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Double the trouble: High levels of both synthetic pesticides and copper in vineyard soils☆

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ABSTRACT

The widespread use of pesticides raises concerns about their impact on soil health. Although vineyard soils are strongly exposed to both, synthetic pesticides and copper, a systematic, detailed, and joint assessment has been lacking. In our study we measured copper and 146 synthetic pesticides in 62 organic and conventionally managed vineyards at high sensitivity.

Up to 60 different pesticides were detected per vineyard. Total pesticide concentrations were almost 13 times higher under conventional compared to organic management. Total copper contamination was high overall with a mean of 371 mg/kg, and no difference between organic and conventional vineyards was found. Pesticide levels declined with increasing years since conversion to organic farming. However, even after 20 years of organic farming, up to 32 pesticides could still be found. Several pesticides showed far higher persistence in soil than expected based on their half-lives. Compared to other land uses, pesticide and copper contamination was clearly higher. Our risk assessment revealed that 50 % of the studied vineyard soils reached pesticide and copper concentrations potentially harmful to soil organisms and only 10 % of vineyard soils were not at risk from either of them. This underscores the urgent need for further research and policy intervention to address these environmental risks.

1. Introduction

Few agriculture-related topics are discussed as controversially as plant protection products. The obvious advantages of increased food production through the suppression of weeds, pests, and diseases (Oerke, 2006) are offset by concerns about human health and environmental hazards (Matthews, 2015). The extensive use of synthetic pesticides (hereafter "pesticides") has led to their widespread occurrence in agricultural and natural terrestrial systems, aquatic systems, and groundwater (Carvalho, 2017). Soil is a primary recipient of pesticides, and an increasing number of studies are unveiling the ubiquitous appearance of pesticide residues throughout European soils (Froger et al., 2023; Geissen et al., 2021; Knuth et al., 2024; Pose-Juan et al., 2015; Silva et al., 2019). Widespread occurrence of pesticides has also been found in long-term organic arable and vegetable fields (Riedo et al., 2021), as well as non-treated grasslands (Riedo et al., 2022).

When studying pesticide residues in European soils, vineyards are of utmost importance. European viticulture is a major agricultural sector, accounting for more than 60 % of global wine production (OIV, 2024). Despite the small cultivation area, due to high revenues viticulture generates over 10 % of production value of all crops in the European Union and even more in some traditional winegrowing countries, such as Italy, France, Portugal, Austria, or Switzerland (Table S1). At the same time, wine grapes are highly sensitive to fungal diseases, especially

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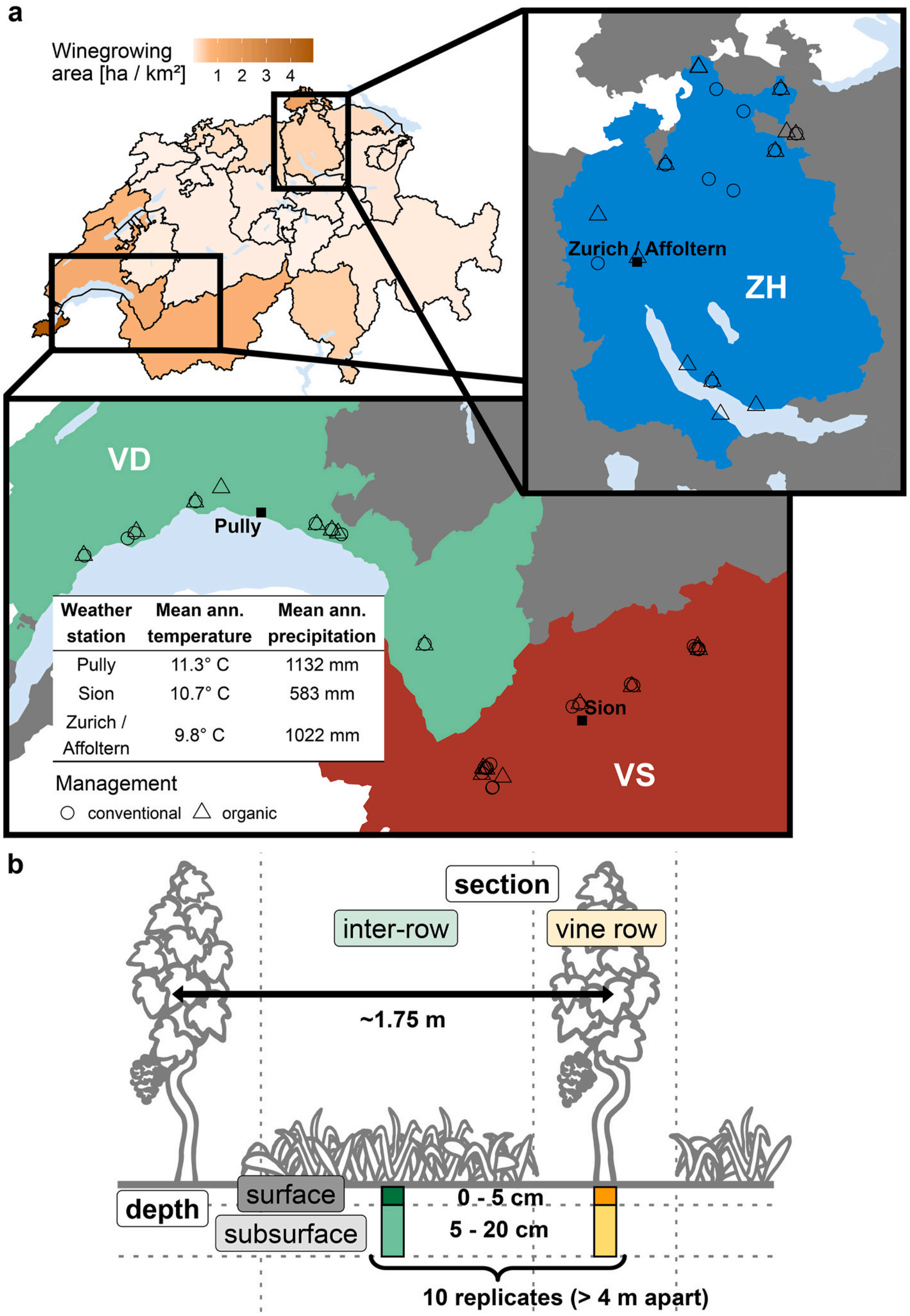
from mildews (Bois et al., 2017). The combination of great crop value and high disease pressure leads to an extensive use of pesticides, mainly fungicides, in wine grapes (Rosenheim et al., 2020). European vineyards often receive more than 20 kg/ha pesticides per year and are responsible for major shares of total pesticide sales, e.g., more than 30 % of pesticides sold in Portugal, France, Italy, and Spain (Table S1). In the case of Switzerland, wine grape accounts for 58 % of overall fungicide use and is the crop with the highest pesticide use per area with over 25 kg/ha per year compared to less than 3 kg/ha in most arable crops (FOAG Federal Office for Agriculture, 2024). As a perennial crop, grape vines are repeatedly treated with pesticides over decades, which further increases pesticide accumulation in soil. In line with this, Knuth et al. (2024) reported that vineyard soils had the highest pesticide concentrations amongst studied crops. While the comparability between crops was limited for several reasons specified in the paper, this finding further supports that pesticide exposure is particularly high in viticultural soils. Vineyards are, therefore, extremely relevant to attain a better understanding of the ecological consequences of pesticide residues in European soils.

Next to a wide range of pesticides, copper is, and has been historically, a central pillar in disease suppression in vineyards, particularly for organic farming where pesticides are forbidden (Dagostin et al., 2011). Copper, a heavy metal, is not degradable and largely immobile in soil, which, over decades of use, has led to its accumulation in many European winegrowing regions (Ballabio et al., 2018). A comprehensive assessment of vineyard soil contamination should therefore cover both



Weather station	Mean ann. temperature	Mean ann. precipitation
Pully	11.3° C	1132 mm
Sion	10.7° C	583 mm
Zurich / Affoltern	9.8° C	1022 mm

Fig. 1. a) Map of Switzerland showing vineyard area density per canton and sampling sites in the three selected regions Vaud (VD), Valais (VS), and Zurich (ZH). Mean annual temperature and precipitation data for a weather station representative for each region shown in the table (data according to Federal Office of Meteorology and Climatology MeteoSwiss (2023)). b) Cross-section of representative vineyard parcel showing separated sampling of section (vine row, inter-row) and depth (surface soil, subsurface soil).

copper and a large number of pesticides which, to our knowledge, has hardly been carried out before in a large-scale study (Vrščaj et al., 2022).

Vineyards are also likely to have a particularly heterogeneous within-field distribution of pesticides and copper compared to non-permanent crops. As applications are usually only to the vine row and the soil is rarely disturbed (Morisod et al., 2018), strong gradients from vine row to inter-row and from surface soil to lower soil layers are expected. While studies on individual vineyards have analyzed spatial distribution of pesticides and copper (Mackie et al., 2013; Silva et al., 2020; Taschenberg et al., 1961), large-scale studies have typically pooled samples across the whole parcel (Froger et al., 2023; Manjarres-López et al., 2021; Pose-Juan et al., 2015), impeding a comparison of within-field differences between different management regimes or regions.

Due to their inherent toxicity, pesticide and copper residues in soil are a potential hazard for soil life (FAO & ITPS, 2017). Numerous studies have found negative effects of pesticides and copper on a wide range of soil organisms including earthworms, arthropods, arbuscular mycorrhizal fungi, and other soil microbes (Bünemann et al., 2006; Edlinger et al., 2022; Hage-Ahmed et al., 2019; Pelosi et al., 2014). Despite the known potential harm for soil organisms, no legal thresholds exist for pesticide residues in European soils thus far (Marti-Roura et al., 2023a; Silva et al., 2023). For copper, thresholds are defined on national level (Ballabio et al., 2018), however, in the case of Switzerland they are based on risk towards humans, livestock, and crops only (OIS, 2016). In the EU context, a guideline value for copper of 150 mg/kg is often used to denote an ecological risk, which is based on Finnish legislations (MEF, 2007). Recent studies have adopted and developed indicators for risk of pesticides towards soil life (Froger et al., 2023; Marti-Roura et al., 2023b; Pelosi et al., 2021; Silva et al., 2023; Vašíčková et al., 2019), however a widely accepted risk assessment approach is still missing and the ecological implications of these risk indicators are largely unvalidated. To facilitate the evaluation of the European Commission's objective to reduce pesticide use and risk by 50 % by 2030 (European Commission, 2020), it is necessary to develop a better understanding of how pesticides affect soil organisms in the field.

To address these knowledge gaps, we studied soil pesticide and copper contamination in 62 vineyards in three major winegrowing regions in Switzerland, including conventional and organic management (Fig. 1a). Switzerland is centrally located between the important wine producing countries France, Italy, Germany, and Austria. It offers wine growing regions in diverse climatic and pedogeological regions within close proximity and with uniform regulations. Swiss vineyards are, therefore, suitable as a case study for central European viticulture. A highly sensitive multi-residue method was used to quantify pesticides in vineyards, covering 146 active substances and transformation products at concentrations down to 0.05 μg/kg soil (Rösch et al., 2023). Multiple samples per vineyard were collected to account for within-field heterogeneity (Fig. 1b) and the dataset was complemented by pesticide application data for five years prior to sampling. To our knowledge this is the first extensive screening study on vineyards combining detailed data on management, copper concentrations, and a broad spectrum of pesticides, including trace concentrations. This powerful setup allowed us to (i) representatively characterize pesticide and copper contamination in Swiss vineyard soils, (ii) analyze their spatial distribution within vineyards, (iii) assess the applicability of persistence data from literature in the field, and (iv) carry out a joint potential risk assessment for soil organisms.

2. Materials and methods

2.1. Study design and sampling

The studied vineyards were part of a farming network and distributed across three major wine-growing regions in Switzerland. Those regions cover different climatic conditions from the canton of Zurich with a relatively cool and humid climate, Vaud with a warmer and humid climate, and Valais with a clearly dryer climate (Fig. 1a). Of the total of 62 parcels sampled, 33 were managed conventionally and 29 organically according to the guidelines of the Federation of Swiss Organic Farmers (Bio Suisse, 2023) which forbids the use of pesticides. Parcels that were not certified but had been managed organically for at least five years were considered organic. Of the 33 conventional parcels, 32 were managed according to the "Proof of Ecological Performance" as recommended by the Swiss Federal Office for Agriculture (FOAG Federal Office for Agriculture, 2023) and 29 had the additional label "Integrated Production Switzerland" which aims to further reduce the use of pesticides and other inputs (IP Suisse, 2023). Soil vegetation cover included fully covered, only inter-row covered, and completely bare soils. Management data were collected directly from the farmers using a questionnaire and included general parcel information as well as pesticide application, fertilizer use, and mechanical treatments for the five growing seasons prior to sampling. There were four conventional parcels that stopped using pesticides before the year of sampling or used a reduced set of pesticides not covered by our method (fungicide folpet and herbicides glyphosate and glufosinate). These years since stopping the application of synthetic pesticides included in our method was combined with the years since conversion to organic farming in a variable that was called "years organic" for simplicity reasons.

Soil sampling was carried out from November to December 2020 to achieve a time interval of 3–4 months since the last pesticide treatment (as an exception three organic parcels in Zurich were sampled in March 2021). This allowed assessing long-term pesticide contamination and improved comparability between vineyards, as an extreme dominance of recently applied pesticides was avoided. Vineyards consist of two sections, the vine row and the inter-row space, which differ in pesticide exposure as applications are usually only to the vine row. Samples were therefore collected from both sections and analyzed separately (Fig. 1b). Ten soil cores per section were taken with a 3 cm diameter auger in five different vine row/inter-row spaces, with a minimum distance of 4 m between soil cores and at least 10 m from the vineyard parcel boundary. The soil cores were split into 0–5 cm and 5–20 cm layers, referred to as surface and subsurface (top)soil, respectively (Fig. 1b), and mixed with the other cores from the same section and depth. This resulted in four pooled soil samples per parcel. The samples were dried to constant weight at 40 °C, sieved to 2 mm, and stored at room temperature in the dark until analysis of pesticide residues and soil physical and chemical properties.

Five parcels were excluded from the initial 67 sampling sites because the distribution of soil properties within the parcel suggested potential sample mix-ups that could not be reconstructed. Exclusion criteria was that depth distribution of soil organic carbon (SOC) and/or pesticides strongly differed between vine row and inter-row (SOC: surface $\ll$ subsurface in one section and surface $>$ subsurface in the other; pesticides: ratio surface/subsurface one section >5*ratio surface/subsurface other section).

2.2. Synthetic pesticide measurements

Pesticide residues were extracted from 5 g of dried soil by an adapted quick, easy, cheap, effective, rugged, and safe (QuEChERS) approach using acidified acetonitrile (2.5 % formic acid) as extraction agent (Lehotay, 2007). Isotopically labelled internal standards were added prior to extraction. Chemical analysis was performed by liquid chromatography coupled to triple quadrupole mass spectrometry with electrospray ionization (LC-ESI-MS/MS). Quantification was based on matrix-matched internal standard calibration. Therefore, calibration standards ranging from 0.05 to 50 ng/g were prepared using a matrix from a standard soil that is largely representative of Swiss agricultural soils. Details on pesticide selection, soil extraction, chemical analysis, instruments used, and method validation are given in Rösch et al. (2023). The calculation of method limit of quantification (MLOQ) and quantification of concentrations above the linear range are explained in the supplementary information (SI) section 1.

2.3. Physiochemical soil analyses and copper

The following soil properties were measured according to the reference methods of the Swiss Federal Agricultural Research Station (https://www.agroscope.ch/referenzmethoden). The texture was split into the fractions clay <2 μm, silt 2–50 μm, and sand >50 μm and determined by pipetting. pH was measured in deionized water. Soil organic carbon was measured by oxidation with dichromate. Total C and N were measured by elemental analysis. The macro-nutrients P, K, Mg, and Ca as well as S, Na, Zn, Mn, Ni, and Pb were extracted in a 0.5 M ammonium acetate−EDTA solution at pH 4.65. Two separate analyses were performed for copper. Extraction in 2 M HNO_3 was used to determine (quasi-)total copper, while the CAT-method (calcium chloride/DTPA extraction) was used to estimate bioavailable copper (CEN European Committee for Standardization, 2001). Both were quantified using inductively coupled plasma optical emission spectroscopy (ICP-OES). Extractions with HNO_3 have been found to yield similar copper concentrations as the common aqua regia extraction or microwave-assisted total digestion (Sabienë et al., 2004; Sastre et al., 2002).

2.4. Data transformation

The number of pesticides was counted using the method limit of detection, while sum concentrations were calculated with the maximum MLOQ per pesticide. To obtain concentrations at parcel level, subsurface soil concentrations were weighed threefold, according to their higher thickness, and sections were weighed equally, assuming constant bulk density over the first 20 cm. Pesticide half-lives (DT50) under laboratory and field conditions were retrieved from the “Pesticides Properties DataBase” (Lewis et al., 2016) (https://sitem.herts.ac.uk/aeru/ppdb/en/index.htm). The website’s recommendations were also used to group pesticides in persistence classes according to their DT50 (<30 days: non-persistent, 30–100 days: moderately persistent, 100–365 days: persistent, >365 days: very persistent).

Risk quotients (RQ) were calculated by dividing pesticide and copper concentrations averaged on parcel level by predicted no effect concentrations (PNEC). For pesticides, we followed the suggestions by the Swiss Centre for Applied Ecotoxicology (Marti-Roura et al., 2023b) referring to the technical guidance document of the European commission (De Bruijn et al., 2002). To derive a PNEC for copper, we used the threshold calculator for metals in soil (version 3) by ARCHE Consulting (https://arche-consulting.be/tools/threshold-calculator-for-metals-in-soil) that takes into account the effect of soil properties on copper toxicity, which lead to a PNEC of 71.2 mg/kg. Details and additional risk assessment approaches are presented in SI section 2.

2.5. Statistical analyses

R version 4.4.1 was used for all statistical analyses (R Core Team, 2023). For analyses at parcel level, analysis of variance (ANOVA) was used to calculate the significance and explained variance of the explanatory variables. The analyses at sample level were performed using a linear mixed model approach due to the nested design. We used the lmer-function (package “lme4” version 1.1–35.5 (Bates et al., 2015)) and included parcel, parcel:depth and parcel:section as random effects. To ease data interpretation by the reader, the different hierarchical levels were displayed separately. We analyzed “simple models” that included only the four design variables region, management, depth, and section. In “extended models” we added further parcel information and soil property data after the design variables and their interaction terms to explain additional variance. As an exception, years organic was added in the first position to correct its regional differences. Soil compositional data were transformed by a centered log-ratio transformation using the pcaCoDa-function (package “robCompositions” version 2.4.1 (Templ et al., 2011)). The first three principal components that explained >75 % of the variation were included in the “extended mixed models” at sample level (Fig. S1). For the total and bioavailable copper data on sample level, no transformation was found that met the assumptions on all hierarchical levels. Therefore, the levels were analyzed separately using ANOVA. On parcel level the data was used non-transformed, whereas depth and section differences were assessed using the concentration ratios between surface and subsurface or vine row and inter-row, respectively, and applying the transformation 1/x - 1. All plots were produced using the “ggplot2” package version 3.5.1 (Wickham, 2016) and the map with the package “ggswissmaps” version 0.1.1 (Petrillo & Stephani, 2016).

3. Results and discussion

We investigated 62 vineyards in three distinct winegrowing regions for soil pesticide residues and found widespread occurrence across all parcels. There were strong management and regional differences (Fig. 2) as well as within parcel differences (Fig. 4) in the number and concentration of pesticides. No management or regional differences were found for copper.

3.1. Unveiling unknown levels of soil pesticide contamination

The high sensitivity of our method allowed us to attain a detailed picture of pesticide contamination in studied vineyards, including transformation products (TP). We found between 4 and 59 pesticides per soil sample (Fig. S2a) and between 11 and 60 pesticides per vineyard with a median of 27.5 (Fig. 2a). After more than 20 years of organic farming, still up to 32 pesticides could be found. Other studies on European soils using multi-residue methods with >100 pesticides typically reported lower number of pesticides with medians below 10 and frequent occurrence of soils without pesticide residues (Froger et al., 2023; Geissen et al., 2021; Silva et al., 2023). The higher numbers in our study are most likely a consequence of lower detection limits and differences in pesticide selection and not primarily of crop management.

Of the 146 pesticides measured, 89 were detected in at least one sample (Fig. S3, Table S2). Of these, 75 were active ingredients (45 fungicides, 14 herbicides, and 16 insecticides), 13 were TP (six of fungicides, six of herbicides, and one of an insecticide), and one was piperonyl butoxide, an insecticide additive (Fig. S3). The most frequent pesticides were boscalid and fludioxonil, found in all studied parcels, followed by the TP cyprodinil CGA249287 (98 % of parcels), TP chlorothalonil-4-hydroxy (97 %), and metalaxyl (95 %). The highest individual concentration per sample was reached by boscalid with a concentration of 1560 μg/kg soil. Boscalid also contributed by far the most to the summed-up pesticide contents across all parcels with 27 %, followed by metrafenone (13 %), fludioxonil (6 %), diuron (5 %) and fluxapyroxad (4 %). All the above pesticides are fungicides or TP of fungicides, except the herbicide diuron.

Few studies have carried out multi-residue analyses of pesticides in a large set of vineyards. Manjarres-López et al. (2021) studied 15 vineyards in Spain and, as in our study, found boscalid the most highly concentrated pesticide frequently reaching concentrations >100 μg/kg. Other common pesticides in both studies were metalaxyl, fluopyram, and dimethomorph (Manjarres-López et al., 2021). In Vrščaj et al. (2022), 176 pesticides were measured in 46 vineyards, but only seven pesticides were detected. While chlorpyriphos was the most frequent and at similar concentrations as in our study, most other substances were at much lower concentrations or remained undetected (Vrščaj et al., 2022).

Pesticide concentrations were summed up as a metric for overall pesticide contamination of the studied soils. Sum concentrations per sample reached up to 3860 μg/kg in a conventionally managed soil and

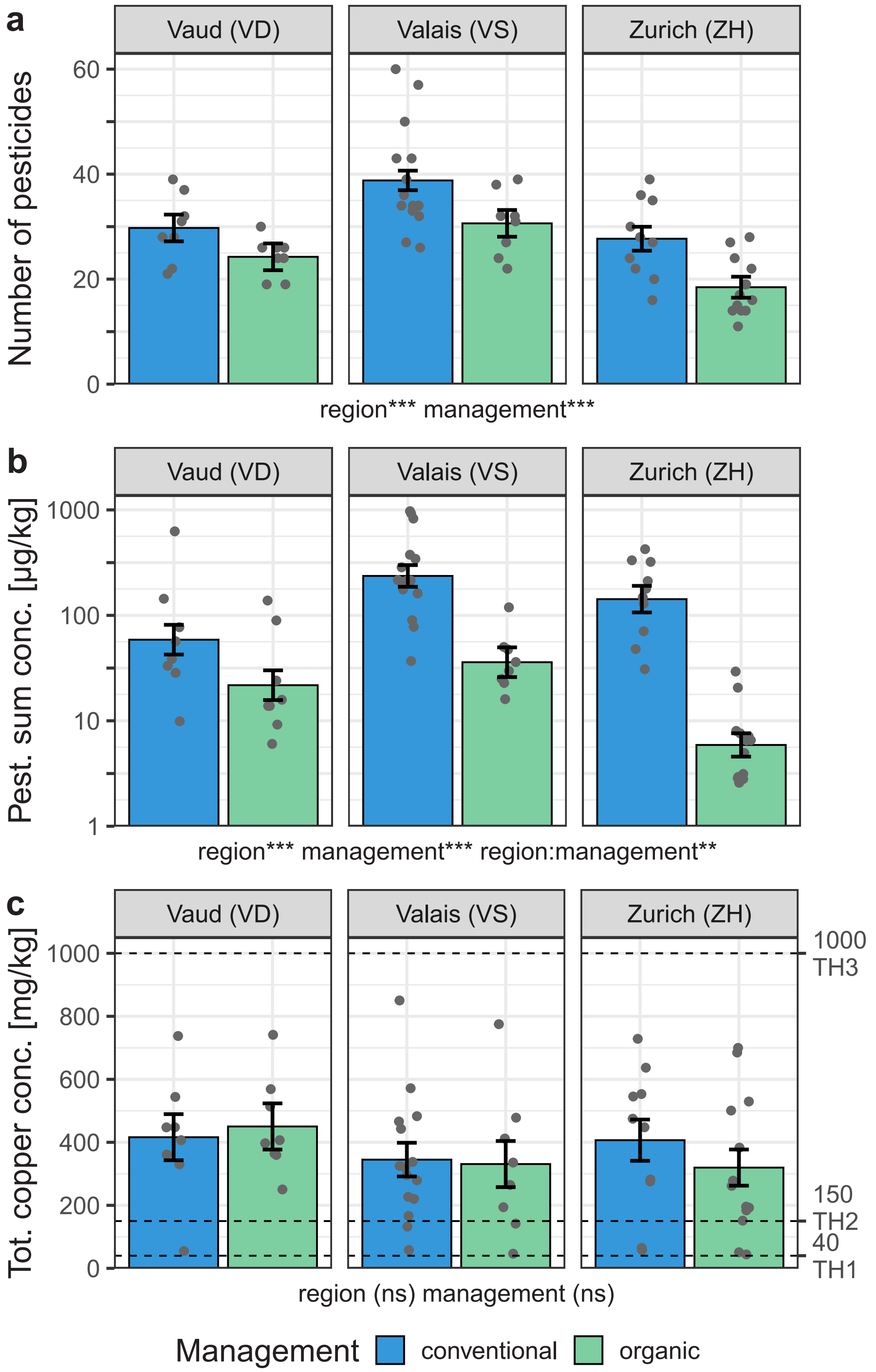


Fig. 2. a) Number of pesticides, b) pesticide sum concentrations, and c) total copper concentrations in vineyard soils per parcel grouped by region and management. Group means, model standard errors, and individual data points are displayed. Significant explanatory variables from 'simple model on parcel level' are displayed below the respective plot using asterisks ($p < 0.001$ (***), $p < 0.01$ (**), and $p < 0.05$ (*)). For copper variables region and management were not significant (ns). Three thresholds (TH) for copper are displayed: TH1 is the guide value by Swiss legislations, TH2 is the guideline value by Finnish legislations indicating an ecological risk, and TH3 is the clean-up value by Swiss legislations.

the lowest concentration was found in an organic vineyard with 0.48 µg/kg soil (Fig. S2b). In organic fields a maximum sum concentration of 271 µg/kg soil was reached.

To compare pesticide residues between vineyards and arable fields, we took the data from Riedo et al. (2021) using the same selection of pesticides and quantification limits. Conventional vineyards had 2.7 times higher median sum concentrations than conventional no-till arable fields (Fig. S4b and c). The samples in Riedo et al. (2021) were collected during the growing season whereas our samples were collected 3–4 months following the last application. As pesticide concentrations typically show pronounced peaks during the growing season (Bucheli et al., 2023), differences could be even greater if samples were collected at the same time of the year. In contrast, the number of pesticides was similar between arable fields and vineyards (Fig. S4a), possibly due to crop rotation in arable farming, which increases the range of pesticides used over the years.

3.2. Copper levels exceeding thresholds

Total copper concentrations in the studied vineyard soils were generally high with a mean concentration of 371 mg/kg and the thresholds based on Swiss and EU legislations were often exceeded (Fig. 2c). Averaged at parcel level, all sites exceeded the Swiss guide value of 40 mg/kg soil (OIS, 2016) and 85 % of the parcels exceeded the EU guideline value for ecological risk of 150 mg/kg soil (MEF, 2007). The highest total copper concentration at parcel level was 850 mg/kg soil, which is just below the clean-up value of 1000 mg/kg in Switzerland (OIS, 2016).

The large-scale study of Ballabio et al. (2018) found much lower concentrations in vineyard soils of the European Union. The overall mean was 49 mg/kg and even in France, the country with the highest copper levels in that study, an average concentration of “only” 91 mg/kg was found. This strong contrast to our study is in line with the study of Droz et al. (2021) that also found especially high copper concentrations in Swiss vineyards and neighboring regions in northern Italy and eastern France. This area is characterized by high precipitation and humidity, and relatively high SOC contents, which were all key variables in the prediction of soil copper contents (Droz et al., 2021) (compare section 3.3). The trends for bioavailable copper were similar to total copper (Fig. S5) with a Pearson correlation coefficient of 0.88 between the two.

3.3. Regional differences from climate and soil properties

The studied vineyards were from the three distinct winegrowing regions Vaud, Valais, and Zurich (Fig. 1a). Region had an especially strong influence on the number of pesticides ($p < 0.001$) explaining 36 % of the total variance (Fig. S6). Highest numbers of pesticides were found in the region Valais with a median of 34 pesticides per parcel, while they were lowest in Zurich with 22 pesticides (Fig. 2a). Also sum concentrations were significantly affected by region ($p < 0.001$) with Valais generally showing highest sum concentrations. Valais showed higher pesticide levels despite not having higher overall application rates (Fig. S7a).

Those differences could be due to climate. Valais has a clearly drier climate compared to the other regions (Fig. 1a), which can reduce microbial activity and, thus, slow down microbial degradation (Schroll et al., 2006). The higher pesticide sum concentrations in Zurich conventional vineyards compared to Vaud could originate from the lower mean annual temperatures (Fig. 1a), which can also hamper pesticide degradation (Wu & Nofziger, 1999).

Climate also influences disease pressure, leading to differences in plant protection strategies. Downy mildew is typically more prevalent in wet climates, whereas powdery mildew prefers warmer and less humid conditions (Bois et al., 2017). In line with this, we found differences in pesticide residues in conventional vineyards depending on the target disease. While fungicides against powdery mildew were most abundant in Valais, fungicides against downy mildew were found at highest sum concentrations in Zurich (Fig. S8b). Similarly, it has been proposed that copper contamination is particularly high in wine-growing regions with a wet climate due to higher disease pressure from downy mildew (Komárek et al., 2010; Neaman et al., 2024). However, we did not find regional differences in soil copper concentrations (Fig. 2c) despite the clear differences in mean annual precipitation (Fig. 1a).

The soils of the investigated regions also differed in SOC content, which is a key driver of pesticide sorption and can prevent pesticide degradation (Arias-Estévez et al., 2008). In Valais, there were particularly carbon-rich surface soils with up to 16 % SOC, which could have undergone strong pesticide accumulation (Fig. S9). In contrast, Vaud was the region with the lowest pesticide sum concentrations and the lowest SOC contents.

3.4. Decline of pesticides after conversion to organic

As expected, management had a strong influence on pesticide exposure. Conventional parcels had 38 % higher median numbers of pesticides than organic parcels and almost 13-times higher median sum concentrations (both $p < 0.001$; Fig. 2a, b). A region-management interaction for sum concentrations ($p = 0.002$) further indicated large differences between organic and conventional management in Zurich and relatively small ones in Vaud (Fig. 2b).

These management differences for pesticides were mainly explained by the time since the last pesticide application. We extended the simple models from above with the number of years since conversion to organic or since terminating synthetic pesticide application (herein after “years organic”). Years organic was strongly negatively related to both number of pesticides and sum concentrations (Fig. 3) with the best model fit for a log(x+1) transformation (Fig. S6). We found that years organic explained most variance in this “extended” model, with 31 % of total variance for number of pesticides and 66 % for sum concentrations (both $p < 0.001$, Fig. S6). Compared to the previous “simple” model, management was no longer significant for both variables. In addition, the effects of region and region-management interaction lost explanatory power, which in the “simple” model was probably partly explained by the differences in mean years organic between regions (Fig. S6).

In contrast, no effect of management (Fig. 2c) or years organic (Fig. S10) was found on total copper concentrations, despite the generally higher dependence of organic viticulture on copper-based fungicides (Dagostin et al., 2011). Although no detailed information about the vineyards’ age was available, we propose that the high copper contents in the soils and the lack of management differences are most

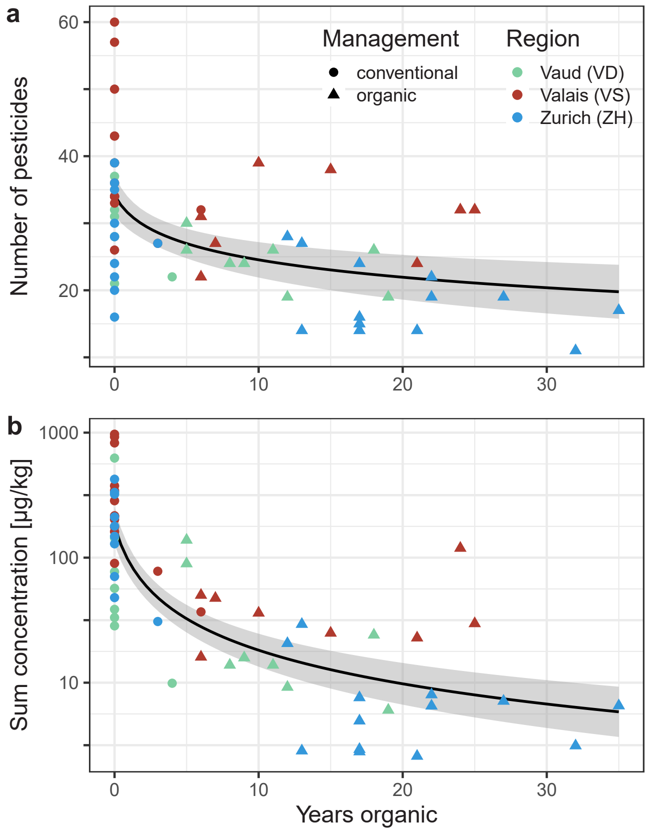


Fig. 3. a) Number of pesticides and b) sum concentrations on parcel level plotted against years organic (including conventional vineyards that terminated application of synthetic pesticides). Regression line between response variable and years organic with log(x+1) transformation added with 95 % confidence interval.

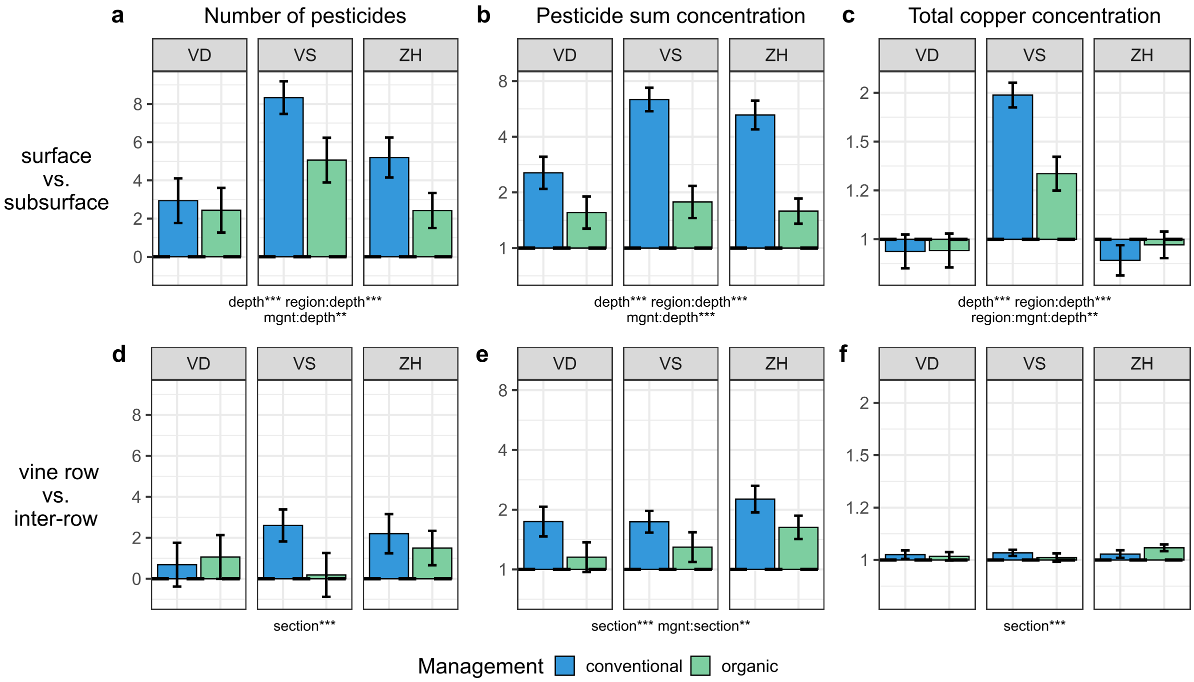


Fig. 4. Within-parcel differences for number of pesticides (a, d), pesticide sum concentrations (b, e), and total copper concentrations (c, f) grouped by region (Vaud (VD), Valais (VS), and Zurich (ZH)) and management (mgnt). Contrasts are expressed as differences for number of pesticides and as ratios for pesticide sum concentrations and copper concentrations, where a ratio of 1 indicates no difference. Group means and model standard errors are displayed. Significant explanatory variables from 'simple mixed model on sample level' are displayed below the respective level using asterisks ($p < 0.001$ (***), $p < 0.01$ (**), and $p < 0.05$ (*)).

likely a result of continuous copper applications over decades. Due to the persistence of copper and its low mobility in most soils, vineyards with a long management history are more likely to have high copper contamination than newer ones (Ballabio et al., 2018; Neaman et al., 2024). Currently, the maximum mean annual copper applications are restricted to 3–4 kg/ha in Swiss viticulture, depending on the management system (Bio Suisse, 2023; Vitiswiss, 2023). Much higher quantities were applied in the past with mean annual copper applications of up to 50 kg/ha between 1920 and 1960 (Speiser et al., 2015). Thus, today's amounts of copper applied over a few years or decades lead to only a small relative increase in already polluted soils (see SI section 3). This can explain why conventional and organic vineyards showed no difference in copper contamination, which has also been observed in Neaman et al. (2024). Additionally, all conventionally managed parcels in our study received frequent copper applications too and the applied copper amounts were not consistently lower than in organic vineyards (Fig. S7b).

3.5. Even non-persistent pesticides can remain in soil for decades

Most pesticide residues did not originate from recent applications. Here we define "residue" as a pesticide occurrence in a specific vineyard. According to the pesticide application records available for 31 conventional vineyards, there were 236 cases of potential residue occurrence from 34 different pesticides that can be detected by our method (most commonly metrafenone, metalaxyl, mandipropamid, and proquinazid). We detected 88 % of these residues, leaving 28 residues undetected (Fig. S11, yellow, and light blue cases, respectively). Cyazofamid and zoxamide both remained undetected in nine cases where they had been applied, partly in the same year of sampling (field DT50 of 4.5 and 6 days, respectively). Additionally, 41 residues of 7 TP from applied pesticides were detected (e.g. cyprodinil CGA249287 and CGA232449, desmethoxy-linuron). In the same 31 parcels, we detected 763 additional residues from pesticides and TP which, according to the application data, had not been applied in the last five years prior to sampling (Fig. S11, orange cases, conventional, e.g. chlorothalonil-4-hydroxy, boscalid, azoxystrobin). In addition, there were 679 residues in the 29 organic fields that had not been treated with pesticides for at least five years prior to sampling (Fig. S11, orange cases, organic, e.g. boscalid, fludioxonil). Compared to recently applied pesticides, residues without application record were generally less concentrated, but elevated concentrations above 10 μg/kg at the parcel level still occurred in 64 cases (Fig. S12). We also commonly found pesticides that had been banned (133 residues from 10 parent pesticides, e.g. atrazine, simazine, orbencarb) or not authorized for viticulture (149 residues from 25 parent pesticides, e.g. pirimicarb, S-metolachlor, imidacloprid) in Switzerland for more than ten years prior to sampling (Table S2).

A long residence time of pesticides in soil can probably explain a large proportion of the numerous residues that were detected but had no application record (Fig. S11). Several studies have already shown that pesticides can persist in soil for much longer than expected from their DT50 alone (Chiaia-Hernandez et al., 2017; Geissen et al., 2021; Riedo et al., 2023). With the high sensitivity of our method, we could now show that a large variety of pesticides with a wide range of DT50 values can remain in the soil over decades at trace concentrations.

Pesticides with low DT50 values are expected to quickly dissipate and, therefore, to be largely absent in soils without recent applications. However, long-term organic vineyards had a similar relative abundance of non-persistent and moderately persistent pesticides as conventional ones (Fig. S13). While persistence seemed to affect how long after the last application pesticides were found at elevated concentrations, trace concentrations (e.g., <1 μg/kg) remained in the soil regardless of their persistence (Fig. S13).

This suggests that DT50 values are not suitable to determine the long-term fate of pesticide traces in soil, which is demonstrated well by the residues of atrazine. Atrazine has a field DT50 of 29 days (=non-persistent) and its use was restricted to maize cultivation since 1987 and finally banned in Switzerland in 2007. Despite this, we detected atrazine in 26 parcels and its TP atrazine-2-hydroxy even in 48 parcels. While this high persistence of atrazine has been described well (Jablonowski et al., 2011), the long-term behavior of many other pesticides has yet to be assessed. Similar findings applied to carbendazim and linuron, although more recently banned. Carbendazim was withdrawn in 2016 with a use-up period until 2018 and linuron was withdrawn from viticulture after 2017 and banned countrywide after 2018. Nevertheless, carbendazim and linuron were detected in 39 and 43 vineyards, including 20 and 18 organic ones, respectively. They were frequently detected, even though carbendazim is considered non-persistent and linuron moderately persistent (field DT50 of 22 and 48 days, respectively).

The persistence of pesticide traces over decades is probably explained by strong sorption to the soil. As pesticides “age” over time, they increasingly sorb to soil particles or diffuse into micropores and become unavailable for biodegradation (Gevao et al., 2000). It is well established that pesticide dissipation often does not follow simple first-order kinetics, instead decreasing dissipation rates over time are frequently observed (Beulke & Brown, 2001). In line with this, we found that pesticide sum concentrations did not show a constant dissipation rate with increasing years organic but a flattening curve (Fig. 3). Similarly, Riedo et al. (2021) only found a difference in sum concentrations between management types but no further decrease with increasing years of organic farming. Better performance is often found by bi-phasic models that take into account the decrease in pesticide dissipation over time (Sarmah & Close, 2009). For a bi-phasic dissipation, DT50 values are not suitable to assess the long-term behavior of a pesticide. Also, DT50 values can vary strongly depending on environmental and soil properties, especially for moderately persistent pesticides (Schäffer et al., 2022). While these findings suggest that many of the detected but not applied residues are indeed remnants from historic applications, pesticides that are still authorized might also originate from other sources, such as spray drift or atmospheric deposition (see SI section 4).

3.6. Within-field differences crucial for sampling strategy

The distribution of pesticides within parcels was highly heterogeneous in both vertical and horizontal dimension, which was analyzed with a linear mixed model on sample level. As pesticide deposition is usually to the soil surface, the surface soil is predominantly more exposed than the subsurface soil. In line with this we found 21 % higher median numbers and 2.5 times higher median sum concentrations in the surface soil than the corresponding subsurface soil layer (both $p < 0.001$; Fig. 4a and b).

A region-depth interaction (both $p < 0.001$) indicated that depth differences were particularly pronounced in Valais and less pronounced in Vaud (4-fold vs. 2.1-fold median differences; Fig. 4a and b). These bigger depth differences in Valais could originate from reduced downward movement of water and dissolved pesticides under dryer climate (Fig. 1a). Strong sorption in the humic surface soil layer in many Valais soils is likely to additionally prevent pesticide movement to the subsurface soil (Arias-Estévez et al., 2008).

There was a management-depth interaction, with conventional fields generally having greater depth differences in numbers and sum concentrations than organic fields (both $p < 0.001$; Fig. 4a and b). While in conventional parcels the median sum concentrations in the surface soil were 4.4 times higher than in the subsurface soil, organic parcels only had a 1.5-fold difference. These management differences in depth distribution were mainly due to years organic, which we analyzed using ratios between sum concentrations in surface and subsurface soil. There was a strong negative linear relationship between log concentration ratios and log(x+1) transformed years organic ($p < 0.001$; Fig. S14). Thus, the sum concentrations in the surface soil and subsurface soil became more similar with increasing years organic, which can partly be explained by the transport of pesticides to lower soil layers over time (Navarro et al., 2007). Moreover, biodegradation of pesticides typically decreases with increasing soil depth (Rodríguez-Cruz et al., 2006). A faster degradation in the surface soil can even cause its concentration to fall below the subsurface soil one as observed in some vineyards (Fig. S14).

For total copper we also found significantly higher concentrations in the surface soil than in the subsurface soil ($p < 0.001$), similarly to pesticides but clearly less pronounced (Fig. 4c). The interaction between region and depth indicated strong depth differences in Valais and smaller differences in Vaud and Zurich ($p < 0.001$, Fig. 4c). These depth differences were especially strong in conventional Valais parcels which could be related to the high age of these vineyards. As copper contamination is largely due to historical applications, large turbation events in the past, such as the renewal of a vineyard, are expected to have a strong effect on its distribution within the field. Vineyards with older vines clearly had higher depth differences in copper concentrations than newer ones (Fig. S15). The pronounced depth differences for copper in Valais (Fig. 4c) could therefore be explained by the longer period without major soil disturbance compared to the other regions. Altogether this would indicate that copper was largely immobile in the studied soils and therefore mainly distributed by soil turbation, while pesticide distribution was mainly driven by dissipation and downward transport. How individual pesticide properties can influence their depth distribution is further explored in the SI section 5.

Pesticides are usually applied to the vine row, leading to a higher exposure of this section compared to the inter-row. We confirmed this by finding higher pesticide numbers, sum concentrations, and total copper concentrations in the vine row compared to the inter-row (all $p < 0.001$; Fig. 4d–f). A management-section interaction for pesticide sum concentrations additionally indicated that section differences were more pronounced in conventional compared to organic parcels (Fig. 4e). In conventional parcels, the median sum concentrations in the vine row were 77 % higher than in the inter-row, whereas in organic parcels this difference was only 39 %.

We show that within-field heterogeneity is not only very pronounced in vineyards, but also affected by a variety of factors such as management, climate, soil properties, or contaminant properties. This implies that the sampling strategy is crucial to representatively assess pesticide and copper contamination in vineyards and other non-tilled soils and avoid over- or underestimations of certain management types or site-specific conditions.

Finally, the model at the sample level was extended with soil properties, which explained some additional variance, especially for the number of pesticides (Fig. S16). Running equivalent analyses with individual pesticides of high frequency yielded similar results as for sum concentrations (see SI section 6).

3.7. Vineyard soils are particularly at risk from pesticides and copper

Compared to other land uses, vineyard soils are particularly exposed to pesticides (Fig. S4b) and copper (Ballabio et al., 2018), which can be translated into a higher potential risk. To compare and cumulate the potential risk from different contaminants to soil organisms, we used risk quotients (RQ). A RQ of one for an individual pesticide would indicate a concentration equal to the potential no effect concentration (PNEC) and an RQ larger than one would indicate a potential risk.

The cumulative RQ for all pesticides attained up to 13.9 per parcel. Over 70 % of conventional parcels, as well as 28 % of organic parcels, had cumulated RQs greater than one (Fig. 5). This is a clear difference to the study of Froger et al. (2023) where a high risk with $RQ > 1$ occurred in only a few cases that were limited to conventional arable fields. The reported RQ can be considered a baseline risk, as sampling took place 3–4 months after the last pesticide application and much higher risks are expected during the growing season (Honert et al., 2025). Chlorpyrifos and boscalid were particularly prominent, together contributing to more than 50 % of the overall risk across all parcels and frequently reaching concentrations larger than their PNEC (Fig. 5, Table S3). Due to its high toxicity, the insecticide chlorpyrifos had the highest mean RQ, despite only contributing 0.8 % to the total concentrations across parcels. Similarly, the insecticide imidacloprid ranked third among all tested

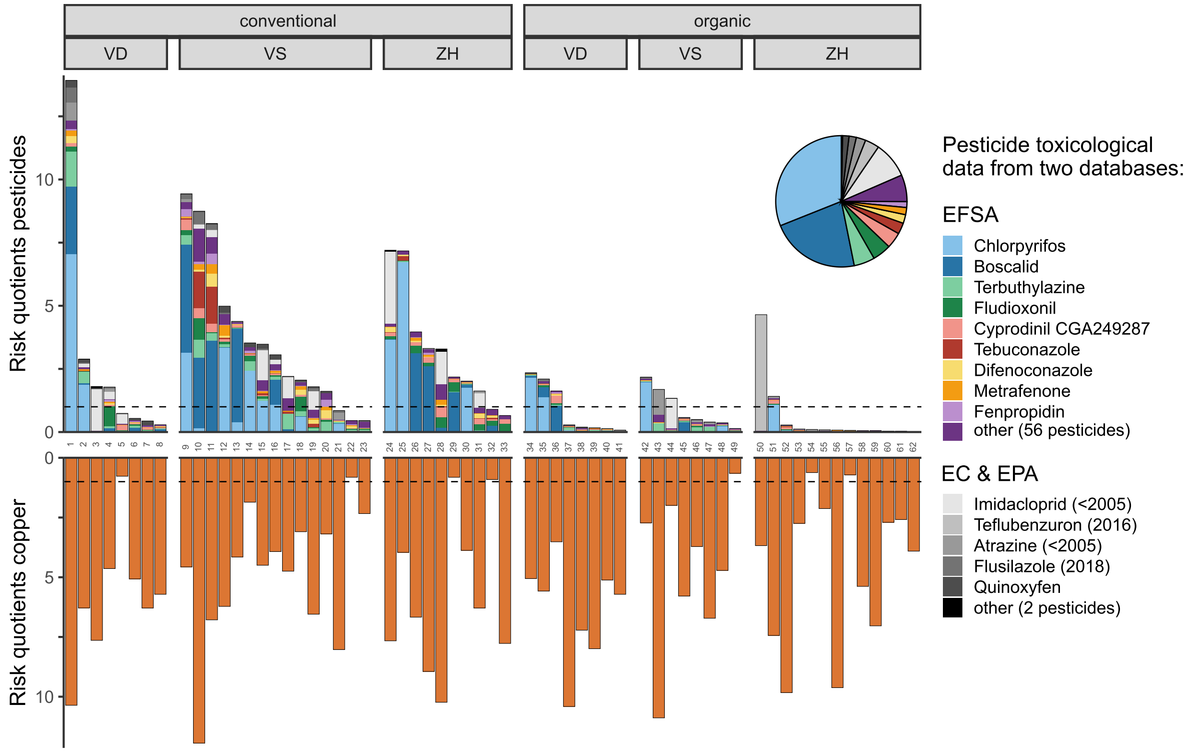


Fig. 5. Cumulated risk quotients (RQ) of pesticides (top) and RQ of copper (bottom) in vineyard soil per parcel grouped by management and region (Vaud (VD), Valais (VS), and Zurich (ZH)). Dashed line indicates an RQ of 1 above which a potential risk for soil organisms is expected. RQ were calculated with individual assessment factors per pesticide. Contribution of individual pesticides to cumulated RQ is displayed with colors and the pie chart displays contribution of pesticides to cumulated RQ across all parcels. Pesticides are separated by database used for toxicological data and ordered by cumulated risk across all parcels. Years in brackets indicate last year of authorization in viticulture if before sampling (2020). Databases: European Food Safety Authority (EFSA) reports, European Commission (EC) reports, and United States Environmental Protection Agency (EPA) ECOTOX Knowledgebase. For copper a PNEC of 71.2 mg/kg was used. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

pesticides, despite being generally present at low concentrations. Hence, insecticides were disproportionally dominant in the risk assessment compared to fungicides and herbicides. While insecticides accounted for only 1.4 % of the total amount of pesticides with toxicological data, they contributed 44 % of the total risk cumulated over all parcels (Fig. S17). Similar observations were made in previous studies (Froger et al., 2023; Pelosi et al., 2021). The two widespread and highly concentrated fungicides boscalid and fludioxonil ranked second and fifth, respectively (Fig. 5, Table S3). The ranking of two additional risk assessment approaches is shown in Table S3 and further discussed in SI section 7.

For copper, we used a PNEC of 71.2 mg/kg. This PNEC was exceeded by 55 out of 62 vineyards and RQ of up to 11.9 were reached (Fig. 5). In most parcels, RQ for copper were higher than cumulated RQ for pesticides, especially in organic vineyards. In accordance with their concentrations, the risk from pesticides was clearly higher in conventional vineyards than in organic ones, while the risk from copper was equivalent. Due to the strong accumulation over decades and high persistence, copper is and probably remains the most important contaminant in most European vineyard soils.

There are currently no recommendations for the joint risk assessment of pesticide mixtures (“cocktail effects”). We therefore assumed additive effects of pesticides and analyzed the risk of copper separately. However, the combination of often several dozen pesticides and high heavy metal concentrations could promote synergistic negative effects (Wang et al., 2015). Many problematic pesticides according to our risk assessments have recently been banned in Switzerland, including chlorpyriphos, imidacloprid, teflubenzuron, diuron, flusilazole, and quinoxyfen (Table S2). The risk posed by those pesticides to agricultural soils is therefore expected to decrease continuously in the future. This indicates also that we identified similar high-risk pesticides as authorities, despite the limitations of our risk assessment approach (see SI section 7).

4. Conclusions and recommendations

The joint analysis of pesticides and copper in vineyards is important in view of recent studies indicating that multiple stresses have an additive negative effect on soil functioning (Beaumelle et al., 2021; Rillig et al., 2023). According to our risk assessment, 50 % of studied vineyards were simultaneously at risk from both pesticides and copper and only 10 % of vineyards were not at risk from either of the two (Fig. 5). While for copper only a further increase can be prevented, pesticides have been shown to decrease over time after application has stopped. Identifying and replacing high-risk pesticides with less problematic alternatives necessitates a collaborative effort between researchers and policymakers and is a first step in reducing pesticide risks that can directly be implemented by wine growers. However, more fundamental changes are probably required to achieve the goal of reducing pesticide risks by 50 % by 2030, as set out in the Farm to Fork Strategy (European Commission, 2020). Our study indicates that conversion to organic farming can achieve a marked decline in pesticide exposure within few years without aggravating risks from copper in the short to medium term. Additionally, a variety of management practices exist already that allow the reduction of pesticide and copper input, such as the use of fungus-resistant wine grape varieties (see SI section 8). Policymakers should set incentives to support the transition to eco-friendly practices while research is commissioned to continuously improve their practicability and efficiency. This knowledge is crucial to maintain crop production while progressing towards healthier soils.

CRediT authorship contribution statement

Elias Barmettler: Writing – original draft, Visualization, Investigation, Formal analysis, Data curation. **Marcel G.A. van der Heijden:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization. **Andrea Rösch:** Methodology. **Lina Egli-Künzler:** Investigation, Data curation. **Pierre-Henri Dubuis:** Investigation, Data curation, Conceptualization. **Kathleen A. Mackie-Haas:** Writing – review & editing, Supervision, Conceptualization. **Stefanie Lutz:** Writing – review & editing, Supervision, Project administration. **Thomas D. Bucheli:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization.

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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.

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Appendix A. Supplementary data

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Data availability

Data will be made available on request.

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