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Spatial Distribution of Agricultural Pesticide Use and Predicted Exposure of the Swiss Landscape.

ESS 511 Master's Thesis

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Abbreviations

AI	Active Ingredient
CTF	Crop Type Fraction
DHM	Digital Height Model
GIS	Geoinformation Systems
hl	Half-life
K_{OC}	Soil organic carbon-water partition coefficient (K_{OC})
POI	Pesticide Occurrence Index
PPI	Pesticide Property Index
PU	Pesticide Use
PUD	Pesticide Use Density
rPPI	Relative Pesticide Property Index
rPOI	Relative Pesticide Occurrence Index
rPUD	Relative Pesticide Use Density
SS	Sum of Squares
TPM	Total applied Pesticide Mass
ws	Water Solubility at 20° degrees

Abstract

Small family farms, which cover roughly 36% of the Swiss landscape, employ mostly intensive agriculture, here defined as agricultural practices which generate high yield per unit area. In these intensive agricultural regions, pest control using pesticides is common, however, pesticides cause major negative effects on various ecosystems. Studies have discovered pesticides in groundwater, surface water and soils all over Switzerland. A continuous spatial approximation of the potential pesticide pollution problem for Switzerland has not yet been established. The goal of this thesis is to calculate and map the distribution of pesticide use and identify areas with higher likelihood of pesticide occurrence. To this regard, I apply pesticide use, precipitation, and physiochemical properties mapped at $1km^2$ spatial resolution to calculate the Pesticide Occurrence Index (POI). In total 1236 tons of fungicide, 498 tons of herbicide and 36 tons of insecticide were applied for a one-year period, consisting of 166 different active ingredients. The POI insecticide and fungicide map showed similar patterns, mainly driven by grapevine production, which was expected due to high fungicide use. The valley of Sion in Valais and the northern region of Geneva showed high POI fungicide and insecticide values. Herbicide pesticide occurrence hot-spots appeared in Valais, Vaud, the Jura Region, and eastern Pre-Alp areas. Herbicide occurrence was highest among pesticide groups driven by the herbicides applied onto fields. Due to the sensitivity of grapevines and their need for fungal control, their impact on the POI of fungicide was in line with expectations. The large effect of herbicide application on fields was, however, unexpected, because only 5.9 tons of herbicides were applied onto fields. However, the high POI herbicide values were in line with Swiss environmental pesticide pollution findings. Overall, these results suggest that the calculated POI in combination with monitoring data, retrieved from monitoring programs such as NAQUA, provide fundamental information that can aid developing future ecosystem monitoring programs, assist in defining monitoring sites, and consequently support pesticide policy development by government agencies.

Keywords: Pesticides, Agriculture, Spatial Accounting, Pollution

1. Introduction

Agriculture and pasture are an integral part of the Swiss landscape. Agriculture is largely dominated by small family farms and covers roughly 36% of Swiss territory, and has become increasingly intensive (Agristat 2016; BAFU 2017). Agricultural intensification arose with the second and third agricultural revolution (mid-19th to 20th century and latter half of the 20th century respectively), during which motorization and chemicalization became a driving force for the ever-increasing yields of Swiss agriculture (Moser 2019). Large-scale use of chemicals for crop fertilization and pesticide use for agricultural yield security continues to this day, resulting in lower costs and more stable agricultural revenue (Moser 2019). However, this intensification has led to an accumulation of pesticides in Swiss ecosystems. A study by Riedo et al. (2021) screening 100 fields under organic and conventional management found 46 pesticides (16 herbicides, 8 herbicide transformation products, 17 fungicides and 7 insecticides). In all fields including 40 organic fields pesticides were discovered. Up to 16 different pesticide residuals were found even after 20 years of organic agriculture. However, not only soils are impacted by pesticide pollution. A study by Wittmer et al. (2014) investigating five medium sized surface rivers found a total of 104 different pesticides. Each sample contained around 40 different pesticides, often transgressing the legal quality limit of 0.1 µg/l, most of them herbicides (Wittmer et al. 2014). Similarly, a study by Gerecke et al. (2002) investigating the effluents of wastewater treatment plants in two rivers during a four month period continuously found concentrations of various pesticides (e.g. atrazine, diuron, mecoprop) within their retrieved samples as well. However, all monitoring efforts so far, have been performed on isolated sampling sites in rivers and fields. Therefore, there is still a gap in the knowledge on the larger continuous spatial extent of the pesticide pollution problem.

Pesticides are defined as substances or mixtures of substances intended for preventing, destroying, repelling, or lessening the damage of any pest (Eldridge 2008). Often, pesticides are named after the target species, i.e. the organism that the pesticide works against, and any other living organism is referred to as a non-target organism (Adisesh 2013). Pesticides are grouped by their target pests and include herbicide against weeds, insecticide against insects, fungicides against fungi. The active ingredients (AI) within the pesticide products are the chemical components that act to control the pests. There are different types of active ingredients, depending on their effectiveness and selectivity: (i) conventional, which are all ingredients other than biological and antimicrobial pesticides, (ii) antimicrobial, which are substances used to destroy or suppress the growth of harmful microorganism for example bacteria, viruses, or fungi, and (iii) biopesticides, which are types of ingredients derived from certain natural materials. All pesticide products contain at least one AI and one "inert ingredient", i.e.

other intentionally added ingredients acting as solvents, surfactants, or preservatives, among many other functions (U.S. EPA 2021).

Inerts can be chemicals, compounds or substances which consist of common food types (edible oils, spices, herbs) and some natural materials, e.g. beeswax, cellulose (Cox and Surgan 2006; U.S. EPA 2021). These substances reach high values in the environment as application doses vary spatially and temporally. The most common are kerosene, propane and other petroleum products, wintergreen oil, peanuts, beeswax, and salt, but it is unclear the extent to which each is applied (National Pesticide Information Center 2021). Pesticides drift and contaminate the air, the soil, and water, and can be toxic to other non-target organisms, including humans (Felsot et al. 2011; Yadav and Davi 2017). Global pesticide application is currently (2018) estimated at 4.1 million tons each year (FAO 2021). Pesticides can enter ecosystems by a point source input or a diffuse source input. Point source inputs correspond to a locations with high concentration of these chemical pollutants and occur at a specific or a set of restricted entry points (Carter 2000). Examples of pesticide point sources are sewage plants, sewer overflows, and losses from farms (Holvoet, Seuntjens, and Vanrolleghem 2007). Diffuse input pesticides correspond to entry pathways into surface waters, such as surface and subsurface runoff, drain flow, drift, atmospheric deposition, soil erosion from treated fields, spray drift application, and deposition after volatilization (Carter 2000; Holvoet et al. 2007). Both of these entry pathways may affect the environment directly or indirectly, however, the research has shown that diffuse pesticide pollution input sources from agricultural areas are one of the greatest causes of contaminated surface waters (Loague, Corwin, and Ellsworth 1998; Müller et al. 2002; Schulz 2004).

In total, there are four different ways humans can be exposed to toxic pesticides: ingestion (consuming polluted food or water), dermal (exposure through the skin), ocular (exposure through the eyes) and inhalation (exposure to polluted air) (Damalas and Koutroubas 2016). Polluted groundwaters can harm humans as well as entire ecosystems when exposed to toxic pesticide concentrations (Margni et al. 2002). Pesticides can cause diseases in humans such as cancer (Lee et al. 2005), respiratory disease (Chakraborty et al. 2009), Alzheimer's disease, Parkinson's disease (Elbaz et al. 2009), reproductive disorders (Petrelli and Mantovani 2002), and several other ailments (Yadav and Davi, 2017).

In Swiss ecosystems pesticide pollution has displayed a multitude of harmful effects (SCNAT 2021). Negative consequences of insecticide and fungicide application on mycorrhizal fungi, which are beneficial for crop growth, have been established by Bünemann et al. (2006) and Riedo et al. (2021). Additionally, a study conducted in Vienna on earthworms and bacterial communities in root systems found that they were negatively impacted by seed treatment (Van Hoesel et al. 2017). Furthermore, soil organisms have shown to recover differently from pesticide application, causing ecosystem imbalances and diversity changes in soils (Kattwinkel et al. 2015). Besides soil organisms, water organisms such as insect larvae, algae, fungi, fish were negatively impaired by pesticide pollution within water systems (Burdon et al. 2019; Chiaia-Hernández et al. 2020; Doppler et al. 2020; Junghans et al. 2019; Schneeweiss et al. 2019; Spycher et al. 2015, 2018, 2019). Due to increased herbicide use, diversity loss of arable flora has been found all over Europe (Andreasen and Streibig 2011; Freemark and Boutin 1995; Richner et al. 2015). Additionally, the use of pesticides was found to be one of multiple factors which led to a significant loss of diversity and frequency of insects (Geiger et al. 2010; Gilburn et al. 2015; Sánchez-Bayo and Wyckhuys 2019; Wagner et al. 2021). This in turn influenced insect-eating bird and mammal populations. In Switzerland, the population of insect-eating bird species in cultivated regions reduced by 60% since 1990 (Knaus et al. 2018). Lastly, small mammals such as bats have been shown to be especially sensitive to pesticide application (Carravieri and Scheifler 2012).

To accurately predict the magnitude of pesticide use, pesticide occurrence, and the associated risks for hydrological and terrestrial ecosystems, it is important to know the extent of (i) the treated areas, which are highly driven by the topography, slope, soil quality and precipitation (Carew, Smith, and Grant 2009; Jiang and Thelen 2004), (ii) the application rate, and (iii) the intervention number applied for individual crops (de Baan, Spycher, and Daniel 2020). Soil fertility has been known to negatively correlate with the topography, resulting in agriculture-free mountainous areas and low pesticide application in the regions (Jiang and Thelen 2004). Pesticide groups with higher application mass will more strongly impact pesticide use than others. In 2019, about 55% of the pesticides sold in Switzerland were fungicides, while only 27% were herbicides, and the rest insecticides (BLW 2020). This is highly indicative of the application mass, application rates, and intervention numbers of the different pesticide groups in Switzerland. In addition to pesticide use, several other factors at the field level can influence the fate of pesticides, such as physiochemical properties that encompass mobility in water and persistence in the soil, as well as factors such as topography, precipitation, soil properties, and hydrological connectivity (Holvoet et al. 2007;

Payraudeau and Gregoire 2012). Important physiochemical properties include (i) half-life, which is defined as the time necessary for 50% of the initial concentration of the pesticide to degrade, and indicates how long a pesticide might impact an ecosystem (Pino and Dîaz 1998), (ii) the water/soil partition coefficient (K_{OC}), which refers to the ratio of pesticide absorbed to pesticide remaining in the soil, and is often used to predict pesticide soil mobility (Weber, Wilkerson, and Reinhardt 2004), and (iii) water solubility, which explains how easily a pesticide is dissolved in water, since water is the vehicle by which pesticides move into ground water (Pino and Dîaz 1998).

Modern insecticide are known to reside in the soil after applications rather than leaching into the ground water, therefore they often display low-to-moderate water solubility levels, but moderate-to-high soil persistence levels (Malaj, Liber, and Morrissey 2020). Fungicides and herbicides show higher diversity of physiochemical property levels within the pesticide groups, depending on their active ingredient (AI). Some are mobile and travel into ground water systems while others persist in the soil. Additionally, crop types have an impact on the pesticide use and occurrence as well, due to their different sensitivities towards pests and the number of pests they are subjected to throughout their growing seasons, resulting in varying amounts of pesticides needed to control the respective pests, application rates, and intervention numbers (de Baan et al. 2020). For example, several pests and diseases have grapevine as their favorite host and the vineyard as preferred environment, leading to increased pesticide application rates and intervention numbers (Pertot et al. 2017). Keeping fungal infestation at bay on certain crop, such as stone fruits, grapevines, and potatoes can drive fungicide application mass into quantities unachieved by herbicide or insecticide use (BLW 2020).

There are several options to monitor pesticides in the environment, which include monitoring stations, maps, and models. Monitoring programs such as National Groundwater Monitoring (NAQUA) deliver information on ground water quality and pesticides therein (FOEN 2019).

Chen et al. (2002) proposed a benchmark pesticide mobility index relevant to surface water runoff and soil erosion for multiple pesticides, i.e. a way to estimate pesticide exposure. Pesticide exposure estimates require information on chemical toxicity concentrations over time, which are often lacking over large spatial areas. Therefore, the authors proposed to use two key pesticide properties: Half-life and water/soil partition coefficient to calculate the surface water mobility index and model pesticide movement. The index allows us to investigate multiple chemicals in one index (Chen et al. 2002).

Process-based models have also become very popular to model pesticides in the environment, as they enable us to capture dominant physical, chemical, and biological processes that explain the movement of chemicals in soils over space and time. For example, the Soil and Water Assessment Tool (SWAT) and the Pesticide Root Zone Model (Akbar, Lin, and DeGroote 2011) have been widely used to predict the environmental impact of land use, land management practices, and climate change, and have shown a great capacity in determining the fate of pesticides in the environment. However, many of these models fail to accurately predict pesticide concentrations in water bodies within a watershed, and may result in predictions orders of magnitude different from the monitoring data (Holvoet et al. 2007). Spatially explicit predictive modeling for pesticide occurrence is rare but necessary to estimate surface waters and environment pesticide exposure (Malaj et al. 2020). For example, Ali Akbar et al. (2011) developed a spatially explicit model called ArcPRZM-3 for analyzing pesticide leaching potential from the soil surface towards groundwater. The model enables simulating the maximum dissolved bentazon concentration at a 0.75m soil depth for a two-year interval, and validation is implemented with bentazon data from monitoring wells from the same area. The results showed that 100% of the wells in which bentazon was detected were within the high-risk category based on the ArcPRZM-3 predictions. Unfortunately, bentazon is only one of many chemicals entering groundwater supplies by pesticide pollution (Akbar et al. 2011) and more models for the different chemicals are urgently needed.

Despite the growing pesticide pollution in Swiss soils, groundwater supplies, and the negative side effects it thereby creates for many ecosystems discussed earlier, little large-scale continuous research exists on the extent and magnitude of the pesticide pollution problem for the Swiss landscape. A spatial analysis for Switzerland of the distribution of pesticide use, and the effect of precipitation and pesticide chemical properties may enable developing a vulnerability map (Akbar et al. 2011), which has yet to be established for Switzerland. To meet this gap in knowledge, in this thesis I establish a (relative) Pesticide Occurrence Index incorporating pesticide application rate and intervention numbers, pesticide chemical properties, and precipitation over Switzerland for the year of 2020. Ultimately, I provide a visualization of areas with higher likelihood of contamination (Malaj et al. 2020). This type of analysis can help prioritize sites for field monitoring programs, used in education and public information, and for pesticide policy development.

1.1. Research Questions

In this thesis I answer the following questions:

1. Which areas of Switzerland experience the highest pesticide occurrence and pesticide use density?
2. Which pesticide group displays the highest pesticide occurrence and pesticide use density?
3. Which crop type has the highest impact on pesticide occurrence and pesticide use density?

1.2. Hypotheses

1. The Pesticide Occurrence Index (POI) values will be strongly negatively correlated with the Swiss topography because higher/lower elevation areas will lead to a decreasing/increasing effect on POI for all pesticide groups. This is due to decreased soil fertility at high elevations and increased slope areas (Jiang and Thelen 2004). Additionally, little agriculture is done in these regions (Jiang and Thelen 2004). Lower quantities of pesticides are therefore applied, potentially resulting in a low POI value at high elevation and higher slope areas.
2. The $rPUD_{Fungicide}$ is higher than the $rPUD_{Insecticide}$ or $rPUD_{Herbicide}$ due to higher application rates and a higher number of fungicide applications relative to herbicides and insecticides. This is due to the impact of different numbers, and the distribution of fungal crop pathogens leading to fungal infections and fungicide use (Fausto, Rodrigues, and Coelho 2019).
3. Regions with extensive vineyards will display higher $POI_{Fungicide}$ values compared to any other crop type in the region. Grapevines are very sensitive to pests and experience one of the highest application rates of all pesticide groups in Switzerland. Vineyards are affected by numerous pests, especially fungal and insecticide infections, such as, *Typholodromus exhilarates* Ragusa (Liguori and Guidi 1995), grape berry moths (Thiery 2011), different kinds of grape powdery mildew (Carisse et al. 2009), and many more.

2. Methodology

2.1. Study Area

This research is conducted for the entire area of Switzerland. The country is dominated by 153 agricultural land cover types (BLW 2020). Swiss elevation varies between 193 m and 4379 m. The southern part of Switzerland is dominated by the mountainous Alps with elevations up to 4379 m, the north experiences lower elevations, however the lowest region of Switzerland can be found in the Lake Maggiore in the South with 193m. The average precipitation varied from 510 mm to 3749 mm from 1981 – 2010.

2.2. Measuring pesticide distribution and use at the country level

To understand the spatial distribution of pesticide occurrence and pesticide use across Switzerland, I chose two indices: Pesticide Use Density (PUD) and Pesticide Property Index (PPI). I chose these indices because the PUD can predict continuous applied pesticide use by means of the crop type area and the applied pesticide mass per crop type. However, the PUD is not able to include local variations in pesticide mass due to changes in climate, natural environment, and pest occurrence. The PPI is able predict the persistence and mobility of the applied pesticides by including three physiochemical properties (water solubility, half-life and K_{OC}). Pesticide fate however is driven by many other factors beyond the three physiochemical properties, such as the application method, application time, and soil composition, among many others (Holvoet et al. 2007; Payraudeau and Gregoire 2012; Reichenberger et al. 2007). For this analysis I chose to calculate the two indices at a 1 km^2 spatial resolution over all of Switzerland. The complete workflow for this analysis can be observed in Figure 1.

2.2.1. Pesticide Use

The first step was to calculate the total quantity of pesticides applied for each pesticide group and crop type. Pesticide Use (PU) was calculated using the cultivated area (BLW 2021b), application rates, and number of interventions (de Baan et al. 2020) (equation 1).

$$PU_{i,j,g} = (Cultivated\ Area_{i,j,g} \times Application\ Rates_{i,j}) \times Number\ of\ Interventions_{i,j} \quad \text{(Equation 1)}$$

Where, i is the crop type, j is the pesticide group, and g each grid cell. Unfortunately, by using only one application rate and intervention number per crop type, the accuracy of this method is highly reduced. The results will therefore not be able to visualize differences of local application mass differences. However, the index is able to visualize and predict the effect of the pesticide mass differences between the individual crop types and pesticide group which is a primary focus of this thesis. Additionally, no distinction was made between organic agricultural areas and non-organic agricultural areas in this calculation.

The application rate and intervention number for grapevine fungicide application was adjusted after comparing the values with a secondary literature source from the canton of Valais (Glasenapp and Bosshard 2013), which displayed a significantly lower intervention number compared to the calculated value retrieved from Agroscope used in this analysis (de Baan, Spycher, and Daniel 2015). Using a weighted mean on the secondary Valais data set resulted in a new intervention number of 1.25 which was significantly lower than the retrieved value of 9.4 by Agroscope. For further calculations, I used a mean of $\frac{1.25+9.4}{2}$ leading to a new intervention number of 5.3. A similar approach was used for the adjustment of the application rate of 23.5 kg/ha. The total applied fungicide mass of grapevines stated in the Swiss agricultural report (BLW 2020) was divided by the total sum of cultivated area of grapevines and the new intervention number, resulting in a new application rate of 17.3 kg/ha.

2.2.2. Total applied Pesticide Mass

The second step was to calculate the Total applied Pesticide Mass (TPM; Equation 2), which summed PU for all pesticide groups (fungicide, herbicide, and insecticide) together. TPM is an indicator of total pollution severity of pesticides in Switzerland.

$$TPM = \sum_{j=1}^{p} \sum_{i=1}^{k} \sum_{g=1}^{n} PU_{j,i,g} \qquad \text{(Equation 2)}$$

Where, g is a grid cell, i is the crop type, j is the pesticide group, k is the number of crop type, n is the number of grid cells (n is 88800 for all of Switzerland), and p is herbicide, fungicide, or insecticide.

2.2.2.Pesticide Use Density

The Pesticide Use Density (PUD) was calculated using PU. However, a crop type fraction was incorporated into the final Index, resulting in a density rather than an absolute value. This approach accounts for area differences between the crop types. Lastly, the Index was scaled twice using (i) overall minima and maxima of all the pesticide groups to rescale the dataset, and (ii) each PUD_j ranges exactly between 0 and 1 by means of the formula $\frac{x - \min(x)}{\Delta \max(x) - \min(x)}$. In this way the pesticide groups are comparable to each other, resulting in a relative Pesticide Use Density (rPUD),

$$PUD_{j,g} = \sum_{i=1}^{k}(PU_{i,j,g} \times CTF_{i,g}) \qquad \text{(Equation 3)}$$

$$\text{CTF} = \text{crop type fraction in \%} = \frac{Cultivated\ Area\ for\ crop\ type\ i_{\ j,g}}{Area\ of\ grid\ cell\ (1000\ m^2\ )}$$

Where, g is 1 km² grid cell, i is the crop type, j is the pesticide group, and k is the number of crop types (k is nine).

2.2.3. Physiochemical Property Index

The chemical properties of the different active ingredients in each pesticide play a crucial role in their pollution capacity (Nicholls 1988; Pino and Dîaz 1998) and are considered in the calculations. Three different properties are included: half-life, water-soil partition coefficient (K_{OC}) and water solubility at 20°C. Half-life and the water-soil partition coefficient were chosen to model and quantify the pesticide mobility capacity, similar to the Surface Water Mobility Index (SWMI) by Chen et al. (2002). In addition to those two parameters, water solubility was added as a parameter motivated by the research by Malaj et al. (2020) modeling a Wetland Pesticide Occurrence Index, who advocated that water solubility is a useful indicator for pesticide mobility within hydrological systems. The properties were approximated for each pesticide group using the sales number (in kg sold) for each active ingredient and information on which crops the pesticides are applied. This information was gathered from the Federal Food Safety and Veterinary Office (BLV 2022). Following equations 4 and 5 results in a weighted mean for each chemical property (Equations 6-8).

First, I calculate the Total Pesticide Use *(TPU)* for each crop. This factor is depending on the cultivated area, the intervention number, and the application rate, and will be used as a weighting factor for the next step.

$$TPU_{i,j} = \sum_{g=1}^{n} PU_{g,i,j} \qquad \text{(Equation 4)}$$

Where, g is 1 km² grid cell, i is the crop type, j is the pesticide group, and n is the number of grid cells (n is 88800 for all of Switzerland).

Second, I assume that the amount of AI used on each crop *(FSA)* is dependent on the sales mass for the AI and a fraction coefficient *(TPU).* The fraction coefficient explains the proportional distribution of how much AI is applied to each crop, i.e. crops with a higher fraction coefficient use more pesticides compared to crops with a lower fraction coefficient.

$$FSA_{a,i,j} = \frac{TPU_{i,j}}{\sum_{i=1}^{k} TPU_{i,j}} \times \ Sales\ Number_{a,j};\ \forall a\ used\ on\ crop\ type\ i \qquad \text{(Equation 5)}$$

Where, a is the active ingredients, *i* is the crop type, *j* is the pesticide group, and *k* is the number of crop types (*k* is nine).

To estimate the final mean value of each physiochemical property (water solubility *(ws)*, half-life *(hl)*, and soil organic carbon-water partition coefficient *(K_{OC})*) for each crop type and pesticide group, I applied a fraction coefficient comprised of the FSA, to approximate the weight of each AI resulting in a weighted mean, see equations 6-8.

$$mean\ ws_{\ i,j}\ =\ \sum_{a=1}^{A}\left(\frac{FSA_{a,i,j}}{\sum_{a=1}^{A} FSA_{a,i,j}} \times ws_a\right); \forall a\ used\ on\ crop\ type\ i \qquad \text{(Equation 6)}$$

$$mean\ hl_{\ i,j}\ =\ \sum_{a=1}^{A}\left(\frac{FSA_{a,i,j}}{\sum_{a=1}^{A} FSA_{a,i,j}} \times hl_a\right);\ \forall a\ used\ on\ crop\ type\ i \qquad \text{(Equation 7)}$$

$$mean\ Koc_{\ i,j}\ =\ \sum_{a=1}^{A}\left(\frac{FSA_{a,i,j}}{\sum_{a=1}^{A} FSA_{a,i,j}} \times Koc_a\right); \forall a\ used\ on\ crop\ type\ i \qquad \text{(Equation 8)}$$

Where, *a* is the active ingredient, *A* is the number of active ingredients, *i* is the crop type, *j* is the pesticide group, *ws* is the water solubility for the active ingredient *a*, *hl* is the half-life for the active ingredient *a*, and *K_{OC}* is the soil organic carbon-water partition coefficient for the active ingredient *a*.

Third, to spatially calculate the final water solubility, half-life, and K_{OC}, the mean physiochemical properties for each crop type and pesticide group are multiplied by the crop type fraction *CTF.*

$$water\ solubility_{j,g} =\ \sum_{i=1}^{k}(mean\ ws_{i,j} \times CTF_{i,g}) \qquad \text{(Equation 9)}$$

$$half-life_{j,g} = \sum_{i=1}^{k}(mean\ hf_{i,j} \times CTF_{i,g}) \qquad \text{(Equation 10)}$$

$$Koc_{j,g} = \sum_{i=1}^{k}(mean\ ws_{i,j} \times CTF_{i,g}) \qquad \text{(Equation 11)}$$

$$\text{CTF} = \text{crop type fraction in } \% = \frac{Cultivated\ Area\ for\ Crop\ Type\ i_{\ j,g}}{Area\ of\ grid\ cell\ (1000\ m^2\ )}$$

Where, *g* is 1 km2 grid cell, *i* is the crop type, *j* is the pesticide group, and *k* is the number of crop types (*k* is nine).

To finally map the Pesticide Property Index (PPI), the physiochemical parameter, water solubility, half-life, and K_{OC} are summed together and divided by three.

$$PPI_{j,g} = (water\ solubility_{j,g} +\ half-life_{j,g} +\ Koc_{j,g}) \times \frac{1}{3} \qquad \text{(Equation 12)}$$

Where, g is 1 km2 grid cell, i is the crop type, j is the pesticide group, and k is the number of crop types (k is nine). Before the parameters were summed, the parameters were scaled twice (i) using overall minima and maxima of each physiochemical parameter of all the pesticide groups to rescale the dataset and (ii) parameter ranged exactly between zero and one for each pesticide group separately using the formula $\frac{x - \min(x)}{\max(x) - \min(x)}$. I purposefully did not rescale PPI, the overall sum of the parameters, from zero to one, to preserve the impact each parameter has independently.

2.2.4. Precipitation

Precipitation can induce pesticide movement and thereby accelerate pesticide distribution over ecosystems and was regarded by Malaj et al. (2020) in their wetland pesticide occurrence prediction efforts. Precipitation is a main driver for pesticide leaching into ground waters especially in structured loamy and heavy clay soils prone to fast preferential water flow in soil macropores (Lewan, Kreuger, and Jarvis 2009). For said reason, precipitation was an important factor to consider when modeling pesticide fate.

The precipitation data retrieved by the Federal Office of Meteorology and Climatology (2019) had an original spatial resolution of $500m^2$. The data was rescaled to match the resolution of the other parameters at 1 km2 spatial resolution. I used the nearest neighbor interpolation (NNI) method, as this method allows aggregating the pixels to the needed spatial resolution. The NNI method is an image interpolation method that uses a source image as a reference to construct a new rescaled image. When performing a digital image interpolation, empty pixels emerge, which are then filled with the nearest neighboring pixel value, hence the name (Rukundo and Cao 2012). Once the pixel resolution was rescaled, I chose to only display the pixels in which the PUDj or PPIj values were positive to avoid creating pesticide occurrence values due to precipitation in areas where no agriculture and therefore pesticide application was even occurring (j stands for the different pesticide groups).

2.2.5. Pesticide Occurrence Index

The Pesticide Occurrence Index (POI) and the relative Pesticide Occurrence Index (rPOI) are comprised of the PUD, PPI and precipitation and rPUD, rPPI and precipitation respectively (Equation 3 and 12). The resulting pesticide occurrence index (POI) is a modeled indicator of pesticide pollution occurrence for Switzerland (Figure 9).

$$POI_{j,g} = (PUD_{j,g} + PPI_{j,g} + Precipitation_g) \times \frac{1}{3} \quad \text{(Equation 13)}$$

$$rPOI_{j,g} = (rPUD_{j,g} + rPPI_{j,g} + Precipitation_g) \times \frac{1}{3} \quad \text{(Equation 14)}$$

Where, j is the pesticide group and g the 1 km^2 grid cell.

Applying a likelihood analysis by means of a composite index can allow different entities to be added together to create one descriptive indicator, the Pesticide Occurrence Index (POI). For this analysis, the three entities pesticide use density (PUD), pesticide property index (PPI), and precipitation are added together to create one descriptive index, analogous to the Wetland Pesticide Occurrence Index by Malaj et al. (2020). The PUD is calculated by means of the pesticide use (PU) of the respective crops, which allows estimations of total applied pesticide mass (TPM). Beside modeling the parameters in an absolute format ranging from zero to one for each separate parameter and pesticide group, the parameters are also calculated in a relative format to compare pesticide group patterns with each other, resulting a relative PUD (rPUD), relative PPI (rPPI) and final relative POI (rPOI).

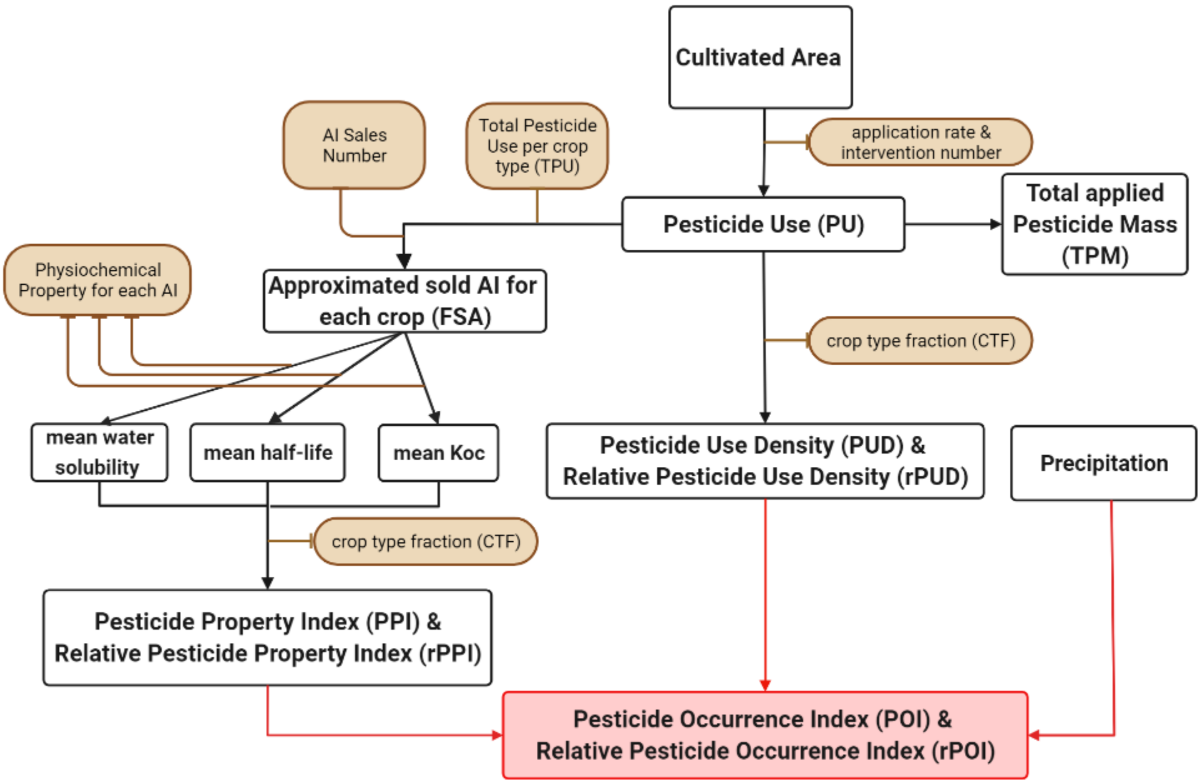


Figure 1: Workflow for the calculation of the Pesticide Occurrence Index. Created with Biorender.com.

2.3. Data

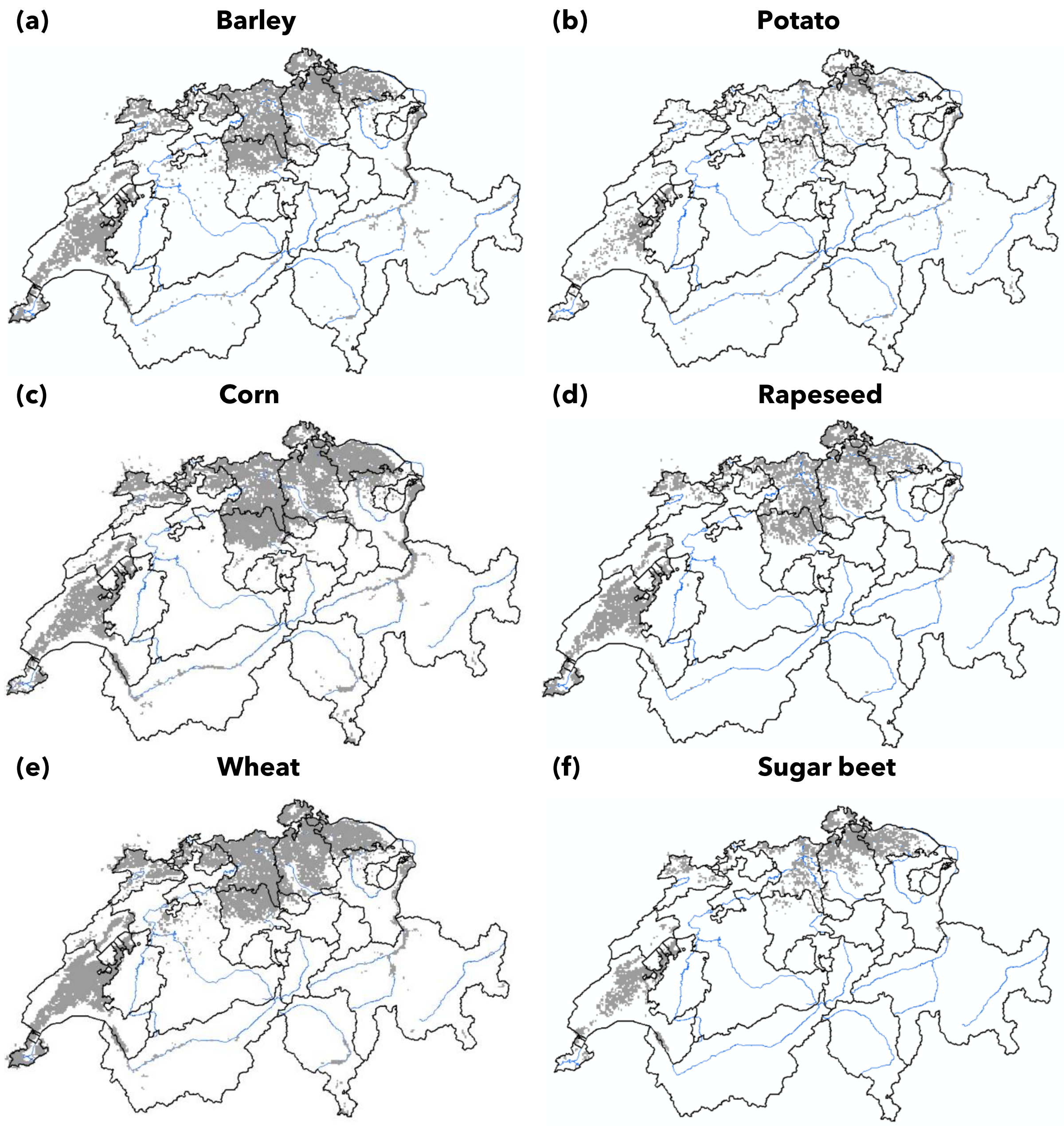
Pesticides: This analysis profited greatly from already aggregated data regarding pesticide application (Table 1). For example, Agroscope was able to provide one single mean application rate and intervention number, using an average from 2009 to 2018 (de Baan et al. 2020) for each crop and pesticide group using an approximation methodology by Baan et al. (2015). For this work, I assume mean application rates and intervention numbers are robust and have therefore not changed since. The application rates and intervention numbers were used to calculate the PU and the PUD. Herbicide, insecticide, and fungicide were the pesticide groups chosen for the analysis in this study as they are the most frequently bought pesticide groups in Switzerland (BLW 2020). The applied AI for each crop type used to calculate the PPI, 166 AI in total, were obtained from the Federal Food Safety and Veterinary Office (state: 2020) providing detailed information on the crops they are applied to and pesticide group they belong. The Federal Office of Agriculture provided sales numbers of the respective AIs for the year 2019. The Pesticide Properties Database created by the University of Hertfordshire supplied the required pesticide properties of each active ingredient which is used in this analysis (Lewis et al. 2016).

Table 1: Intervention number and application rate for the nine investigated crop types and fungicide, herbicide, and insecticide. Adjusted grapevine application rate and intervention number. Data provided by de Baan et al. (2020) from Agroscope.

Intervention number									
	Grapevine	Potato	Stone fruit	Sugar beet	Barley	Wheat	Rape-seed	Corn	Fields
Herbicide	1.2	2.1	1.3	4.1	1.2	1.2	1.3	1.1	0.1
Fungicide	5.3	5.3	5.6	1.2	1.6	1.4	0.8	0.0	0
Insecticide	0.4	1.1	2.1	0.2	0.0	0.1	1.9	0.0	0
Application rate (kg/ha)									
	Grapevine	Potato	Stone fruit	Sugar Beet	Barley	Wheat	Rape-seed	Corn	Fields
Herbicide	1.2	2.4	0.9	5.1	1.5	0.8	1.7	1.4	0.1
Fungicide	17.3	6.3	5.9	0.4	0.9	0.8	0.3	0.0	0
Insecticide	0.3	3.0	2.6	0.1	0.0	0.0	0.2	0.0	0

Land cover types: The Minimal Geodata Model (state: 2021), mapping the different land use areas of Switzerland, was provided by the Federal Office of Agriculture and includes 153 different land use categories (BLW 2021b). The model was used to calculate the cultivated area of each crop type and was utilized in several steps of the analysis (Equation 1,3, 9-11). Agroscope provided mean application rates and intervention numbers for exactly 12 crop types, however the crop types (legumes, pome fruits, and "other grains" were excluded since no respective land use category was designated to them, and therefore I was unable to estimate their cultivated area. The remaining nine crop types which are included in this analysis and possessed land use categories in the Minimal Geodata Model were barley, potato, corn, rapeseed, wheat, sugar beet, grapevine, stone fruits, and fields.

An empty 1 km^2 spatial resolution raster grid covering the extend of Switzerland and containing 88800 empty pixel was intersected with the Minimal Geodata Model map, providing the cultivated area of each crop in each grid cell. The resulting spatial files contains the cultivated area in each grid cell (Figure 2).



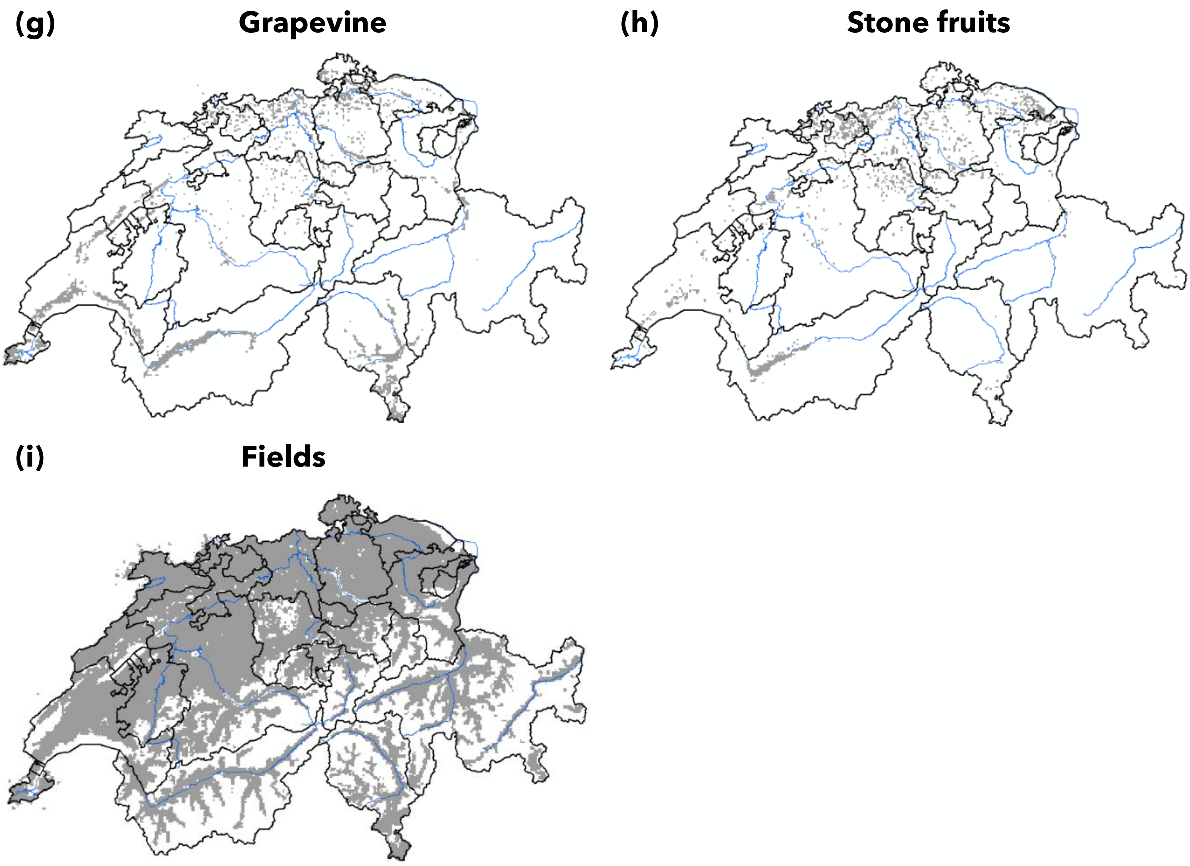


Figure 2: Cultivated areas for the nine investigated crop types (a)-(i).

Climate: A major climate factor for pesticide fate is precipitation, which induces pesticide movement and is a main driver for pesticide leaching into ground waters (Lewan et al. 2009) and was therefore included in this analysis. The precipitation raster data was retrieved from the Federal Office of Meteorology and Climatology (2019) with 500m^2 spatial resolution and represented the average precipitation of the years 1981-2010 (Figure 3). Using the average precipitation values can reduce yearly variation in the data and thereby give a more distinct picture of the expected precipitation. For this analysis, I assume no major changes in precipitation in the mean values.

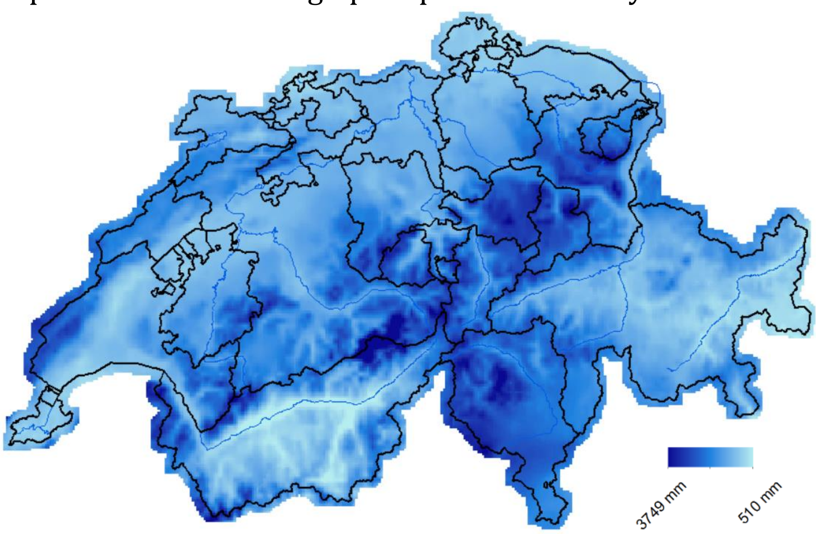


Figure 3: Average precipitation map of 1981 – 2010 of Switzerland in millimeters provided by Federal Office of Meteorology and Climatology.

Table 2: The data collection table presents the descriptions, units, sources, links and in which equations the data was used. The data was collected from various Federal Offices encompassing different years and time frames.

	Description	Units	Source	Link	Equation
Cultivated Planted	Minimal Geodata Model displaying the agricultural landcover types for 2021.	[m^2]	Federal Office of Agriculture (2021) Received by Beat Tschumi on the 30.09.21 via E-Mail	https://www.blw.admin.ch/blw/de/home/politik/datenmanagement/geografisches-informationssystem-gis/minimale-geodatenmodelle.html	1,3,9-11
Application Rates & Number of Interventions	Mean value of intervention and application rate per crop and active ingredient group (2009 - 2018).	[kg/ha] [-]	de Baan et al. (2020)	https://www.agrarbericht.ch/de/umwelt/wasser/verkauf-und-einsatz-von-pflanzenschutzmitteln?_k=4yLaidwL	1
Precipitation	Average precipitation for 1981 – 2010.	[mm]	Federal Office of Meteorology and Climatology (2019) Map: Precipitation 1981-2010 (climate normal)	https://map.geo.admin.ch/?layers=ch.meteosmeteos.klimanormwertk-niederschlag_aktuelle_periode&lang=en&topic=ech&bgLayer=ch.swisstopo.pixelkartefarbe&E=2692020.08&N=1201301.63&zoom=1.217460617038049&catalogNodes=532,628	13,14
Physicochemical Properties					
Utilized active ingredients	Plant Protection Directory describing which active ingredients are used on each crop type.	[-]	Federal Food Safety and Veterinary Office (2020)	https://www.psm.admin.ch/de/produkte	5-9
Sales number of active ingredients	Sold amount of each active ingredient in 2019.	[tons]	Federal Office of Agriculture (2019)	https://www.blw.admin.ch/blw/de/home/nachhaltige-produktion/pflanzenschutz/verkaufsmengen-der-pflanzenschutzmittel-wirkstoffe.html	5
Water solubility at 20° C	Physiochemical property of AI. Explains how easily pesticides are dissolved in water.	[mg/L]	PPDB: Pesticide Properties DataBase (Lewis et al. 2016) State: Jan. 2021	http://sitem.herts.ac.uk/aeru/ppdb/en/atoz_fung.htm#A	6
Half-life (DT_{50})	Physiochemical property of AI. Necessary time for	[days]	PPDB: Pesticide Properties DataBase	http://sitem.herts.ac.uk/aeru/ppdb/en/atoz_fung.htm#A	7

	50% of the initial concentration to degrade.		(Lewis et al. 2016) State: Jan. 2021		
Soil organic carbon-water partition coefficient (K_{OC})	Physiochemical property of AI. Refers to the ratio of pesticide absorbed to pesticide remaining in the soil.	[-]	PPDB: Pesticide Properties DataBase Lewis et al. (2016) State: Jan. 2021	http://sitem.herts.ac.uk/aeru/ppdb/en/atoz_fung.htm#A	8
Topography Digital Height Model (DHM)	DHM25 is a dataset representing the 3D form of the Swiss surface without vegetation and buildings.	[m]	Federal Office of Topography (2004)	https://www.swisstopo.admin.ch/de/geodata/height/dhm25.html#download	Finding POI patterns related to the Swiss topography

2.4. Statistical Analyses

2.4.1. Which areas of Switzerland experience highest pesticide occurrence and pesticide use density?

To find areas and patterns with high pesticide use density and pesticide occurrence, the two parameters were mapped over Switzerland and visually analyzed. Furthermore, to test my first hypothesis that POI will strongly negatively relate to topography, I applied a focal correlation method between the Digital Height Model (DHM) and the POI map. First, I rescaled the DHM of Switzerland from 25m^2 to 1km^2. Second, I applied the focal correlation method, which allows me to detect local relationships between topography and pesticide occurrence. The focal correlation method does not use all the data values of the two maps at once to calculate one overarching correlation value, but instead utilizes each pixel and their respective surroundings separately. This allows us to investigate local patterns more precisely. Correlation is a statistical measure of the linear relationship between two variables X and Y, resulting in a value ranging from -1 to 1, and references the degree to which the variables are linearly related to each other (Akoglu 2018).

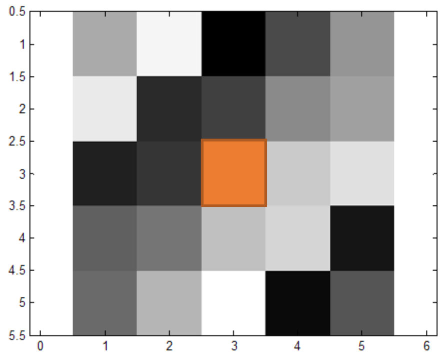


Figure 4: Visualization of a 5×5- pixel neighborhood of the investigated main center orange pixel. Image retrieved from MathWorks, January 2020[1].

For every single pixel location in both parameter maps, a 5×5 pixel neighborhood around it was chosen and correlated with each other, meaning for example that 25 pixels from the POI map and 25 pixels from the DHM map were correlated, resulting in a single correlation value for the investigated central pixel (see orange pixel Figure 4). The degree of correlation therefore indicates what influence elevation has on local pesticide occurrence patters and can be interpreted as to which extent the POI varies with the elevation.

[1] https://ch.mathworks.com/de/company/newsletters/articles/how-matlab-represents-pixel-colors.html

2.4.2. Which pesticide group displays the highest pesticide occurrence and pesticide use density?

To answer my question regarding which pesticide group displays the highest pesticide occurrence and pesticide use density, I used a relative scaling approach, rescaling the parameters to produce the rPUD and rPOI, which allows us to compare the parameter between the three pesticide groups. Additionally, the pixel values of the PUD and POI were plotted in frequency histograms to numerically compare the pesticide groups.

2.4.3. Which crop type has the highest impact on pesticide occurrence and pesticide use density?

To investigate which crop type has the greatest impact on the different pesticide pollution parameters in Switzerland and to answer my research question, I applied an ANOVA analysis in Rstudio. A type III SS analysis, i.e. an analysis that tests the effect of the different crops on the final parameter using the 88800 pixel values as sample size, was chosen to ensure that the crop types were examined independent from their model order. I tested whether PUD, PPI, and the POI varied with crop type.

Throughout the analysis, model errors arose because the final parameters (sum PUD, PPI, and POI) ran out of residuals to compute the sum of squares with, because the sample size of 88800 was rather high. Therefore, the final parameters in each model were rescaled respectively by a factor 1×10^{12} for PUD, 1×10^{6} for PPI, for 1×10^{6} for $POI_{Herbicide}$ to ensure that the models did not run out of residuals to calculate the SS with. Only crop types onto which the respective pesticide group was applied to were included in the ANOVA analysis. For fungicide, the crop types corn and fields, and for insecticide barley, corn, wheat, and fields were removed in the models.

3. Results

3.1. Total applied Pesticide Mass

The total applied pesticide mass (TPM) was calculated to a total of 1770.7 tons over Switzerland for 2019 to 2020, with the assumption that application rates, intervention numbers, and precipitation are robust mean values and therefore did not include major changes. The 1770.7 tons of pesticide were 1235.9 tons of fungicide, 498.8 tons of herbicide, and 36 tons of insecticide (Table 3). Grapevine pesticide application was the highest compared to the other investigated crops with 908.6 tons, which is roughly 51% of the total applied pesticide for all of Switzerland. Sugar beets comprised the second highest pesticide use with 257 tons of pesticides, mainly herbicides.

Table 3: Pesticide Use for each crop type and pesticide group. Pesticide Use numbers from the Swiss agricultural report were reported in the last row for comparative measures.

[tons]	Fungicide	Herbicide	Insecticide	Total
Barley	26.3	32.7	0	58.9
Potato	183.8	28.9	18.2	230.9
Corn	0	69.5	0	69.5
Rapeseed	4.3	39.6	6.8	50.8
Wheat	64.3	55.1	0	119.4
Sugar beet	5.8	251.0	0.2	257.0
Grapevine	893.4	14.0	1.2	908.6
Stone fruit	58.2	2.1	9.6	69.9
Fields	0	5.9	0	5.9
Sum	1235.9	498.8	36.0	1770.7
Agricultural Report 2020*	1006	575	280	1771

3.2. Pesticide Use

I found that the distribution of pesticide use varied greatly over Switzerland (Figure 5). Insecticide displays the lowest pesticide input with 148 kg per grid cell, while fungicides show the highest with 5741 kg per grid cell.

$PU_{Fungicide}$ per grid cell varies from 0 to 5741 kg, which is the highest application amount out of all pesticide groups, and seven times the maximum for herbicides, the second highest pesticide group. The valley of Sion in Valais following the river system of the Rhone, the cantons of Vaud and Geneva, and the river system of the Rhein in the canton of Grisons were all areas in Switzerland with high pesticide use (Figure 5a). These regions correspond to vineyard production in the country (Figure 2g). Therefore, it is not surprising that grapevine production accounted for roughly 72% of the fungicide input (Table 3).

The values for $PU_{Herbicide}$ varied from 0 to 813 kg per grid cell. Areas with high $PU_{Herbicide}$ values are mainly the canton Vaud, northern areas of Thurgau and Zurich, as well as small parts of Fribourg.

The $PU_{Insecticide}$ values in Switzerland ranged from 0 to 178 kg per grid cell which is the lowest compared to any other pesticide group (Figure 5c). The main areas of insecticide input were the valley of Sion in Valais and the Rhein river system. The main driver of $PU_{Insecticide}$ input is, however, potatoes, as they account for half of the insecticide input (Table 3). It can be observed that the high $PU_{Insecticide}$ values are located in areas with grapevine production (Figure 2g, 5c). Furthermore, a large area of Switzerland was not affected at all by any $PU_{Insecticide}$.

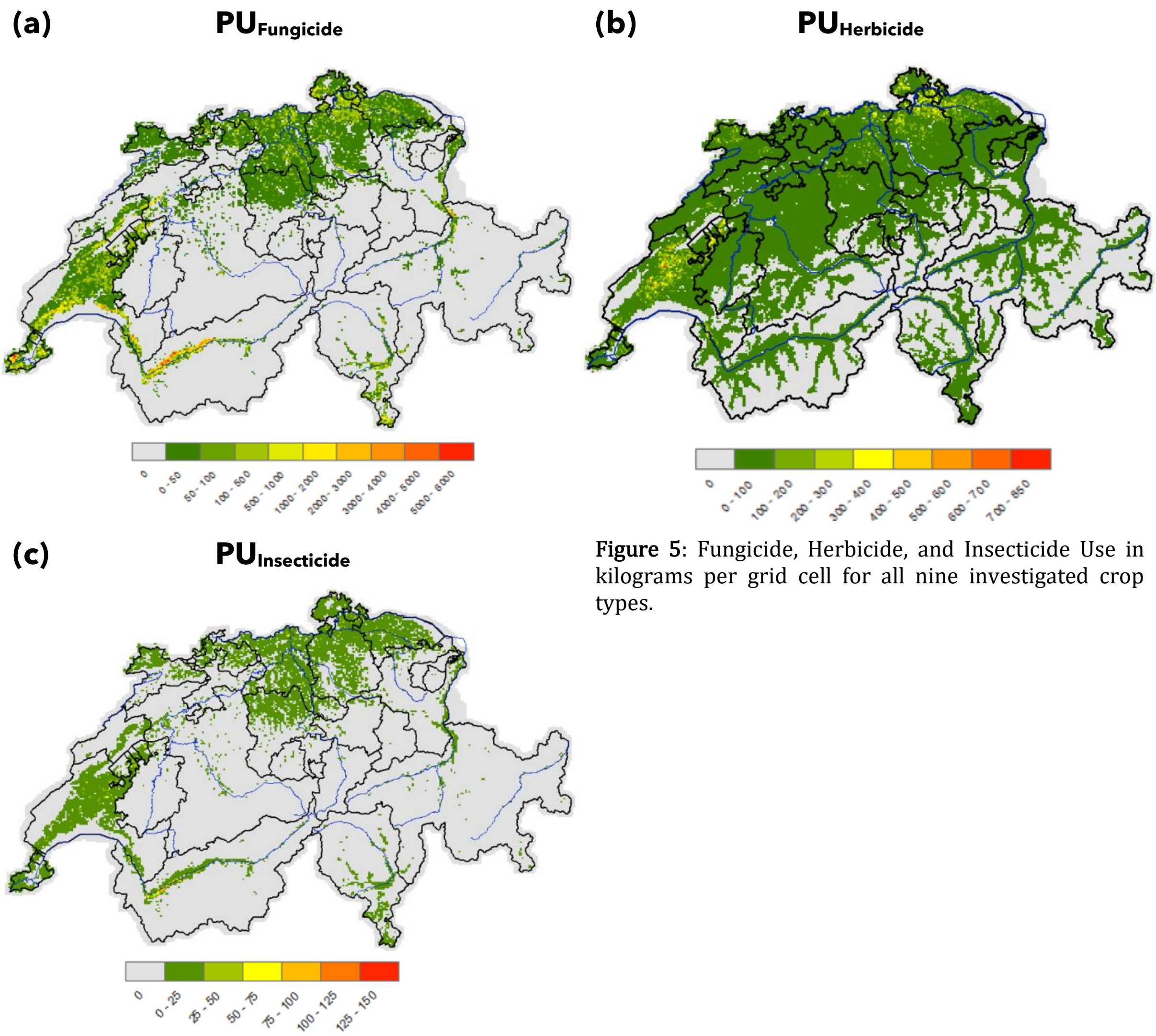


Figure 5: Fungicide, Herbicide, and Insecticide Use in kilograms per grid cell for all nine investigated crop types.

3.3. Pesticide Use Density and relative Pesticide Use Density

3.3.1. Pesticide Use Density

Observing the PUD for the different pesticide groups, the patterns and areas of concern appear to be similar as in the PU map, which is not surprising as the bases of both calculations are similar (Figure 7). However, differences can be allocated for each pesticide group (Figure 5 and 7).

Many areas of high $PU_{Fungicide}$ have now lower $PUD_{Fungicide}$, however, the valley of Sion and following the riverbanks of the Rhone up to the lake of Geneva still display high values. The region of Schaffhausen and Rhein display lower relative values compared to the $PU_{Fungicide}$ values. The $PUD_{Fungicide}$ pattern follows agricultural grapevine production (Figure 2g and 7a). Additionally, the $PUD_{Fungicide}$ experiences the highest number of grid cells with densities above 0.2 compared to herbicide and insecticide (Figure 6).

Areas of high $PUD_{Herbicide}$ values include the region of Schaffhausen, northern Geneva, Zurich, Thurgau, as well as the riverbanks of the Rhone. Yet, Vaud is the canton with highest $PUD_{Herbicide}$ values in Switzerland. The hotspots of high $PUD_{Herbicide}$ visually overlap with agricultural sugar beet production of Switzerland (Figure 2f and 7b).

The $PUD_{Insecticide}$ displays the least amount of grid cells above 0.1 compared to the other pesticide groups (Figure 6b). Only the valley of Sion, the Rhein, and isolated grid cells in Vaud experienced high $PUD_{Insecticide}$ values, very likely due to agricultural grapevine production in these regions (Figure 7c and 2g).

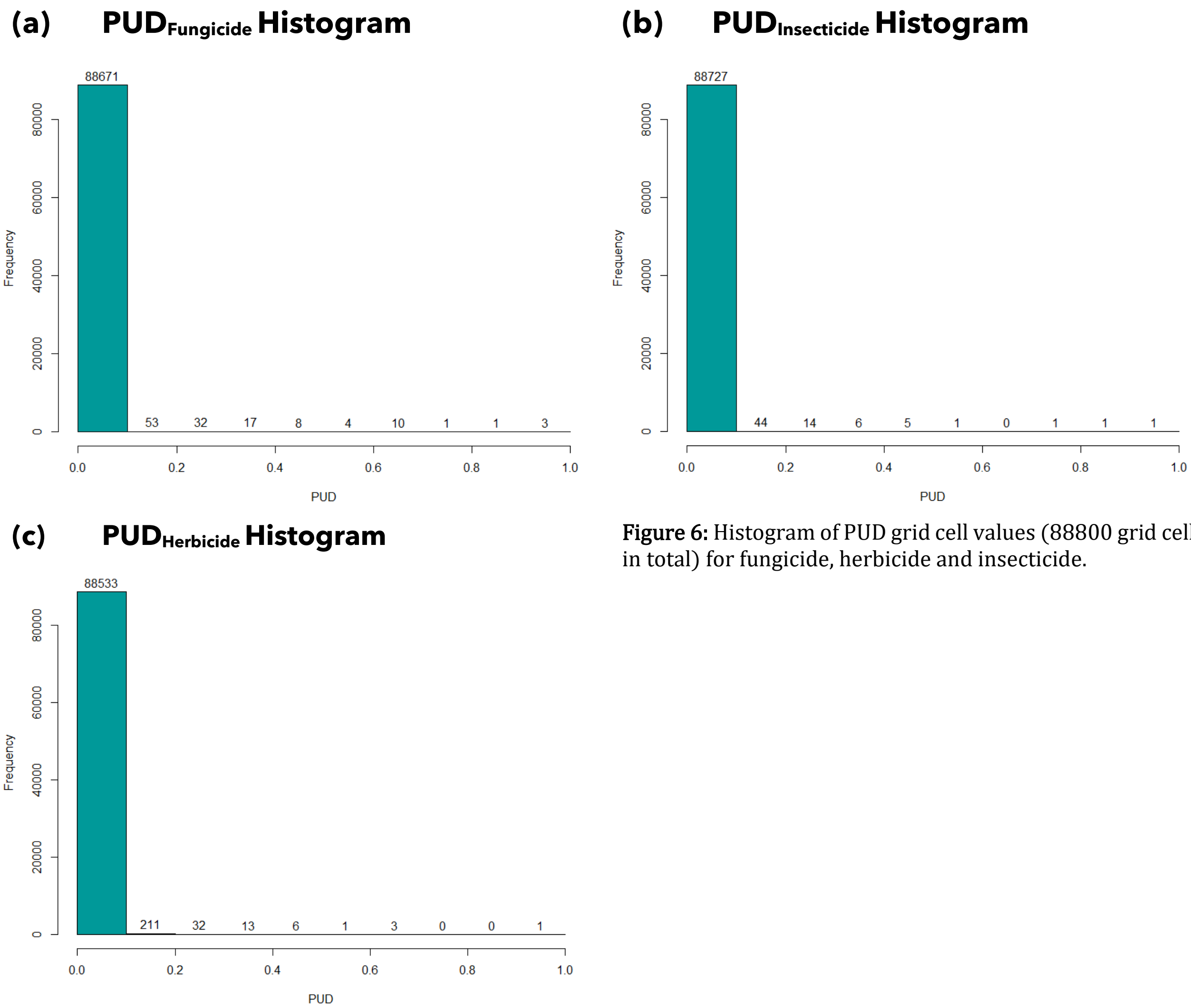


Figure 6: Histogram of PUD grid cell values (88800 grid cells in total) for fungicide, herbicide and insecticide.

3.3.1. Relative Pesticide Use Density

Fungicides display overall the highest $rPUD_{Fungicide}$ values, which range from zero to one, whereas the maximal value for herbicides was 0.079 and 0.011 for insecticides (Figure 7d-f). Because of the scaling method, the $rPUD_{Fungicide}$ and $PUD_{Fungicide}$ are identical.

To find potential areas of high pesticide pollution, only high levels of PUD values will be examined. These values were only found for $PUD_{Fungicide}$, mainly in the valley of Sion, following the Rhone River system to Geneva, and by the Rhein close to Chur, all areas with intensive agricultural grapevine production (Figure 2g). The rPUD for herbicides and insecticides did not display any values above 0.1.

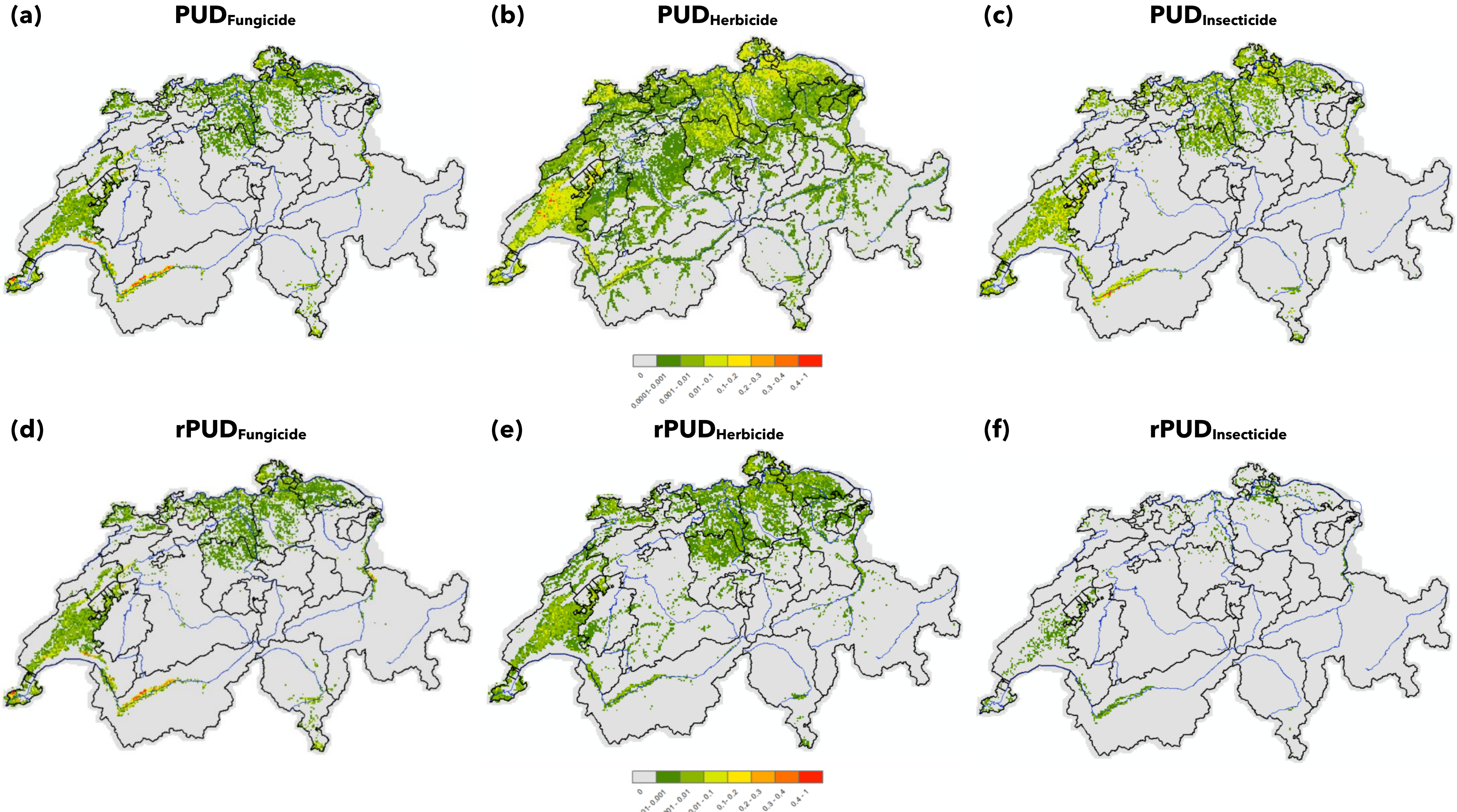


Figure 7: Pesticide Use Density (a) – (c) and relative Pesticide Use Density (d) – (f) for fungicide, herbicide, and insecticide for all nine crop types. The final densities were scaled twice, first in an absolute fashion so that each PUD pesticide map ranged exactly from 0 to 1 (a) – (c) and the second time so that the pesticide groups are comparable to each other (d) – (f).

3.4. Pesticide Property Index and relative Pesticide Property Index

3.4.1. Pesticide Property Index

To examine which properties of pesticides most impact Switzerland, the Pesticide Property Index (PPI) and relative Pesticide Property Index (rPPI) display patterns of negative environmental impacts by accentuating areas of damaging physiochemical pesticide properties, as some active ingredients are more harmful to natural systems than others. The PPI and rPPI for Switzerland are shown in Figure 9.

High $PPI_{Fungicide}$ values are mainly distributed in the mid-region of Vaud and the southern edge of the Neuchatel Lake in Fribourg. A secondary hotspot can be observed in the north of the cantons Zurich and Thurgau, as well as parts of Schaffhausen, all of which are regions of expanded agricultural potato production (Figure 9a). Potatoes had the highest mean water-solubility as well as half-life values of all crop and pesticide groups, which is a potential driver for the high values in the region (Table 4). In the valley of Sion, following the river system of the Rhone, increased $PPI_{Fungicide}$ values are found, most likely due to the intensive agricultural grapevine production in these regions (Figure 2g and 9a). In total, 72 fungicide AIs were applied onto Swiss crops, however, the crops corn and fields did not receive any fungicide treatment, and therefore had zero as their mean chemical property values (Table 4). The PPI for herbicide had the largest number of values above 0.1 followed by fungicide and insecticide (Figure 8a-c).

The $PPI_{Herbicide}$ map displays a pattern unlike any other map plotted thus far. High values are found in the Pre-Alp regions, especially in the cantons of St. Gallen, Appenzell Innerrhoden, Appenzell Ausserrhoden and Fribourg, as well as on the Jura plateau (Figure 9b). The high occurrence of fields in those regions could be the potential driver of high $PPI_{Herbicide}$ values (Figure 2i). In total, 73 herbicide AIs were utilized on crops and herbicides were applied onto all the investigated crop types. The physiochemical properties of the herbicide AIs applied onto fields, stone fruits and grapevines were extremely high, with half-lives for all three crop types above 250 days (Table 4). The combination of high physical pesticide properties and larger areas impacted by these properties most likely led to large regions of high $PPI_{Herbicide}$ values.

The amount of AIs applied in insecticide is only a fraction of those for fungicide or herbicide, with 21 AIs. The insecticide AIs display exceptionally high K_{OC} values, with the overall maxima of all pesticide groups in stone fruits (Table 4). The applied AIs therefore attach themselves efficiently onto organic soil particles rather than water, however the half-life is rather low and does not range above 100 days.

Areas of high $PPI_{Insecticide}$ values are the Valley of Sion at the river system of the Rhone, and in the northern corner of Geneva, both areas of extensive agricultural grapevine production (Figure 2g and9c).

Table 4: Calculated mean water-solubility at 20°C (ws), half-life (hl), and K_{oc} for each crop type. Blue highlighted cells display the high physiochemical properties of grapevine, stone fruits, and fields. Bold numbers represent the maxima of the respective physiochemical property.

[ml, days, -] **Fungicide**	Barley	Potato	Corn	Rape seed	Wheat	Sugar Beet	Grapevine	Stone Fruit	Fields
Mean ws	36	**228665**	0	29	33	30	37096	5765	0
Mean hl	63	**323**	0	54	68	290	129	70	0
Mean K_{oc}	4011	2233	0	8040	3976	4810	2846	1446	0
Herbicide									
Mean ws	29958	514	10833	4012	29690	1573	42073	107857	114756
Mean hl	21	43	24	52	21	17	302	273	259
Mean K_{oc}	1939	4272	1743	3337	1917	323	110486	100499	94690
Insecticide									
Mean ws	0	325	0	645	0	19	0.1	2	0
Mean hl	0	15	0	13	0	64	64	65	0
Mean K_{oc}	0	166086	0	74702	0	8989057	8760301	**9069139**	0

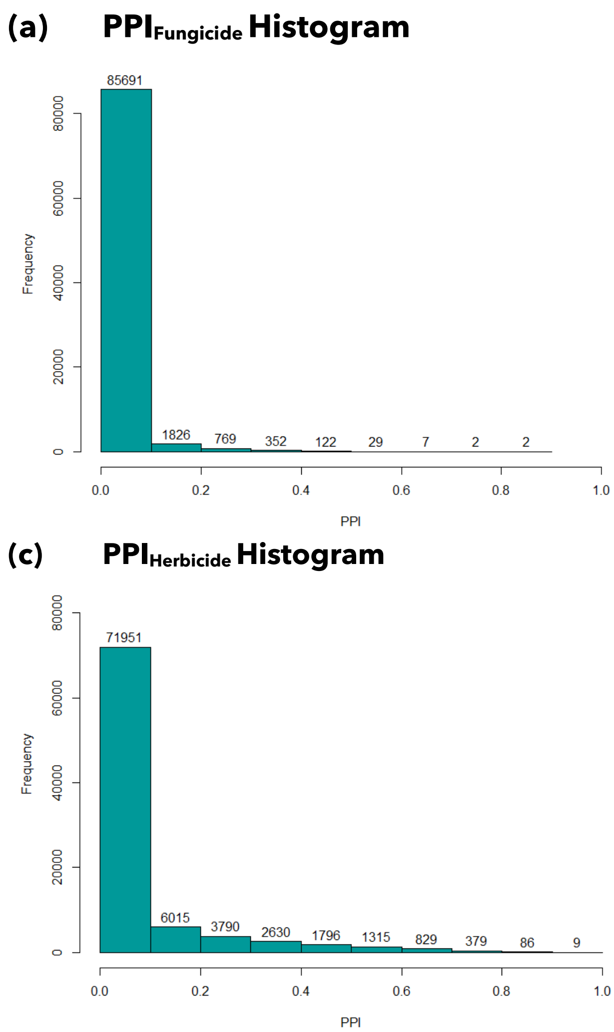


Figure 8: Histogram of PPI grid cell values (88800 grid cells in total) for fungicide, insecticide and herbicide.

3.4.2.Relative Pesticide Property Index

Visually examining the different rPPI maps, it is evident that $rPPI_{Herbicide}$ more frequently displays values above 0.2 (Figure 9d-f). The maximum for $rPPI_{Herbicide}$ is 0.68 and is the highest among all pesticide groups. The hotspot areas for the $rPPI_{Herbicide}$ can be found in the Pre-Alp regions as well as on the Jura plateau in Neuchatel (Figure 9d).

The $rPPI_{Fungicide}$ map displays the lowest values relative to the other pesticide groups, as the maximal value is only 0.11 (Figure 9d). Furthermore, the general grid cell values of the map are comparatively low as well. The maximal $rPPI_{Insecticide}$ value is a little higher with 0.35. The rest of the map is very similar to the $rPPI_{Fungicide}$ patterns (Figure 9f).

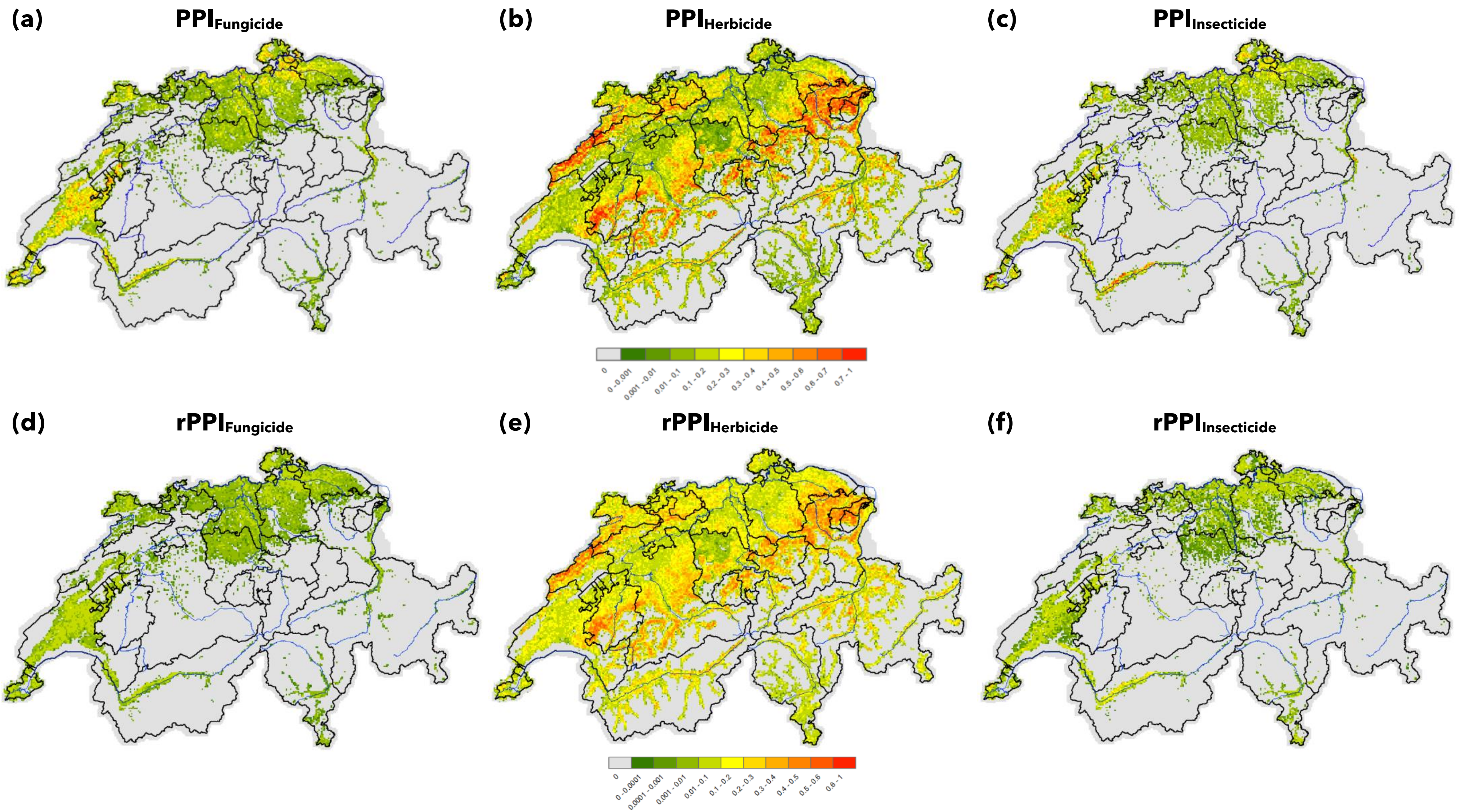


Figure 9: Pesticide Property Index (a) – (c) and the relative Pesticide Property Index (d) – (f) for fungicide, herbicide, insecticide for all nine investigated crop types. The color code used for the maps range from 0 to 1 and was chosen to display extremely small as well as high values. This was achieved by exponentially increasing the values until 0.1 followed by a steady linear increase up to 0.7 for PPI, 0.6 for rPPI. Only few values were found lager than these thresholds, therefore the last coloring interval ranged from 0.7/0.6 to 1.

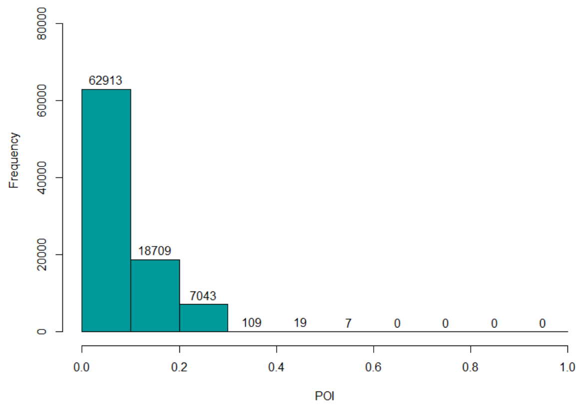
3.5. Pesticide Occurrence Index and relative Pesticide Occurrence Index

3.5.1. Pesticide Occurrence Index

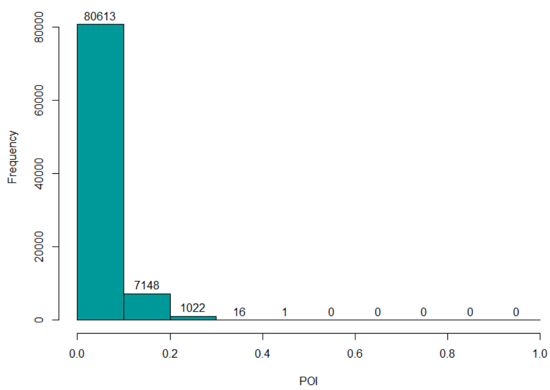
I found that the highest $POI_{Fungicide}$ values occur in the valley of Sion at the river side of the Rhone, and at a single region in the northern part of Geneva (Figure 11a), both locations of agricultural grapevine production (Figure 2g). Similar patterns showed high values for $POI_{Insecticide}$, in the valley of Sion and following the river system of the Rhone as well as the northern part of Geneva (Figure 11c).

In the region of southern Zurich and Zug, the northern parts of Schwyz, the west of St. Gallen as well as areas of Jura show the highest values of $POI_{Herbicide}$ (Figure 11b). The $POI_{Herbicide}$ patterns clearly follow the Pre-Alp region as well as the Jura region, which are both areas with a high frequency of the crop type fields, which yielded high values for the $PPI_{Herbicide}$. The $POI_{Herbicide}$ dispayed the most values above 0.3, clearly indicating the strong effect of herbicide compared to fungicide and insecticide on the pesticide occurrence in Switzerland (Figure 10).

(a) $POI_{Fungicide}$ Histogram



(b) $POI_{Insecticide}$ Histogram



(c) $POI_{Herbicide}$ Histogram

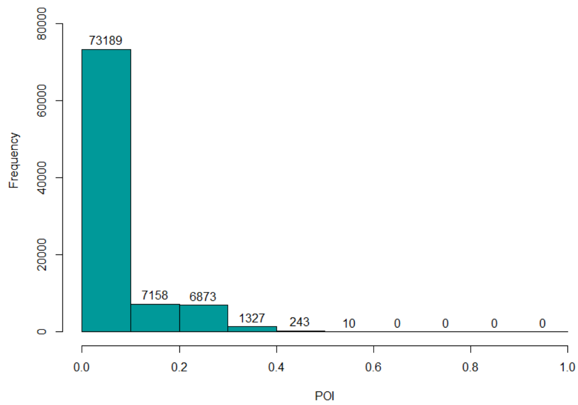


Figure 10: Histogram of POI grid cell values (88800 grid cells in total) for fungicide, herbicide and insecticide.

3.5.2. Relative Pesticide Occurrence Index

The $rPOI_{Herbicide}$ displayed the highest values at 0.46, closely followed by $rPOI_{Fungicide}$ which had a maximum of 0.45. The $rPOI_{Insecticide}$ maximum, however, was considerably lower with 0.29 (Figure 11d-f). The pesticide group with higher values was herbicide, covering the Pre-Alpine and Jura region with high values (Figure 11e).

The $rPOI_{Fungicide}$, $rPOI_{Insecticide}$, and $rPOI_{Herbicide}$ maps all displayed medium-high values in the canton of Ticino. Other areas of high $rPOI_{Fungicide}$ values were found on the river sides of the Rhone, and isolated hotspot regions in northern Geneva (Figure 11d).

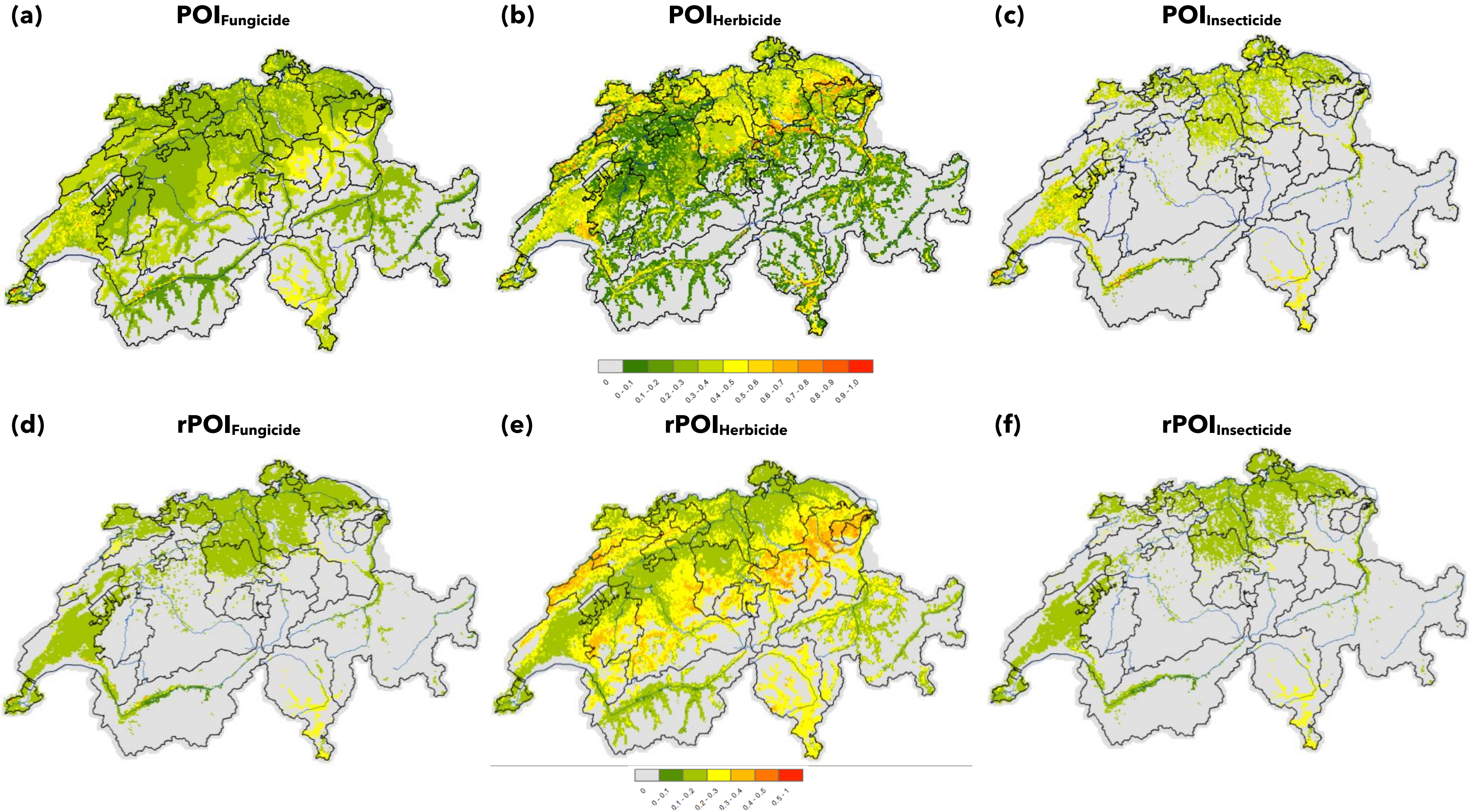


Figure 11: Pesticide Occurrence Index (a) – (c) and relative Pesticide Occurrence (d) – (f) for fungicide, herbicide, and insecticide for all nine investigated crop types. The resulting map was displayed in a linear color code because few small values were present in this index. The legend for the rPOI only reaches 0.5 as higher values were scarce and no further division increased visualization of the index.

3.6. ANOVA

The ANOVA was used to test whether PUD, PPI and POI were influenced by the crop types. Results showed that all the investigated crop types had a significant effect on the final indices.

3.6.1. Pesticide Use Density

I found that for $PUD_{Fungicide}$, all crop types showed a significant effect on the resulting density of the different pesticide groups (Table 5). Out of the tested crop types, grapevines had the strongest effect, followed by potatoes, stone fruits and wheat; these results are in line with the $PUD_{Fungicide}$ map (Figure 7a). The ANOVA analysis for $PUD_{Herbicide}$ showed that for this type of pesticide group, again all crops had significant effect, with the strongest being attributed to sugar beets, followed by grapevines, wheat, and corn. However, the crop types explaining the highest variation for $PUD_{Insecticide}$ were stone fruits and potatoes followed by grapevines and rapeseeds.

Table 5: ANOVA table of all pesticide use densities for all pesticide groups.

$PUD_{Fungicide}$

Response: PUD_F$PUDSmFn * 1e+12

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000e+00	1	1.1288e+02	< 2.2e-16	***
Grapevine	PUD_F$PUDRBFn	1.6486e+25	1	1.1740e+28	< 2.2e-16	***
Potatoes	PUD_F$PUDKTFn	2.4961e+22	1	1.7775e+25	< 2.2e-16	***
Stone Fruit	PUD_F$PUDSOFn	2.0351e+22	1	1.4492e+25	< 2.2e-16	***
Wheat	PUD_F$PUDWFng	2.2970e+21	1	1.6357e+24	< 2.2e-16	***
Barley	PUD_F$PUDGFng	9.4129e+19	1	6.7031e+22	< 2.2e-16	***
Sugar Beet	PUD_F$PUDZRFn	2.5341e+19	1	1.8045e+22	< 2.2e-16	***
Rapeseed	PUD_F$PUDRFng	5.8096e+18	1	4.1371e+21	< 2.2e-16	***
	Residuals	1.2500e+02	88792			

$PUD_{Herbicide}$

Response: PUD_H$PUDSmHr * 1e+12

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000e+00	1	6.8262e+02	< 2.2e-16	***
Sugar Beet	PUD_H$PUDZRHr	7.6309e+24	1	9.7341e+28	< 2.2e-16	***
Grapevine	PUD_H$PUDRBHr	6.4789e+23	1	8.2645e+27	< 2.2e-16	***
Potatoes	PUD_H$PUDKTHr	9.5548e+22	1	1.2188e+27	< 2.2e-16	***
Corn	PUD_H$PUDKMHr	2.9940e+23	1	3.8191e+27	< 2.2e-16	***
Wheat	PUD_H$PUDWHrb	2.6022e+23	1	3.3194e+27	< 2.2e-16	***
Rapeseed	PUD_H$PUDRHrb	7.8492e+22	1	1.0012e+27	< 2.2e-16	***
Barley	PUD_H$PUDGHrb	2.2674e+22	1	2.8923e+26	< 2.2e-16	***
Stone Fruit	PUD_H$PUDSOHr	4.0662e+21	1	5.1869e+25	< 2.2e-16	***
Fields	PUD_H$PUDWWHr	8.9228e+21	1	1.1382e+26	< 2.2e-16	***
	Residuals	7.0000e+00	88790			

PUD$_{Insecticide}$

Response: PUD_I$PUDSmIn * 1e+12

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000e+00	1	1.2627e+02	< 2.2e-16	***
Stone Fruit	PUD_I$PUDSOIn	4.4746e+24	1	4.9224e+27	< 2.2e-16	***
Potatoes	PUD_I$PUDKTIn	1.9992e+24	1	2.1992e+27	< 2.2e-16	***
Grapevine	PUD_I$PUDRBIn	2.2737e+23	1	2.5012e+26	< 2.2e-16	***
Rapeseed	PUD_I$PUDRIns	1.4584e+23	1	1.6043e+26	< 2.2e-16	***
Sugar Beet	PUD_I$PUDZRIn	3.8578e+20	1	4.2438e+23	< 2.2e-16	***
	Residuals	8.1000e+01	88794			

3.6.2. Pesticide Property Index

The crop types explaining the highest SS for the PPI$_{Fungicide}$ were almost identical to the PUD$_{Fungicide}$, which were potatoes and grapevines, followed by sugar beets and wheat. For PPI$_{Herbicide}$ however, fields and grapevines were the crop types explaining the most SS, followed by wheat. Grapevines also explain most of the variation for PPI$_{Insecticide}$, followed by sugar beets and rapeseeds. All crop types were again highly significant for all pesticide groups (Table 6).

Table 6: ANOVA table of all Pesticide Property Indices for all pesticide groups.

PPI$_{Fungicide}$

Response: PPI_F$PPISmFn * 1e+06

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000e+00	1	1.5083e+03	< 2.2e-16	***
Potatoes	PPI_F$PPIKrtF	1.3569e+13	1	2.9545e+21	< 2.2e-16	***
Grapevine	PPI_F$PPIRbnF	1.1639e+13	1	2.5344e+21	< 2.2e-16	***
Sugar Beet	PPI_F$PPIZckF	6.8056e+12	1	1.4819e+21	< 2.2e-16	***
Wheat	PPI_F$PPIWznF	6.7205e+12	1	1.4634e+21	< 2.2e-16	***
Rapeseed	PPI_F$PPIRpsF	3.9845e+12	1	8.6761e+20	< 2.2e-16	***
Barley	PPI_F$PPIGrsF	1.3355e+12	1	2.9080e+20	< 2.2e-16	***
Stone Fruit	PPI_F$PPIStnF	1.1332e+11	1	2.4675e+19	< 2.2e-16	***
	Residuals	0.0000e+00	88792			

PPI$_{Herbicide}$

Response: PPI_H$PPISmHr * 1e+06

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000e+00	1	1.0565e+03	< 2.2e-16	***
Fields	PPI_H$PPIWdWH	1.6543e+15	1	6.2354e+22	< 2.2e-16	***
Grapevine	PPI_H$PPIRbnH	1.5635e+13	1	5.8932e+20	< 2.2e-16	***
Stone Fruit	PPI_H$PPIStnH	8.0734e+11	1	3.0431e+19	< 2.2e-16	***
Wheat	PPI_H$PPIWznH	7.6033e+10	1	2.8659e+18	< 2.2e-16	***
Corn	PPI_H$PPIMsHr	1.6434e+10	1	6.1945e+17	< 2.2e-16	***
Barley	PPI_H$PPIGrsH	1.5783e+10	1	5.9490e+17	< 2.2e-16	***
Rapeseed	PPI_H$PPIRpsH	1.0025e+10	1	3.7787e+17	< 2.2e-16	***
Potatoes	PPI_H$PPIKrtH	1.2672e+09	1	4.7766e+16	< 2.2e-16	***
Sugar Beet	PPI_H$PPIZckH	6.0016e+08	1	2.2621e+16	< 2.2e-16	***
	Residuals	0.0000e+00	88790			

$PPI_{Insecticide}$

Response: PPI_I$PPISmIn * 1e+06

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0	1	5.2159e+03	< 2.2e-16	***
Grapevine	PPI_I$PPIRbnI	3351358463	1	1.6934e+17	< 2.2e-16	***
Sugar Beet	PPI_I$PPIZckI	1205168847	1	6.0895e+16	< 2.2e-16	***
Rapeseed	PPI_I$PPIRpsI	637482359	1	3.2211e+16	< 2.2e-16	***
Stone Fruit	PPI_I$PPIStnI	196903050	1	9.9492e+15	< 2.2e-16	***
Potatoes	PPI_I$PPIKrtI	81987108	1	4.1427e+15	< 2.2e-16	***
	Residuals	0	88794			

3.6.3.Pesticide Occurrence Index

The ANOVA results for POI showed that all crop types had a significant effect. Agricultural grapevine production displayed the highest SS for $POI_{Fungicide}$, followed by potatoes and stone fruits. A high amount of SS for the $POI_{Herbicide}$ ANOVA analysis was explained by fields, followed by grapevines, sugar beet and stone fruit. This is in line with the $POI_{Herbicide}$ maps (Figure 11b). The crop types with the strongest effect on $POI_{Insecticide}$ were stone fruits and potatoes, followed by grapevines. All crop types had a strong significant effect on the final index (Table 7).

Table 7: ANOVA table of all Pesticide Occurrence Indices for all pesticide groups.

POI Fungicide

Response: POI_F$POISmFn

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000	1	5.0670e-01	0.4766	
Grapevine	POI_F$POIRBFn	5.8926	1	6.1169e+06	<2e-16	***
Potatoes	POI_F$POIKTFn	1.6600	1	1.7232e+06	<2e-16	***
Stone Fruit	POI_F$POISOFn	1.4580	1	1.5135e+06	<2e-16	***
Sugar Beet	POI_F$POIZRFn	0.8008	1	8.3125e+05	<2e-16	***
Wheat	POI_F$POIWFng	0.7029	1	7.2971e+05	<2e-16	***
Rapeseed	POI_F$POIRFng	0.4712	1	4.8910e+05	<2e-16	***
Barley	POI_F$POIGFng	0.2359	1	2.4484e+05	<2e-16	***
	Residuals	0.0855	88792			

POI Herbicide

Response: POI_H$POISmHr * 1e+06

		Sum Sq	Df	F value	Pr(>F)	
	(Intercept)	0.0000e+00	1	1.1132e+02	< 2.2e-16	***
Fields	POI_H$POIWWHr	1.7222e+14	1	6.5817e+22	< 2.2e-16	***
Grapevine	POI_H$POIRBHr	2.4007e+12	1	9.1748e+20	< 2.2e-16	***
Sugar Beet	POI_H$POIZRHr	8.6094e+11	1	3.2902e+20	< 2.2e-16	***
Stone Fruit	POI_H$POISOHr	1.0582e+11	1	4.0443e+19	< 2.2e-16	***
Wheat	POI_H$POIWHrb	7.1092e+10	1	2.7169e+19	< 2.2e-16	***
Corn	POI_H$POIKMHr	4.5909e+10	1	1.7545e+19	< 2.2e-16	***
Potatoes	POI_H$POIKTHr	1.8160e+10	1	6.9401e+18	< 2.2e-16	***
Rapeseed	POI_H$POIRHrb	1.7566e+10	1	6.7131e+18	< 2.2e-16	***
Barley	POI_H$POIGHrb	1.2751e+10	1	4.8730e+18	< 2.2e-16	***
	Residuals	0.0000e+00	88790			

POI Insecticide

Response: POI_I$POISmIn

		Sum Sq	Df	F value	Pr(>F)
	(Intercept)	0.0341	1	278.41	< 2.2e-16 ***
Stone Fruit	POI_I$POISOIn	5.8596	1	47847.81	< 2.2e-16 ***
Potatoes	POI_I$POIKTIn	5.4475	1	44482.55	< 2.2e-16 ***
Grapevine	POI_I$POIRBIn	1.7696	1	14450.01	< 2.2e-16 ***
Rapeseed	POI_I$POIRIns	1.6117	1	13160.65	< 2.2e-16 ***
Sugar Beet	POI_I$POIZRIn	0.8084	1	6601.55	< 2.2e-16 ***
	Residuals	10.8740	88794		

3.7. Effect of elevation

I found a clear north-south trend with north facing areas showing more positive correlation values and the south predominately negative correlation values between -1 and 1 (Figure 12). The degree of correlation indicates what influence elevation has on the local pesticide occurrence patterns and can be interpreted as to which extent the POI varies with the elevation. Positive correlation relates to local increasing pesticide occurrence with increasing elevation, meanwhile negative correlation values refer to local decreasing pesticide occurrence with increasing elevation.

This trend is clearer for the $POI_{Fungicide}$ cor. DHM map: the plateau and the Pre-Alps, as well as the eastern parts of Jura follow a distinct positive correlation trend between 0.5 and 1. However, the western part of Jura, the Alps and the southern Alps display negative values between -0.5 and -0.92. The cantons of Zurich and Aarau display little-to-no clear correlation between -0.5 and 0.5. Additionally, it can be observed that along the river systems, there are strong positive correlation values in locations near rivers, while the surrounding shows negative correlation values (Figure 12a).

For the $POI_{Herbicide}$ cor. DHM map, the north displays few positive correlations with index values between 0.5 to 1. The south also shows negative correlations between -0.5 and -0.94. However, the Jura region changed to almost solely negative correlation between -0.5 and -1, while the Vaud plateau remains displaying positive values from 0.5 to 1 (Figure 12b).

The least visible trends were observed in the $POI_{Insecticide}$ cor. DHM map, as the north-south trend is barely-to-non- existent. Only the region of Jura experiences negative values from -0.5 to -0.89, while the west side of the plateau displays positive correlation values ranging from 0.5 to 1. Additionally, the Rhone and Rhein River systems display clear negative correlation values comprised between -0.5 to -0.89 (Figure 12c).

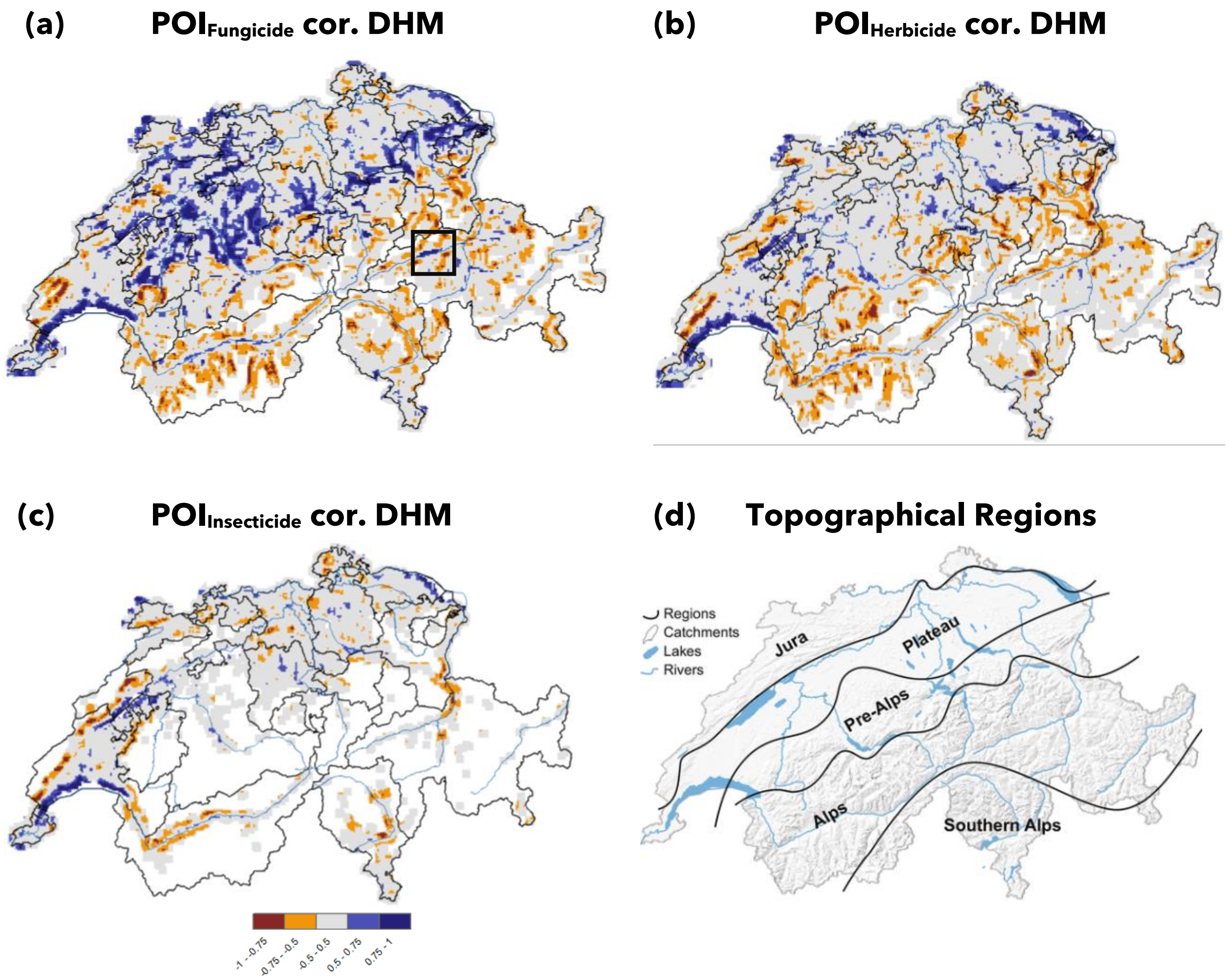


Figure 12: Map of the Pesticide Occurrence Index correlation with Digital Height Model for fungicide, herbicide, and insecticide (a) – (c). The box in the first map, represents the phenomenon of positive values in the riverbed and immediate negative surroundings. In figure (d) the map of Switzerland divided into Topographical Regions, image retrieved from Brunner et al. (2019).

4. Discussion

The total applied pesticide in Switzerland was calculated to be 498 tons of herbicides, 1236 tons of fungicides, and 36 tons of insecticides. The POI and PUD patterns vary by canton and by pesticide groups as well as by elevation. The absolute PUD for fungicides and insecticides was higher in areas in the valley of Sion and following the riverbanks of the Rhone up to the lake of Geneva- all areas of intensive agricultural grapevine production, which I found to be the land use with the strongest effect on PUD (Table 5). For $PUD_{Herbicide}$, the plateau of Vaud experienced the highest densities, driven by increased frequency of agricultural sugar beet production in the region. $POI_{Fungicide}$ and $POI_{Insecticide}$ patterns were very similar, displaying increased values in the plateau of Vaud, the valley of Sion following the river flow of the Rhone up north, and the north corner of Geneva. In contrast, the $POI_{Herbicide}$ patterns displayed different high areas in the Pre-Alp region in the northern part of Switzerland and most of the Jura region, as well as the Rhein and Ticino river systems.

The rPUD and PUD analysis revealed clearly that fungicides accounted proportionally for the highest Pesticide Use. For the POI and rPOI, herbicides presented the highest index values, mainly driven by fields, but all crop types had a significant effect on the POI values in general. I found that grapevines had the strongest effect, followed by potatoes, stone fruits and wheat for the $PUD_{Fungicide}$. However, stone fruits and potatoes had the strongest effect on $PUD_{Insecticide}$. Furthermore, the analysis for $PUD_{Herbicide}$ showed that the highest explaining variable can be attributed to sugar beets, followed by grapevines, wheat, and corn. Sugar beets contributed 251 tons of herbicide input, which is about half of the calculated total herbicide input of Switzerland. Grapevines accounted for 893 tons out of the 1235 tons of applied fungicide input and had the strongest impact on the $POI_{Fungicide}$. More than half, 18.2 tons of out of the 36 tons, of the insecticide input were accounted for by potato cultivation (Table 3), which had the second strongest influence on the $POI_{Insecticide}$ after stone fruits. Fields accounted for only 5.9 tons of herbicide application however had the highest impact on the $POI_{Herbicide}$ followed by grapevines.

The calculated Total applied Pesticide Mass resulted in values very similar to those reported by the Swiss Agricultural Report of 2020 (BLW 2020); however, fungicide application values were overestimated by about 230 tons while herbicide and insecticide numbers were underestimated. However, there is a difference in the data that was used in the calculations presented and those in the Swiss agricultural report from 2020: here I approximated the mass through application rate, intervention numbers and cultivation area while the agricultural report used sales numbers and not application amounts due to a lack of specific application data (BLW 2020). Even though adjustments were made, fungicide application numbers were still overestimated which can potentially be attributed to an overestimation of commercial grapevine production. Organic grapevine production today uses little to no fungicides, and accounts for 13.3% of the total agricultural grapevine production in Switzerland (BLW 2021a), and was not considered in this analysis but nevertheless decreases fungicide application mass. Still, my results agree with those from the Swiss Agricultural Report (2020) where fungicide and herbicide were identified as the main contributors to pesticide application mass. An overall decreasing trend in sales of pesticides was identified from 2008 to 2020 in Switzerland, with herbicide sales dropping by 33%, fungicide sales increasing by 6%, and with no clear trend for insecticide (BLW 2020). This was due to current agricultural policies in Switzerland in place since 2018, which incentivize farmers through compensation strategies, to use production systems with no herbicides in combination with prevention efforts against soil degradation (Böcker, Möhring, and Finger 2019).

As expected, the $rPUD_{Fungicide}$ displayed the overall greatest densities compared to the other pesticide groups due to high application rates and intervention numbers in stone fruits and grapevines. The regions with extensive vineyard production displayed, as expected, higher $POI_{Fungicide}$ values compared to any other crop in the region. This is most likely a consequence of the multitude of pests and diseases that have grapevines as their favorite host and preferred niche. Therefore, intensive pesticide application is generally required to upkeep the quality and quantity of the grapes (Pertot et al. 2017). Additionally, in contrast to other annual crops, most grapevine production cannot benefit from crop rotation or radical change of the cropping system, making it harder to protect the crop from pests (Pertot et al. 2017).

The pesticide occurrence shows a strong north-south correlation pattern within the Swiss landscape. This contradicts the expectations as it was expected that the Swiss topography would be negatively correlated with the POI values, meaning increasing elevation would lead to lower pesticide occurrence due to decreased agricultural suitability. This was the case for mountainous areas leading to negative correlation values. Decreased agricultural suitability with increasing elevation can be caused by shorter growing seasons, decreased temperature (Holzkämper, Calanca, and Fuhrer 2013), reduced soil fertility, lower soil carbon content (Jiang and Thelen 2004; Prasuhn et al.

2013), increased erosion and mass movement due to slopes (Prasuhn et al. 2013), and the difficult terrain for machinery However, focusing on non-mountainous areas, agriculture suitability and therefore pesticide occurrence increased with rising elevation. Potential drivers of increased suitability with rising elevation in these regions could be rising soil thickness and stronger solar radiation, however further research on the drivers of agriculture suitability with elevation in non-mountainous areas could help predict pesticide occurrence. The elevation threshold which separated the two opposite trends can be observed in the clear Pre-Alp line (Figure 12).

Even though applied herbicide mass was only 5.9 tons for fields, it had a larger effect on the POI for herbicide compared to the 251 tons of applied herbicides by sugar beet cultivation. The impact of fields on the POI even surpassed the effect of the 893.4 tons of applied fungicide on grapevines. The strong effect of fields on the POI and rPOI values was a surprising find, considering that the application rate of herbicides on fields was only 0.1 kg/ha. However, fields cover the largest agricultural area in Switzerland and, in combination with the exceptionally high physiochemical properties of the active ingredients, led to substantial areas with increased $PPI_{Herbicide}$ values and subsequently POI values. Future adjustments, potentially reducing the impact of the PPI to the PUD or on-site validation of the herbicide impact due to fields could increase the accuracy and predictability of the index.

Nevertheless, the high risk of herbicides compared to fungicides and insecticides represented by high POI and rPOI values was in line with recent field measurement findings. A study by Riedo et al. (2021) analyzing 100 fields under organic and conventional management found 46 pesticides, 16 herbicides, 8 herbicide transformation products, 17 fungicides, and 7 insecticides. Doppler et al. (2017) and Spycher et al. (2019) each investigated five small surface waters sites with agricultural catchment areas across Switzerland. Doppler et al. (2017) found 128 pesticides, 61 herbicides, 45 fungicides and 22 insecticides. In total 32 pesticides transgressed the legal 0.1µg/l legal quality limit, of which 17 were herbicides, 11 were insecticides and 4 were fungicides. Spycher et al. (2019) found 31 pesticides which transgressed legal quality limit within their samples and a total of 145 pesticides across all sites, 50 herbicides, 39 fungicides, 10 insecticides and 1 molluscicide. Wittmer et al. (2014) investigated five medium sized surface waters for polar organic-synthetic pesticide, which represent half of the sold pesticides in Switzerland, and found 104 different pesticides in total, 54 herbicides, 31 fungicides and 17 insecticides.

The POI was able to predict the risk of herbicide occurrence by incorporating the PPI into the Pesticide Occurrence Index, which was validated by the literature above. An analysis only focusing on the pesticide use of Switzerland could not have predicted the herbicide risks for the Swiss environment. The potential of toxicity through persistence and mobility clearly overshadowed the impact of application mass on the index, which was a surprising finding.

While these results show for the first time a spatial map approximating the distribution of pesticide incidence over the country, several considerations about data quality need to be discussed. First, this analysis was only possible due to the availability of the open-source data in Switzerland, which is plentiful. However, the precision of the calculation could profit from more comprehensive data detail in many regards. For example, about 25 % of the polygons in the land cover map provided by the Federal Office of Agriculture were empty and lacked any description, potentially resulting in uncertainties, underestimations, and imprecisions regarding the extent of the agricultural areas. In total, the computed cultivated area was underestimated by 613 km^2, which is a deficit of about 7.5%. Furthermore, in the calculations presented herein, I have main differences in the calculated agricultural areas of rapeseed, potatoes, barley, and grapevines compared to the Swiss agricultural report (2020) (Table 8), which could be due to the "empty" cells in the land use dataset. All cantons besides Jura, Neuchatel and Valais had "empty" polygons in their extent, some of which could be agricultural areas. Basel displayed the highest count of empty polygons with nearly 60% of undefined polygons (see Appendix, Table 1).

Table 8: Calculated cultivated area for each crop type compared to reported area in km^2 retrieved from the Swiss Agricultural Report (2020).

	Agricultural Report [km^2]	Calculations [km^2]	Δ	%
Barley	269	182	- 87	- 32
Potatoes	110	55	- 55	- 50
Corn	627	451	-176	- 28
Rapeseed	1	179	178	178
Wheat	803	574	- 229	- 29
Sugar beets	176	120	- 56	- 32
Grapevines	147	97	- 50	- 34
Stone fruits	17	18	1	6
Fields	6019	5880	- 139	- 2
Total cultivated area	8169	7556	- 613	- 7.5

Second, many other crop types beside the nine considered are cultivated in Switzerland e.g. spelt, oats, millet, soy, sunflowers, peas and many others which account for roughly 370 km2 of agricultural land which was not accounted for in this thesis (BLW 2020). Third, one cause for overestimation of use and impact of pesticides might be rooted in the disregard for organic agricultural areas. Their production tends to use little to no pesticides, however no distinction between commercial and organic agricultural production was made for this analysis. This distinction potentially led to an overestimation in pesticide use, particularly in agricultural grapevine production, as organic grapevine production has become more popular in Switzerland (BLW 2020).

Fourth, fungicides, herbicides and insecticides are not the only pesticide groups applied onto crops in Switzerland. In the country, bactericides (against bacteria), algicides (against algae), molluscicides (against snails), nematicides (against nematodes), rodenticides (against rodents), miticides (against mites) are a few of many other important pesticide groups that can contain pests and contaminate the environment, but were not regarded in the analysis, potentially further underestimating the likelihood of pollution.

Fifth, pesticides can be applied by different application methods, and the contamination risk with each method varies greatly. For this study I made no distinction regarding the different application methods or point source vs. diffuse source input. Besides the general arrangement and makeup of the land cover types, crop types, and pesticide groups, more specific refinements could be implemented specifically for the PUD and the PPI. One of the fundamental elements for the PUD calculations is the application rate and intervention numbers for each crop type and pesticide group. The same number was applied regardless of the location in Switzerland. This can cause inaccuracies since regional variation due to climate, soil quality, and pest occurrence should be accounted for. This generalization greatly reduces regional precision and could be greatly improved by using local pesticide application rates and intervention numbers. Besides increasing the precision of the PUD, increasing the precision of the PPI could further raise the accuracy of the calculations of the POI. The PPI visualized the potential risks that AIs display, by persisting in the soil or moving offsite and therefore polluting waterbodies, using the physiochemical properties of water-solubility, half-life and K_{OC} of the AI. Many studies have shown the long-term environmental risks of persistent and water-soluble pesticides compared to rapid degrading pesticides (Bonmatin et al. 2015; Hladik, Kolpin, and Kuivila 2014).

Finally, due to the lack of exact application numbers for AIs on crops, sales numbers were again used as a proxy. However, knowing the exact amount of AIs that are applied on each crop type would give insight into the risk factor they pose. Unfortunately, exact application numbers of the AIs were not available, hence sales data and the plant protection register of the Federal Food Safety and Veterinary Office were used to estimate the AIs applied. The pesticide property databank helped to identify the pesticide properties of the AIs. 166 AIs were applied in total, along with 72 fungicides, 73 herbicides and 21 insecticides. However, missing data within the databank was a recurring problem as 16 AIs displayed one or more NaNs in their properties. Additionally, for the K_{OC} two different isotherms (the Linear and Freundlich isotherms) were used interchangeably. Even though they are very similar and explain a parallel process, they refer to two different approaches and ideally should not be compared to each other. Water solubility, half-life and K_{OC} are meaningful descriptors for soil and groundwater pollution risk, however no specific chemical toxicity values were included in the PPI. Overall, the discrepancies emphasize the urgency for more accurate data regarding pesticide use and their respective active ingredients. This would allow for more accurate and regional variability to be displayed that relies less on approximation and extrapolation of incomplete and coarsely-scaled datasets.

Precipitation highly impacts pesticide fate as more frequent and intense precipitation events have shown to lead to movement of pesticides from their application point to surface waters (Lewan et al. 2009). The nature of the water movement is greatly interlinked with the corresponding soil structure. Linking those two parameters together would greatly improve the precision of the predictability of the pesticide fate through water movements. Besides precipitation close to mountain surfaces, runoff from snowmelt, for example from spring snow melt, may be crucial to pesticide movement and should be included in future analysis in these regions. I acknowledge that many other factors could improve the calculations, for example specifying vegetation, topography, and chemical features of the AIs.

To visualize the variability within the datasets and between the parameters in the final POI, the parameters need to vary within the same interval (zero to one) and ought to follow a normal distribution. Neither the PUD nor the POI displayed such a distribution. The visualization of the POI would greatly profit from modifying the parameter distributions. To be sure, implementing different approaches could improve the data distribution. Throughout my work I tried logging the respective parameters, but this proved more counterproductive as it did not yield the intended results. For further research, I highly suggest a scaling approach in which certain value intervals are scored and only afterwards scaled. Especially the PPI would profit from such an approach, as using a ranking system could make the results of the PPI more meaningful. One could clearly identify within which chemical property and risk interval the grid cell is located, compared to the current investigation technique.

Overall, the relatively simple Pesticide Occurrence Index incorporating Pesticide Use, Physiochemical Properties and precipitation varied with topography, crop type and pesticide group. Most of the utilized pesticides in Switzerland, however, were fungicides and were applied on grapevines in the valley of Sion, following the river system of the Rhone. Herbicides were applied onto all nine investigated crops, resulting in a large application area. High herbicide use mainly converged in the canton of Vaud and was driven by sugar beet production. Insecticide use was negligibly low compared to the other two pesticide groups and mainly driven by potato cultivation. Crop types had varying strong effects on the PUD, PPI and POI, however, grapevines, fields, potatoes, and stone fruits were the crop types that influenced the parameters the strongest.

The POI was able to predict the impact of herbicides on the Swiss environment discovered through field research efforts by modeling the spatial patterns of the persistence and mobility of the applied active ingredients using the PPI. The PPI revealed high physiochemical properties of the active ingredients within herbicides applied on fields, grapevines, and stone fruits. Mean half-life for the active ingredients applied on said crops all surpassed 250 days. High PPI values were found in the northern Pre-Alp regions as well as in the canton of Vaud and Jura, all areas of cultivated fields. Consequently, the POI for herbicide which included the PPI for herbicide was strongly driven by the PPI distribution. Investigating and adjusting the disproportionate large impact of the PPI through fields, in respect to the application amount of only 5.9 tons, could advance the precision of the POI.

The Swiss topography correlated positively with the POI in the north, and negatively in the south, separated by the Pre-Alp line (north-south trend). The contradictory trends are driven by agricultural suitability of the topography. However, further research regarding the driver of agricultural suitability with increasing elevation in non-mountainous areas could indicate factors which influence pesticide occurrence in these regions.

The POI provides a unique tool for the assessment of early identification of probability of pesticide occurrence which can incentivize field sampling efforts and help policy makers make informed decisions. However, the POI would greatly profit from on-site validation to further examine the strength and weakness of the index. Additionally, incorporating soil types in combination with the precipitation could better predict the effect of leaching.

5. Conclusion

Transgressing legal quality limits, pesticides have been found in Swiss surface waters and fields and have shown to negatively impact a diverse group of Swiss ecosystems, including humans (Doppler et al. 2017, 2020; Riedo et al. 2021; Spycher et al. 2018, 2019; Wittmer et al. 2014). However, monitoring efforts thus far have been performed on isolated sampling sites. A model of the continuous spatial extent of the pesticide pollution problem in Switzerland was missing. In this thesis, I established an index which mapped the occurrence of pesticides over all of Switzerland utilizing (i) application rates and intervention numbers for fungicide, herbicide and insecticide, and cultivated area of barley, potato, wheat, corn, sugar beet, rapeseed, stone fruit, grapevine, and fields (ii) three physiochemical properties of the applied active ingredients and (iii) precipitation. I was interested in finding areas and patterns with high pesticide occurrence, regarding elevation as a potential factor. Additionally, I questioned the impact of the nine different crop types and three pesticide groups on the pesticide occurrence index.

Little-to-no pesticide occurrence was found in the Swiss Alps. In the Alps, pesticide occurrence decreased with increasing elevation, due to declining agricultural suitability of the landscape following rising elevation. In the low, elevated north, however, rising elevation did not increase pesticide occurrence. Further research into the driving elements of agricultural suitability and elevation impacting pesticide occurrence could increase predictability of the index. Separating these two opposite trends is a threshold elevation level which follows the Pre-Alp line. The north-east and west of Switzerland as well as river systems and lake sides all displayed a high likelihood of pesticide occurrence. The different crop types had varying effects on the pesticide occurrence and use. Crops with higher pesticide application, such as grapevines, or crops which were treated with active ingredients possessing high physiochemical properties, for example fields, impacted the pesticide occurrence substantially.

Fungicide application accounted for roughly half of the applied pesticide mass in Switzerland, most of which applied to grapevines; however high pesticide occurrence was driven by herbicide use, due to high persistence and mobility of the active ingredients within herbicides. Future research regarding establishing and implementing mitigation strategies to reduce fungicide use, specifically on grapevines, could be valuable work to reduce pesticide pollution. Further incentivizing the creation and use of less damaging herbicides could lower environmental contamination in Switzerland significantly.

Future efforts must also include model validation by means of a cross correlation of the index with field measurements to provide important insight regarding the quality and precision of the index. In addition, combining the soil type and precipitation of the cultivated area into a single parameter could increase predictability of leaching more accurately.

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Appendix

Table 1: Overview of number of polygons with undefined land use categories for each canton within the Minimal Geodata Model from BLW (2020).

Canton	Empty polygons	Total polygons	%
Basel	45361	77769	58.33
Grisons	243862	445763	54.71
Appenzell Ausserrhoden	19614	40952	47.9
Schaffhausen	20619	43839	47.03
Bern	152415	324206	47.01
Solothurn	31053	66543	46.67
Zug	16148	35231	45.83
Fribourg	35435	82914	42.74
Thurgovie	45422	110625	41.06
Lucerne	81368	205612	39.57
St Gall	86156	218520	39.43
Glarus	2046	15596	13.12
Nidwald	2885	24831	11.62
Appenzell Innerrhoden	1315	13403	9.81
Vaud	36808	435678	8.45
Schwyz	2354	37942	6.2
Obwald	959	18231	5.26
Uri	251	12368	2.03
Ticino	714	59594	1.2
Zurich	673	129145	0.52
Argovie	82	101008	0.08
Geneva	0	11190	0
Jura	0	173147	0
Neuenburg	0	125676	0
Valais	0	422453	0
Total	825540	3232236	25.54

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Personal Declaration

I hereby declare that the submitted Thesis is the result of my own, independent work. All external sources are explicitly acknowledged in the Thesis.

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Zurich, 31.03.2022 Noemi Shavit