 15-fold, for folpet up to fivefold and for meptyldinocap up to threefold (Table 3).



Discussion

To the best of our knowledge, the present study and our previous one [12] are the first to demonstrate pesticide contamination of public sites in agricultural landscapes using grass samples. For soil samples, pesticide contamination of playgrounds was reported in one European

[17] and one US study [47]. The US study [47] exclusively focused on residues of persistent organochlorine pesticides which have been banned since the 1970s, while the European study performed on playgrounds in Sarajevo [17] investigated also the variety of contaminants reported for public sites in South Tyrol. The soil samples of these studies were contaminated with up to 42 different pesticide residues with concentrations up to 0.1 mg kg⁻¹ soil for Europe and up to 0.06 mg kg⁻¹ for playgrounds in the USA [17, 47]. We argue that pesticide residues of grass samples might give a more immediate picture of actual pesticide contamination and off-site drift than soil samples due to possible faster degradation of residues on the surface. Our previous study showed that irradiance can be a driving factor to lower pesticide contamination by photodegradation [12, 48, 49].

Concerning pesticide classes and concentrations, the findings of the current study are comparable with an Austrian study sampling field margins near apple orchards [50] showing that 73% of the detected residues were fungicides, the dominant pesticide class used in intensive apple and wine production. Furthermore, 23 of the detected residues reported in our current study were also detected in a recent monitoring study in Germany using passive air samplers which analyzed >500 substances in 116 sites across Germany [51]. Because glyphosate was the only pesticide found in all German sites we assume that glyphosate might also be present at our study sites

Table 3 Maximum residues (MR in mg kg⁻¹) in grass samples of frequently detected pesticides and surrogate maximum residue limits (MRL) according to EU database [30], assuming similar contamination levels for vegetables and fruits from home gardens

Pesticide	Current MR (grass)	MRL according to EU database [30]			Relationship to lowest surrogate MRL
		Lettuce	Spinach	Strawberry	
Chlorpyrifos	<i>0.71</i>	0.30	0.01	0.01	71-fold over
Fluazinam	<i>0.24</i>	0.05	0.01	0.01	24-fold over
Dodine	<i>0.23</i>	0.01	0.01	0.01	23-fold over
Captan	<i>0.46</i>	0.03	0.03	5.00	15-fold over
Oxadiazon	<i>0.64</i>	0.05	0.05	0.05	13-fold over
Chlorpyrifos-methyl	<i>0.06</i>	0.06	0.01	0.01	Sixfold over
Folpet	<i>0.15</i>	0.03	0.03	1.50	Fivefold over
Penconazole	<i>0.04</i>	0.01	0.01	0.50	Fourfold over
Meptyldinocap	<i>0.15</i>	0.05	0.05	3.00	Threefold over
Phosmet	<i>0.07</i>	0.05	0.05	0.05	1.4-fold over
Imidacloprid	0.03	2.00	0.05	0.05	1.7-fold under
Difenoconazole	0.12	3.00	2.00	0.40	Threefold under
Penthiopyrad	0.10	15.00	30.00	3.00	30-fold under
Fluxapyroxad	0.09	4.00	3.00	4.00	33-fold under
Methoxyfenozid	0.03	4.00	4.00	2.00	67-fold under
Fludioxonil	0.04	40.00	30.00	4.00	100-fold under

Italic print indicates concentrations exceeding surrogate-MRLs. Comparisons were made to the lowest reported MRL

if the samples had been analyzed with the appropriate methods [52].

Origin of detected pesticides

We assume that the main sources of pesticide residue contamination in our study were drifting during application or secondary drift via volatilization and/or dust from soil or plants after application [53, 54]. Of the 33 substances identified in the grass samples in the present study, 97% were approved for use in fruit orchards and vineyards in Italy at the time of sampling [55]. Propiconazole and diphenylamine are not approved for fruit growing, but are approved for general agriculture and are additionally contained in wood preservatives. The origin of the two detected preservative agents (diphenylamine and 2-phenylphenol) is also most likely agricultural, as both are used for post-harvest treatments to control storage disease in apples and citrus fruits. In Italy, diphenylamine is used as an anti-scald agent to avoid skin browning [55]. Diphenylamine is also used for non-agricultural dyes.

Only 10 of the detected pesticides, or 30%, have approval for non-professional users, nine fungicides, and one insecticide [55–57]. Therefore, we assume that the majority of residues in our samples very most likely derived from agricultural applications, though we cannot rule out contributions from other sources.

According to the official risk assessments, all detected residues (except the preservative-agent 2-phenylphenol, Table 1) are characterized by low volatility and thus are expected not to move more than a few meters beyond the application site [58]. However, our results suggest that evaporation from plant surfaces or particle drift may be significant sources for off-site contamination. It is important to point out that the European Food Safety Authority (EFSA) builds its risk assessment on expectations and modeling. Our data show that these models seem to underestimate the real-life situation and we, therefore, plead for a stricter application of the precautionary principle.

Potential impact of pesticide residues on human health

We assume that vegetables and fruits from nearby private gardens are as likely to be similarly contaminated by pesticide drift as the grass on public sites. Hence, we argue that the residue levels in grass samples might also be a valid indicator for overall contamination in public sites and private gardens.

Comparing residue levels in the grass with MRLs for lettuce, spinach, and strawberries, the levels for fluazinam and captan detected in this study, exceeded the surrogate MRLs 24-fold and 15-fold, respectively. Moreover, fluazinam is an endocrine active substance of category 2

[43] and captan is classified as a category 2 carcinogen, according to the EU pesticide database [56].

The multiple detections of fluazinam on three sites on the following sampling dates (spring and summer) at the end of May and the end of July with decreasing concentrations indicate a possible persistence of this residue. The half-life of fluazinam is up to 69 days [59] (Table 1) and the concentration decline from spring to summer was 80% (Fig. 3). Residues of the fungicides captan and dodine did not decline and their half-lives are only between 3 and 5 days, indicating that both are less persistent and that the higher concentrations detected in summer occurred through repeated pesticide application (Fig. 3).

Generally, residue concentrations in grass samples were low, but the year-round detection and the reported contamination of the last years [12, 27, 28] indicate chronic exposure of humans and the environment. Among the detected pesticides, chlorpyrifos and chlorpyrifos-methyl are the most hazardous. Residues of these chemicals were detected in the grass samples of 2018 and in samples from earlier years [12, 27, 28]. Low doses of chlorpyrifos have been shown to lead to brain anomalies in fetuses and children [60], and to affect locomotor activity, behavior, and neurotransmitter systems in rats [61]. Recently a study [62] re-evaluated the low dose effects of chlorpyrifos in developmental neurotoxicity and corrected the misleading results of the original assessment [63]; meanwhile, these two insecticides are banned in the EU.

Overall, 25 of the 33 identified compounds are EDAs [43, 64], which were present in 96% of the investigated sites, while in 53% of them, EA substances were detected all year round. Endocrine disruptive compounds frequently exhibit non-monotonic dose–response relationships and, therefore, can be effective in concentrations several orders of magnitude below common residue thresholds [65]. Relative to their body weight, children inhale more air, drink more, and eat more food than adults, and some activities of children on playgrounds are likely to bring them in contact with contaminants present in the grass or sand [24, 66]. Endocrine active pesticides interfere with oestrogen or androgen receptors [1] and have been linked to an increased risk of thyroid, breast, and prostate cancer [1, 67]. Simultaneous exposure to a variety of pesticides might also trigger synergistic effects [68], which are extremely difficult to examine in risk assessments [69]. Due to the low-dose effect, the non-monotonic dose–response relationship, and the interaction of EDCs with endogenous hormones or other EDCs, it is questionable whether a low dose can ever be considered safe [45, 65]. Moreover, EDCs can cause multigenerational effects [70, 71] even in grandchildren of exposed pregnant women [72].

Exposure of pregnant women and children to endocrine active pesticides—as were detected in our grass samples—and/or their metabolites have been described in several publications. For example, di-ethyl phosphate and 3,5,6-trichloro-2-pyridinol, both metabolites of chlorpyrifos and chlorpyrifos-methyl, were detected in urine samples from children [73] and hair samples from pregnant women [74, 75], as well as cypermethrin, cyprodinil, difenoconazole, imidacloprid, oxadiazon, penconazole, propiconazole, pyraclostrobin, pyrimethanil, tebuconazole, tetraconazole, thiacloprid and zoxamid [74, 76]. Many of the EDCs are fungicides—in this study 75%— which is also the most prevalent group of detected residues in the current sites. In the European Union, fungicide sales represent more than 40% of total pesticide sales, and in areas dominated by fruit growing, such as vineyard regions, fungicides account for more than 90% of all pesticide applications [77].

Fungicides such as propiconazole and tebuconazole have also been identified as potential human health risks by exposure through drinking water [78]. Indeed, contamination of surface water within the study area had been reported for the same pesticides and at similar concentrations as in grass samples from the public sites investigated [79].

Multiple pesticide contamination occurred on 19 sites at least once during the entire year. The effect of mixtures of active compounds and their specific formulations/additives is still not clear. Exposure due to pesticide drift raises the issue of multiple exposures from various sources [80] and the still unknown extent thereof. It is obvious that people living in the study area of this survey are simultaneously exposed to the same pesticides in commercial food, vegetables, fruits, and herbs from private gardens, as well as through inhalation of contaminated air.

Plant protection products consist, besides the active compounds, of specific formulations to enhance the effect of the ingredients. It is likely, for instance, that formulations on tween-basis can have an influence on the mobilization, bioavailability, and bioaccumulation of organochlorine contaminants and its metabolites [81, 82]. However, the concealment of the full list of ingredients in formulations makes a detailed investigation impossible.

Impact on the environment

Besides affecting human health, many of the detected pesticides have a proven impact on various non-target organisms and are in general a driving factor in biodiversity decline [83]. Non-target effects of pesticides affect soil biota [84–86], bees [87, 88], and other organisms important for the functioning of the agroecosystems [89].

A recent biodiversity assessment conducted in the same region as the presented study showed that butterflies are endangered through pesticide exposure [90].

We intended to estimate the association between environmental factors and pesticide residues in public sites, as we did in our previous study [12]. Due to a nearly three-fold higher number of different fungicides detected in spring 2018 compared to our previous study [12] and a higher proportion of public sites contaminated with fungicides, we hypothesized that such findings could relate to the higher rainfall in 2018. However, the relationship between precipitation and detected fungicide residues was not significant, pointing to multifactorial influences.

One public site was without any pesticide residue throughout the year, even though the site is located only 40 m from the closest agricultural area. However, we know from the previous study [12] that, besides chemical drift, the characteristics of the pesticides applied, as well as the prevailing wind direction and wind speed, may have affected the contamination of these public sites.

Our results show that even a distance of more than 100 m to agricultural sites does not lower the contamination level significantly (Additional file 1: Figure S1) and the relationship between residue load and distance is not linear (Additional file 1: Figure S2). Therefore, an evaluation of the extent of exposure requires further studies and a subsequent risk assessment.

There are already sufficient arguments to ban or at least drastically reduce pesticide application. The most important arguments therefor concerning human and environmental health, sustainability, and costs. The impact of chronic pesticide exposure on human health and the environment is proven [1, 89]. All in all, the health costs to the EU caused by endocrine active compounds, mainly pesticides, have been estimated at more than € 150 billion annually [91, 92]. However, sustainable and ecological agriculture is possible and economically. It generates farm incomes exceeding those from conventional and industrial farms, providing more employment and thus supporting regional economies by using less fossil fuel and contributing to preserving biodiversity [93].

Conclusions

Our findings suggest that pesticide contamination of public sites in intensively managed agricultural areas is a year-round potential health issue, in particular for the most vulnerable groups of society, including infants and pregnant women. The long-term effects of exposure to such contamination are entirely unknown.

Our results are particularly critical because the majority of the detected pesticides are known EDAs and several samples contained hazardous compounds such as

chlorpyrifos (recently banned because of its neurotoxicity) or captan (classified in the EU as a “suspected human carcinogen”).

It is likely that other non-crop sites within intensively managed agricultural areas such as private gardens, nature conservation areas, or fields of organic farms are contaminated in a similar way. This is an aspect that is rarely addressed in the scientific and public debate. Our findings also suggest that the guidelines of the European Food Safety Authority (EFSA) concerning the exposure of operators, workers, residents, and bystanders [58] are not sufficiently protective, because they base their risk assessment on volatility, disregarding other mechanisms of drift, e.g., via particulate matter. Therefore, we suggest to include field studies, such as our study, in the risk assessment.

Hence, we recommend an overall reduction of pesticide use and improvements in agricultural practices as precautionary measures to protect human and environmental health from uncontrolled pesticide exposure.

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1186/s12302-020-00446-y>.

Additional file 1. Additional figures and tables.

Abbreviations

EDs: Endocrine disruptors; EDA: Endocrine active; EDC: Endocrine disruptive compounds; MRL: Maximum residue limit/level; IQR: Interquartile range.

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Authors' contributions

CL planned and conducted data analyses, generated tables, and graphs and did the literature search. JGZ, PC, SP, and FB provided literature and substantial expert knowledge. KH organized the study data, did the preliminary data analysis, organized explanatory data and provided literature. All authors read and approved the final manuscript.

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Availability of data and materials

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Ethics approval and consent to participate

Not applicable.

Consent for publication

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Competing interests

The authors declare no financial and non-financial competing interests.

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