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Albrecht Benzing (albrechtbenzing@yahoo.com)
CERES GmbH
Hans-Peter Piepho
University of Hohenheim
Waqas Ahmed Malik
University of Hohenheim
Maria R. Finckh
University of Kassel
Manuel Mittelhammer
CERES GmbH
Dominic Strempel
CERES GmbH
Johannes Jaschik
Eurofins Dr. Specht International GmbH
Jochen Neuendorff
GfRS GmbH
Liliana Guamán
CERES GmbH
José Mancheno
CERES GmbH
Luis Melo
CERES GmbH
Omar Pavón
CERES GmbH
Roberto Cangahuamín
CERES GmbH
Juan-Carlos Ullauri
CERES GmbH

Research Article

Keywords: Organic certification, Pesticide residues, Spray-drift, Sampling, Discriminant analysis, Supervised learning

Posted Date: March 30th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-357778/v1

Version of Record: A version of this preprint was published at Scientific Reports on July 20th, 2021. See the published version at https://doi.org/10.1038/s41598-021-93624-8.

Telling Apart the Bad from the Good Guys Behind the Spraying Mist

Improved Sampling and Pesticide Residue Testing for Organic Farm Compliance Assessment

Albrecht Benzing[1*], Hans-Peter Piepho[2*], Waqas Ahmed Malik[2], Maria R. Finckh[3*], Manuel Mittelhammer[1], Dominic Strempel[1], Johannes Jaschik[4], Jochen Neuendorff[5], Liliana Guamán[1], José Mancheno[1], Luis Melo[1], Omar Pavón[1], Roberto Cangahuamín[1], Juan-Carlos Ullauri[1] (all addresses below are in Germany)

[1]CERES GmbH, Vorderhaslach 1, 91230 Happurg, albrechtbenzing@yahoo.com

[2]Biostatistics Unit, Institute of Crop Science, University of Hohenheim, 70593 Stuttgart

[3]Department of Ecological Crop Protection, University of Kassel, Nordbahnhofstraße 1a, 37213 Witzenhausen

[4]Eurofins Dr. Specht International GmbH, Am Neulaender Gewerbepark 2, 21079 Hamburg

[5]Gesellschaft für Ressourcenschutz (GfRS), Prinzenstr. 4, 37073 Göttingen

Abstract

Residues of pesticides not allowed in organic farming are often found in organic food. A large number of samples are being tested by organic certifiers, but the sampling methods often do not allow to determine if such residues stem from prohibited pesticide use by organic farmers, from mixing organic with conventional products, from short-range spray-drift from neighbour farms, from the ubiquitous presence of such substances due to long-distance drift, or from other sources of contamination. Eight case studies from different crops and countries are used to demonstrate that sampling at different distances from possible sources of short-distance drift allows in most cases to differentiate deliberate pesticide application by the organic farmer from drift. Datasets from 67 banana farms in Ecuador, where aerial fungicide spraying leads to a heavy drift problem, were subjected to statistical analysis. A linear discriminant function including four variables was identified for distinguishing under these conditions application from drift, with an accuracy of 93.3%.

Keywords

Organic certification, Pesticide residues, Spray-drift, Sampling, Discriminant analysis, Supervised learning

1 Introduction

1.1 Pesticide Residues in Organic Products

Non-use of synthetic pesticides is a major characteristic of organic farming, with the objectives of protecting (a) the environment, (b) consumer health, and (c) farm worker health. In consumer studies, "no chemical pesticides" is usually mentioned as one of the most important criteria for buying organic food.[1,2] These consumer expectations are mostly met in what is referred to in objective (b), because the levels of pesticide residues in organic food offered in retail stores are low. The food authority in Baden-Württemberg, Germany, has been comparing pesticide residues between organic and conventional products since 2002. Taking the example of fruits and vegetables from 2013 to 2019, on average the residues were more than 100 times lower than in conventional vegetables[3] (Figure 1).

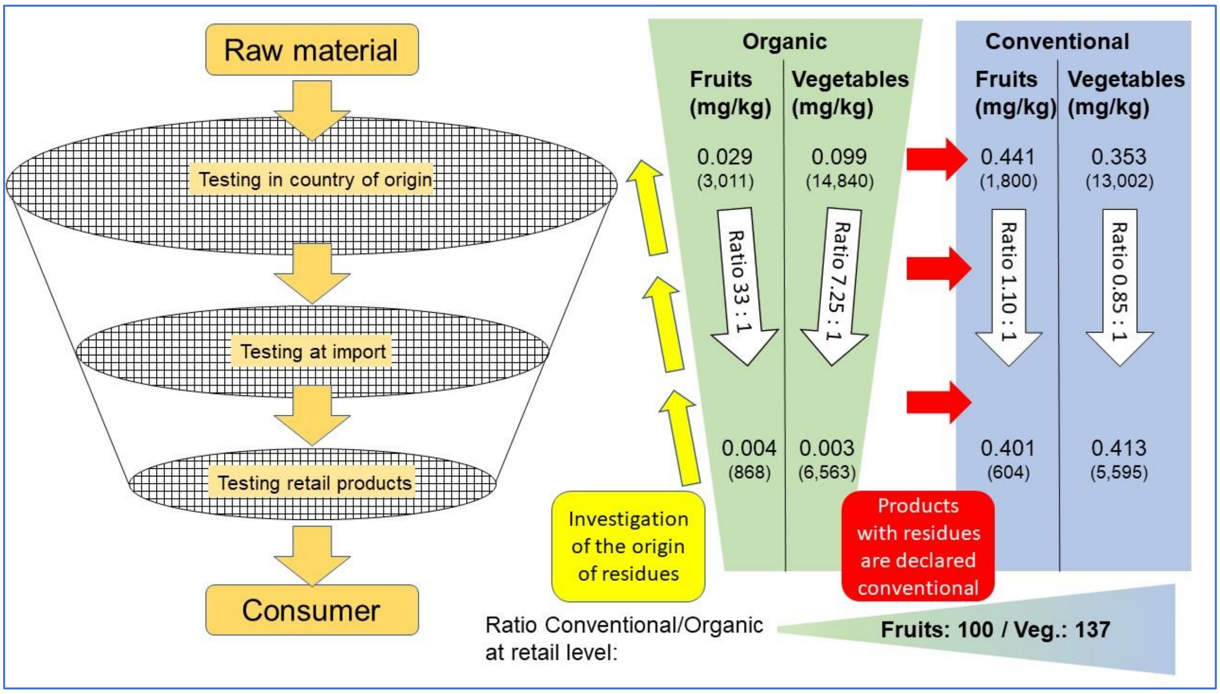


Figure 1: Multi-layer sieving model for residue testing at different points of the organic supply chain. The figures for organic and conventional fresh fruits and vegetables, respectively, represent the average total amount of pesticide residues found per sample. Above the white arrows: values found by the accredited commercial laboratory Eurofins Dr. Specht International, representing samples from products taken at different points of the supply chain. Below the white arrows: values found by the food authority CVUA,[3] representing retail products sampled from supermarkets, farmer markets and organic

food stores. Data in brackets represent the number of samples tested. Eurofins figures are from 2020 only, CVUA figures from 2013 through 2019. Ratios from wholesale to retail are shown in the white arrows. While the figures for conventional products remain in the same range in the process from wholesale to retail (blue rectangle to the right), the residues for organic products are very substantially reduced during this process (green trapezium). As a result, residues at retail level are 100 and more times lower in organic than in conventional products (trapezium at the bottom). This shows that the process represented by the red arrows works fairly well – which is not always the case for the investigation of the origin of such residues, as represented by the yellow arrows.

Unfortunately, this good news for consumers with respect to objective (b) does not always mean that objectives (a) and (c) are also met. With the steady growth of the organic market and globalisation of supply chains, fraud has also grown.[4,5] Pesticide residues in organic products can be the result of fraudulent spraying by farmers, commingling organic with conventional products, selling conventional products as organic, spray-drift, or different (avoidable or unavoidable) sources of contamination along the supply chain.

1.2 Two Forms of Spray-Drift

Over the past decades, a distinction has been made between short distance **primary** spray-drift **during** the application, and long distance **secondary** spray-drift occurring **after** the application.[6] The latter was attributed to evaporation and considered to play a role only for pesticides with high vapour pressure.[7] On the one hand, recent studies have shown that evaporation and long-distance transport can already play a role **during**, not only after application.[8] On the other hand, long-distance transport has been found to be linked not only to evaporation. Pesticides adherent to dust from wind erosion can contaminate large areas.[9] In the present context, we use the terms **short-range** and **long-range** drift, instead of primary and secondary drift (Figure 2).

1.3 Long-range Drift

Long-range drift is so far poorly understood, can lead to (normally very low) residues at distances as far as thousands of km,[10] and happens in the form of vapour or molecules adhered to dust. The main factors influencing long-range drift are vapour pressure of the pesticide, capacity of adherence to dust, incidence of wind erosion, and temperature inversion in the atmosphere.[7] Long-range pesticide drift has recently received more attention.[11,12,13,14,15] Examples have been used in the context of organic certification for supporting the argument of ubiquity of pesticides, linked to the assumption that low- or even medium-level residues in

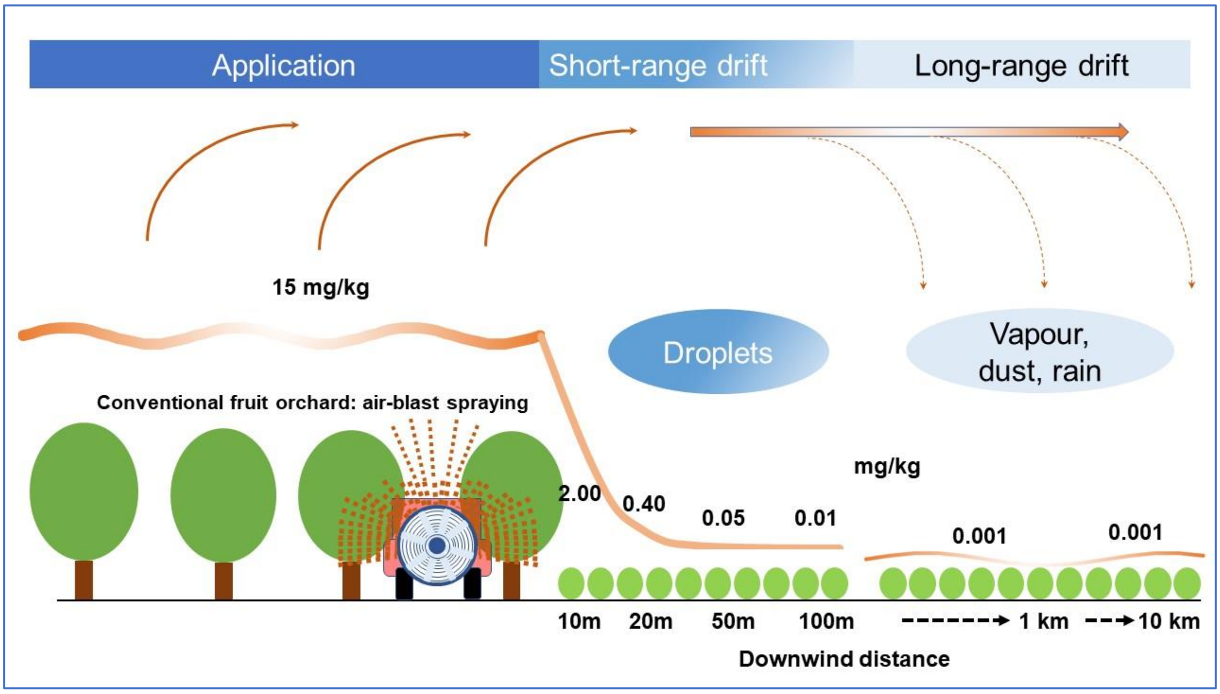


Figure 2: Simplified model of short-range vs. long-range drift originating from air-blast spraying in a fruit orchards. The specific values for pesticide concentrations (mg/kg) expected for different down-wind distances from the orchard can vary by a factor 10 or more, depending on the applied substance, dose, weather conditions, vegetation, etc., but the graph provides an approximate estimate of the ratios that can be expected. In the case presented here, pesticide concentration in fruit leaves immediately after the application is 15 mg/kg. In the area of short-range direct drift, deposit decreases exponentially, so that at 100 m distance, we can expect to find only 0.01 mg/kg. At further distances, deposits are often below this level.

organic products are often derived from their omnipresence in the environment.[16,17] Cases from Brazil (endosulfan in soybeans), Montana (USA) and Saskatchewan (Canada) (glyphosate in khorasan wheat) and Germany (pendimethalin and prosulfocarb in different crops) have been quoted to demonstrate the ubiquity of pesticides.[17] None of these case studies, however, provides solid evidence for the assumption that long-distance transport of pesticides leads to residues in organic food above the level of, say, 0.01 to 0.03 mg/kg. The problem of the herbicides pendimethalin and prosulfocarb being subject to long-distance drift because of their high vapour pressure, has been known for a long time,[18] but this phenomenon cannot be extrapolated to other substances. Even for these herbicides, there is no evidence that residues at larger distances could be above the indicated levels. On an average of 15 vegetation samples from nature reserves in Germany, 0.009 mg/kg pendimethalin and 0.004 mg/kg prosulfocarb were found.[19] Exceptions may exist, e.g., when pesticide applications are followed by heavy wind erosion, as seems to be the case in some of the North American wheat growing areas, where glyphosate is used for cereal desiccation shortly before harvest.

In a survey in Switzerland,[20] neonicotinoid residues were found in 93% of plant samples from organic farms (as compared to 100% of samples from conventional farms), thus supporting the ubiquity suspicion. But there were substantial quantitative differences between organic and conventional farms (Figure 3). The average sum of neonicotinoid residues in plant and soil samples from organic farms was lower by a factor of 11 than that of plant samples from conventional farms. For soil samples, this factor was as high as 71. Even the highest value for one single substance (imidacloprid) found in organic plants (2.13 µg/kg = 0.00213 mg/kg) would be below the limit of quantification (LOQ) used for this substance in most screenings (0.01 mg/kg).

In a study in Germany[19], total mean pesticide concentration in natural vegetation in five reference areas (average distance from arable fields > 3 km) was 0.003 mg/kg, and in 15 nature conservation areas (average distance from arable fields 143 m) it was 0.006 mg/kg, but in three buffer zones (average distance 54 m) it was 5.4 mg/kg. (To make figures comparable with other data in this article, we have deducted the concentration of non-agricultural pesticides from the total amounts, and divided the numbers by a factor five, because the residues in this study refer to dry matter, while all the others use fresh matter). Although 5.4 mg/kg at 54 m distance is a disturbingly high value, the survey confirms that concentrations at larger distances do not exceed the "traces" level. The intention of this article is not to put in doubt the environmental damage caused by such traces. We try to show that the "ubiquity" argument may sometimes be hiding cases of fraudulent pesticide use by organic farmers.

1.4 Short-range Drift

As opposed to long-range drift, short-range drift is well understood, has its impact mainly in a range from 1 m up to a maximum of 1,000 m (for aerial spraying), happens in the form of droplets, and is not substance specific. The main factors influencing this form of drift are droplet size, windspeed, and height of the boom (nozzles) above soil.[7,10,21,22,23] The fact that long-range drift is poorly understood and leads to low concentrations of certain substances over wide areas, should not stop certification bodies (CBs) from using the available

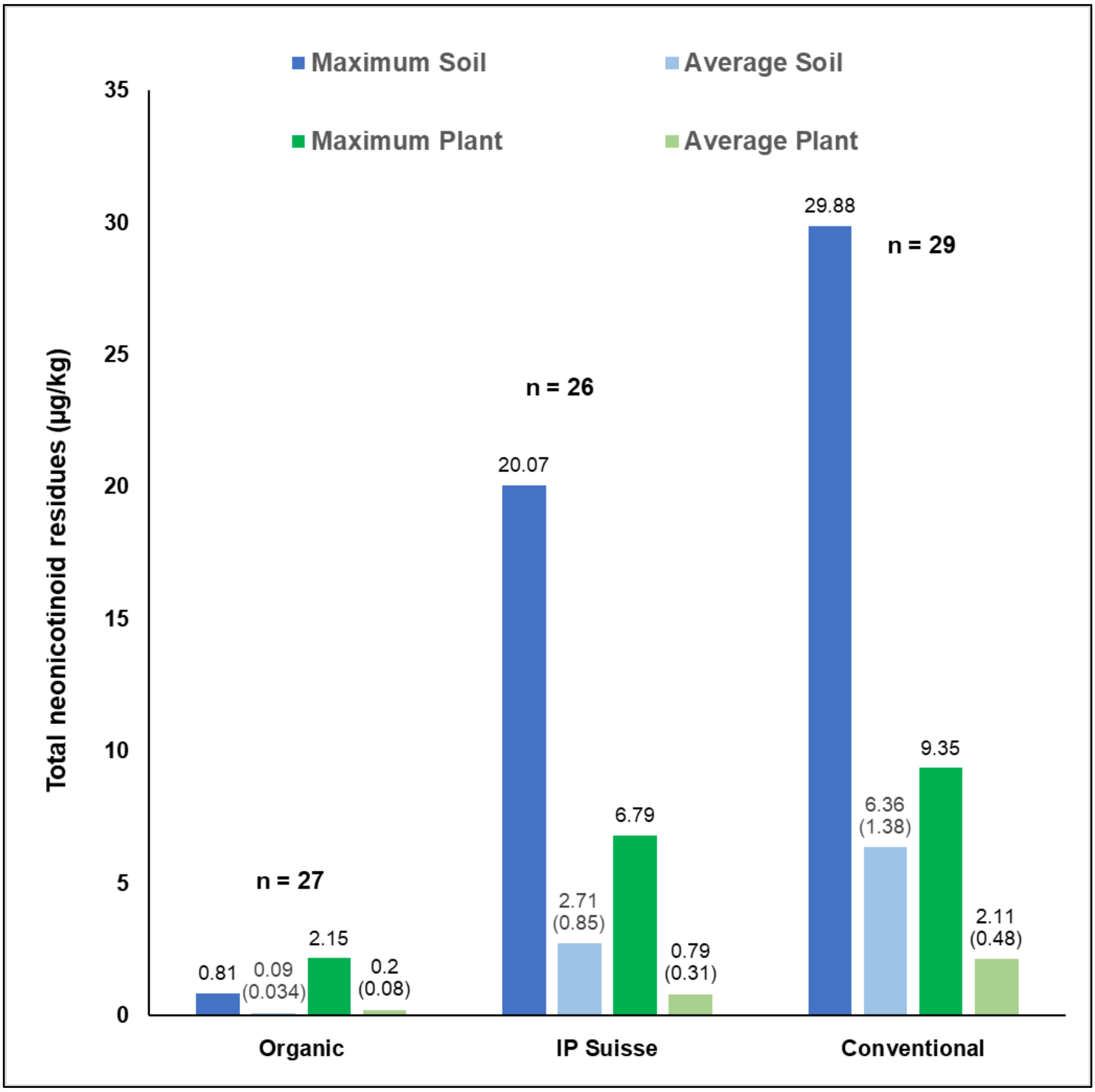


Figure 3: Maximum and average residues of neonicotinoid insecticides in soil and plant samples from organic, integrated ("IP Suisse") and conventional farms in Switzerland. The figures represent the sum of acetamiprid, chlothianidin, imidacloprid, thiacloprid and thiamethoxan. Figures in brackets represent standard errors.[18]

knowledge about short-range drift as a tool for assessing farmers' compliance with organic production rules. The dynamics of short-range spray-drift have been widely studied in the context of preventing liability problems due to herbicide damage, contamination of water bodies and natural habitats, and direct risks for human settlements.[10,21,22,23,25,26,27] Pesticide deposit decreases exponentially with increasing distance from the field, on which the substance is applied. With a tractor boom sprayer, deposit at 25 m distance is expected to be only 1% of that in the target field. While distances are greater for air-blast or aerial spraying, the basic principle of exponential decrease is the same (Figure 2 and Supplementary Fig. 1).

1.5 Certifiers' Testing Strategies

Both the EU Regulation on organic farming and the US National Organic Program (NOP) require certification bodies (CBs) to take samples from at least 5% of their clients every year. A large amount of data is being generated through this mechanism, but the sampling procedures and interpretation of results often do not allow to derive clear results.

Testing at different points along the organic supply chain could be an excellent tool for detecting non-compliance with organic production rules. The idea behind this is depicted in Figure 1. The filter process as such, and the exclusion of contaminated batches from the organic market, as represented by the red arrows, often work well. Thus, there are significantly lower average amounts of residues after undergoing this filtering process across the supply chain. The fact that residues in organic produce reported from the wholesale and processing levels are massively higher than those reported from retail samples (33 times higher for vegetables and 7 times higher for fruits) while they vary little for conventional produce, shows that market actors often remove problematic produce by declaring it conventional. However, the information about these switches is not always reaching the CBs, thus impeding the investigation of the origin of residues and the exclusion of excluding fraudulent actors from the market (yellow arrows in Figure 1).

Not only for market actors, however, but also for many CBs, the purpose of sampling and testing is limited to ensuring that food sold on the market with an organic claim, is free of pesticide residues. A recent unpublished BSc thesis at the University of Kassel revealed that 80% of the samples by CBs in ten EU member countries are taken of final products, but only 20% from the field or during the production process.

The differentiation between active use and non-intentional contamination is difficult, though, if only final products are tested. Plant (mainly leaf) samples from the field have several advantages in this regard: (a) Often, there is a long time span between pesticide application and harvest. Because of dissipation of the residues, nothing or only traces may be found in the final product (Supplementary Table 1). Field samples can be taken during or shortly after a suspected pesticide application, which leads to better results. (b) Leaves have a surface/weight ratio between 10 and 118 cm^2/g[28], whereas for fruits this ratio is between 0.6 and 2.2[29], and for seeds between 2 and 10 cm^2/g only[30,31,32]. Residues in leaves are therefore normally higher than in seeds, fruits or roots, which makes interpretation of test results easier. (c) Field sampling allows to take separate samples from centre and margin of the field, as explained below in more detail.

Unfortunately, if CBs take field samples at all, they often take them **only** from field margins[33,34] ("let's see if there is a drift problem"). Positive results are then attributed to spray-drift, and farmers are required to establish buffers – without even considering the possibility of residues originating from an application by the organic farmer. Such procedures open the door for fraudulent use of pesticides by organic farmers.

Other CBs have established so-called "action levels", below which they consider the presence of residues in organic products to be the result of ubiquitous environmental contamination, with no need to investigate their origin.[33] While such "action levels" may be necessary for specific cases (see below concerning the banana industry), using this approach in a general way disregards not only the spatial distribution, but also temporal dynamics of pesticides in plant tissue. As opposed to soil, half-lives in plant tissue exposed to UV radiation and weather, are relatively short for most modern pesticides[35]. A residue level of 0.02 mg/kg, used by some CBs as "action level", is typically reached one to two months after the application of a pesticide, in some cases even after only five days (Supplementary Table 1).

1.6 Objectives

The objectives of our study are: (I) to demonstrate that appropriate field sampling methods can differentiate the effects of fraudulent pesticide application by the organic farmer, from the results of both short-range and long-range spray-drift, and (II) for the specific case of aerial fungicide spraying in the banana industry, identify appropriate variables, which allow us to interpret the test results correctly for the purpose of this differentiation.

2 Materials and Methods

2.1 Case Studies from Different Crops and Countries

To demonstrate the appropriateness of differentiated field sampling (objective I), in a first part of our study we selected from the CERES archive six cases (Table 1), and one case from the GfRS archive (N° 6 in Table 1).

Table 1: Overview of case studies

Country	Crop	Type of sample	Approx. distance border to centre samples	Specific conditions to be considered
1. Chile	Apples, Blueberries	Leaves	100 m	High level of spray-drift because of air-blast spraying on conventional neighbour farms
2. Togo	Soybeans	Dry plants	20 – 40 m	Small fields, low level of spray-drift because farmers use manual knapsack sprayers
3. Thailand	Rice	Straw	50 m	Small field, good buffers between organic and conventional fields, irrigation
4. Ecuador	Cocoa	Leaves, cocoa beans, weeds	100 to 300 m	2,4-D had been found by a Belgian importer in cocoa beans. Conventional banana farms with a high level of drift in the neighbour-hood. 2,4-D is selective for control of dicoty-ledonous weeds, mainly in cereals, but is fre-quently used in Latin America also for weed control in perennial crops such as cocoa.
5. Bulgaria	Oil-bearing roses	Leaves	200 to 600 m	The inspector had been made believe that a risk of spray-drift did not exist, because con-ventional neighbour fields were semi-aban-doned. Therefore, he had taken only one sample from the centre of the farm, com-posed of several sub-samples. Later, it turned out that one neighbour had sprayed his rose plantation, using air-blast equip-ment, four days before the organic field had been sampled.
6. Germany	Grapes	Leaves	10 – 15 m	Small fields, very heavy drift, samples taken during spraying season.
7. Moldova	Walnuts	Kernels, fresh nuts, leaves	100 to 400 m	This case is different from the others, be-cause the walnut trees are wild or aban-doned. Requirements for organic certification of wild collection include, among others, har-vest from areas, where no pesticides have been applied during three years, and which are not exposed to spray-drift from conven-tional farming.
8. Ecuador	Bananas	Leaves and fruit peels	Ca. 200 m	Very high level of spray-drift because of aer-ial fungicide spraying. In addition, terrestrial spraying of insecticides and herbicides is common (insecticides and herbicides were not considered for this case study, however).

For differentiating drift from an application by the organic farmer, it would be preferable to test samples taken at **several** distances from the potential source of spray-drift, to find out if a gradient similar to the theoretical exponential decrease exists. In most cases, however, this would be too costly. Therefore, only two samples are normally taken: one close to the possible source of spray-drift, and one at the centre of the organic field. Exceptions are those cases, where an organic field is surrounded by several conventional fields.

Detailed sampling records are kept. Samples are stored in PE bags. When there is a risk of samples rotting during storage and transport, paper bags are used. Bags are sealed and sent to accredited laboratories for multi-residue screening following DIN EN 15662:2018-07 mod. LC-MS/MS, GC-MS/MS, GC-NCI-MS.

2.2 Variables that allow Interpretation of Test Results on Aerial Spray-Drift in the Banana Industry in Ecuador

Frequent aerial spraying in this industry leads to strong spray-drift, often with overlaps from several conventional neighbours on different sides of the organic farm. In addition, the high frequency of 17 to 35 applications per year[36] often leads to temporal overlaps of residues derived from several spraying events. To identify appropriate variables that allow under these conditions to discriminate between fraudulent application and spray-drift (objective II), a total of 476 residue tests from Ecuador from 2018 and 2019 were analysed. From these, 24 were excluded because of sampling mistakes. In most cases, to reduce testing costs, first a mixed sample from border and centre is tested (Supplementary Fig. 3). Based on the assumption that, due to the overall heavy drift problem, residues in mixed samples below 0.1 mg/kg were derived from drift and did not require separate testing, 119 mixed samples were identified as "drift" and no separate testing of margin and centre done. These were excluded from the statistical analyse. Residues in 20 mixed samples were so high that they were immediately identified as "application" and were also not considered. This left datasets from 67 farms with 222 individual samples (i.e. 67 centre and 155 border samples), which were analysed separately and then subjected to statistical analyses. Of the 67 cases, 14 had been identified as "application", 48 as "drift", while five had remained "unclear".

Thirty-nine variables (Supplementary Table 2) were tested for their suitability for telling apart spray-drift from deliberate fungicide use by organic farmers. When the laboratory had found only "Traces < LOQ" for a specific substance, a default value of 0.005 mg/kg was used instead. For testing the variables, multivariate statistical analysis based on logistic regression, discriminant analysis and support vector machines were performed to find rules that would classify farms into the two groups.[37] Here, we only report results of the discriminant analysis, which provided the most satisfactory results. The variable selection and performance of models and their prediction accuracy were assessed using leave-one-out and *k*-fold cross validation. The selected variables are graphically displayed using a biplot.[38] A one-way ANOVA was used to test the statistical significance of these variables between the "application" and the "drift" farms. The analysis was performed in *R* programming language using MASS, caret and klaR libraries.

3 Results and Discussion

3.1 Case Studies from Different Countries

3.1.1 Apple and Blueberry Orchard in Chile

The organic apple orchard from Chile has borders with a conventional cherry plantation. While in the sample taken close to the cherries, 11 different pesticides were found, with different residues adding up to 13.41 mg/kg (the highest value for a single substance was 9.1 mg/kg for captan), at 100 m distance only seven substances were found, with a concentration of only 3% of the border sample – leaving no doubt that the residues were derived from drift. Yet, the values were so extremely high that the orchard lost its organic status under NOP, while under the Chilean organic standard, the farmer had to establish broad buffer zones. In a nearby blueberry plantation, however, we had the opposite picture: the concentration of imidacloprid in the margin sample was 0.15 mg/kg, while in the centre of the field, at 100 m from the margin, it was 1.8 mg/kg. This was a clear case of fraud, and the farm lost its certification.

3.1.2 Soy-Beans from Togo

In the centre sample from farmer 1, 0.023 mg/kg chlorpyrifos was detected, and traces of dichlobenil, but no lambda-cyhalothrin. In the margin sample from SH1, however, no chlorpyrifos was found, but 0.076 mg/kg lambda-cyhalothrin and 0.005 mg/kg dichlobenil. Thus, most probably there was an overlap of an application (chlorpyrifos) and drift (for the other two substances). In the case of farmer 2, only traces of deltamethrin were found in the margin sample, but no residues in the centre, thus this was a clear "drift" situation. From farmer 3, only one sample was taken, because there was no conventional neighbour. The sample had relatively high residues of fipronil, clearly showing an application (Supplementary Table 3). These results demonstrate that even for small fields of less than 1 ha, the difference between residues derived from spray-drift and from application by the organic farmer can often be established, especially when neighbours use manual knapsack sprayers. As a result, the group's internal control system excluded several member farmers from the group and had to improve its internal member monitoring.

3.1.3 Rice Field in Thailand

The insecticides bifenthrin and chlorpyrifos were detected at levels of 0.005 to 0.013 mg/kg in samples from centre and margin, respectively, of a 4 ha rice field (Supplementary Fig. 2). The on-site inspection did not reveal any evidence for use of these substances by the organic farmer. Short-range drift could be ruled out, because in this case the residues in the centre would be expected to be by a factor 10 lower than in the margin sample. Both insecticides have a low vapour pressure, therefore long-range drift through evaporation is also excluded. The residues could theoretically originate from an application two to three months prior to sampling, but also from long-range drift through dust, or contaminated irrigation water. Many conventional rice farmers in the region use these insecticides. Under the principle of "innocent until proven guilty", the farmer remains certified.

3.1.4 Cocoa Plantation in Ecuador

For finding the origin of 2,4-D residues detected in organic cocoa beans, different samples were taken from the cocoa farm. Weed samples from the centre of the plantation had low levels (0.023 mg/kg) of the herbicide 2,4-D, while weed samples from the field margins and cocoa leaf samples were free of residues. Several fungicides found in the cocoa leaves probably came from aerial spraying on nearby banana plantations, but this could not have been the case for 2,4-D, because aerial spraying of this herbicide would kill the banana plants, while spray-drift from manual knapsack sprayers used in between banana plants, with the nozzle turned downwards, is almost zero. Also, long-range drift could be ruled out, because considering the dense canopy of cocoa trees, this would lead to higher residues in the canopy itself than in the weeds growing beneath (Supplementary Table 4). Dry weeds observed by the inspectors in between the cocoa trees provided further evidence of herbicide application on the organic plantation. Therefore, the certificate was suspended in spite of the low residue level.

3.1.5 Oil-bearing Roses from Bulgaria

A leaf sample from an organic oil-bearing rose (*Rosa damascena*) field in Bulgaria in 2020 had penconazole residues of 0.62 mg/kg. The farmer's claim of the neighbour spraying at a wind speed of 11 to 13 m/s seemed unlikely (not only because spraying under such conditions is not effective, but also because data from regional weather stations show a maximum wind speed of 3.8 m/s for the entire month). Using a spray-drift equation for air-blast spraying[27]:

$$y = 3908x^{-2.42} \tag{1}$$

with: x = distance from the target field and y = deposit at distance x, expressed as a fraction of the deposit on the target field,

combined with approximate data concerning the impact of wind speed,[10] CERES concluded that the assumption of these residues being derived from drift, was not plausible (Supplementary Table 5). Penconazole was also detected in a sample of rinse water from the organic farm's sprayer, further supporting the presumption that it was a case of deliberate application. The farm lost its organic status.

3.1.6 Vineyards in Germany

Grape leaves were sampled from eight organic vineyards during a period when conventional farmers were applying fungicides for preventing different fungus diseases. Samples from seven farmers had residues with a maximum of 0.75 mg/kg for folpet and 0.52 mg/kg for substances from the dithiocarbamate group. The small size of the vineyards, combined with air-blast spraying by neighbours and possibly air swirling caused by thermal lift in the hilly landscape, did not allow for a clear distinction between margin and centre samples. On farm N°8, however, the folpet concentration reached 73 mg/kg, clearly indicating a direct application by the organic farmer. This was confirmed later by a sample taken from sprayer rinsing water. This farmer lost the organic status, while the others remained certified. This decision was correct assuming that under the given weather conditions, all farmers in the region had sprayed more or less at the same time, so that drift effects were not confounded with dissipation effects.

3.1.7 Walnuts from Moldova

Between 2017 and 2019, eight out of eight walnut samples from a company in Moldova dedicated to wild collection had low residues of the herbicide 2,4-D (0.013 to 0.031 mg/kg; average 0.016 mg/kg). Four hypotheses were considered regarding the origin of this phenomenon: (a) Ubiquity due to long-range transport: 2,4-D is known to be taken up by plant roots and transported via the xylem.[39] Because of its lipophilic condition[40] it is often found in walnuts. This, together with consistently low residues in all samples from three harvest seasons, at a first glance made ubiquity in the region the most plausible explanation. (b) For facilitating harvest, collectors might remove vegetation below the walnut trees with the help of 2,4-D: This could be ruled out, because it would have been easily visible during on-site visits. (c) Short-range spray-drift from nearby cereal fields: To verify this hypothesis, leaf samples were taken from margins and centres of the collection areas. Indeed, the two margin samples had traces of 2,4-D < LOQ, while the six centre samples were free of residues. However, the finding did not seem to be a plausible explanation for the presence of 2,4-D in **all** walnut samples from three seasons. (d) Collectors might be delivering nuts from non-certified areas: This was confirmed through collector interviews. Due to the pressure from the CB, the company implemented strict measures for preventing delivery of nuts from non-certified areas. As a result, nine out of nine nut samples from the 2020 harvest were free of residues, thus refuting the ubiquity hypothesis and showing that most probably (d) was the main cause of the problem, possibly in combination with (c). After implementing the necessary measures, the company kept its organic status.

3.1.8 Two Examples from the Banana Industry

Sampling banana leaves is a time-consuming effort (Figure 4d). On the first plantation, the same six fungicides were found in the centre and in two border samples. Not only the sum of all pesticide residues was substantially higher in the centre than in the margins, but also the values for most individual substances (Figure 4a). This did not leave any doubt that the residues were derived from an application by the organic farmer, whose certificate was then suspended. On the second plantation, however, only the sample taken close to the conventional banana neighbour had residues, while the samples from the centre and close to a plantain orchard were free of residues. The residues were derived from drift and the farm kept its organic status (Figure 4b and Supplementary Fig. 4).

3.2 Statistical Analysis of Banana Sample Test Results from Ecuador

3.2.1 Results by CERES

For many cases, however, the decision between "drift" and application was not as clear as in Figure 4 a and b. As an example, one more difficult case is presented in Figure 4c. In total, residues of 25 different fungicides were detected, with a group of nine substances (difenoconazole, epoxiconazole, fenpropidin, fenpropimorph, propiconazole, pyrimethanil, spiroxamine, tebuconazole, triadimenol) each occurring in more than one third of the 222 samples from the 67 farms. The highest value for one single substance was 4.8 mg/kg; 21% of the centre samples, but only 1.3% of the border samples were free of residues (Supplementary Table 6).

CERES had decided that of the 67 farms included in the analysis, on 14 farms fraudulent pesticide applications had taken place, while 48 were classified as drift, and five had remained "unclear".

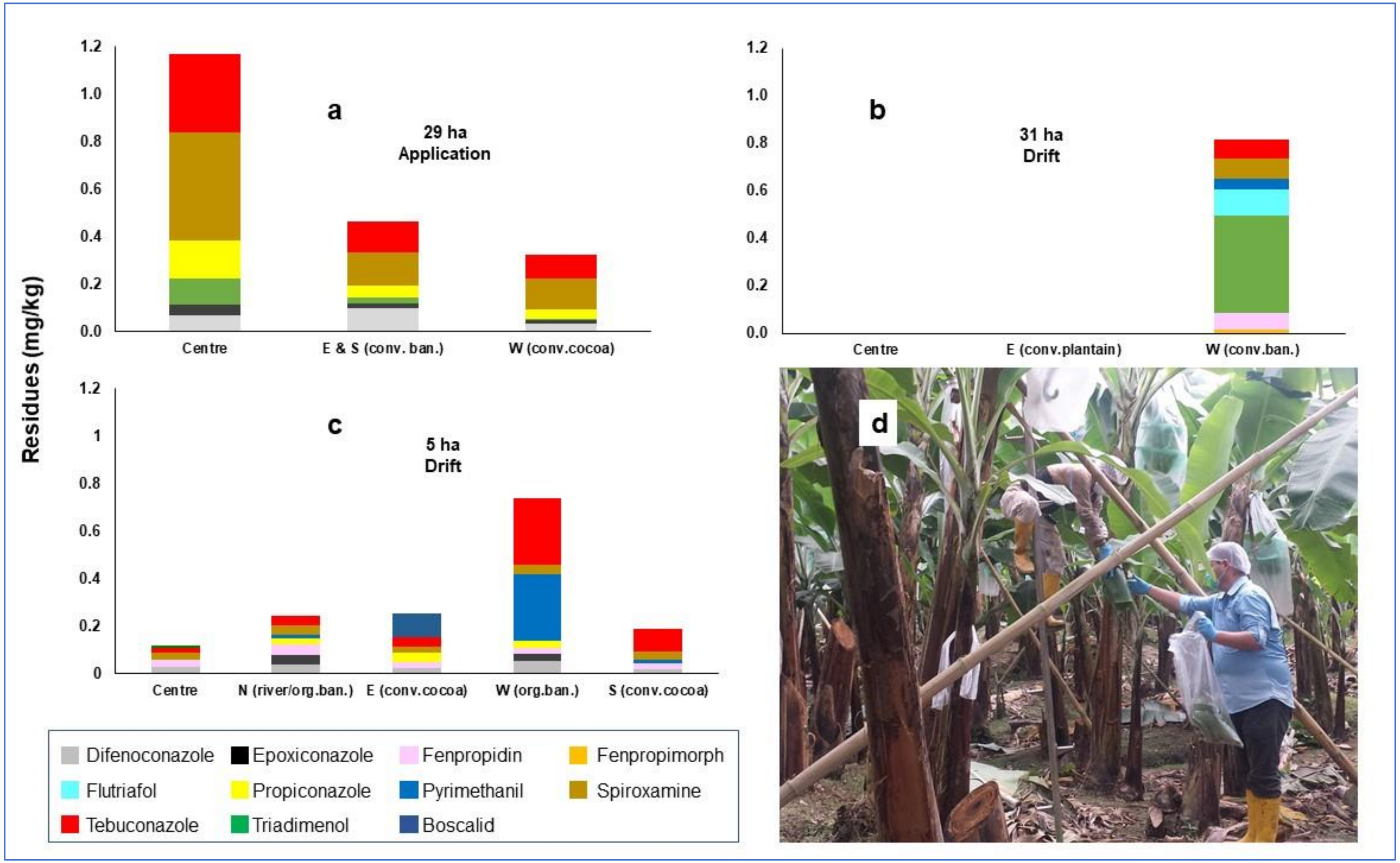
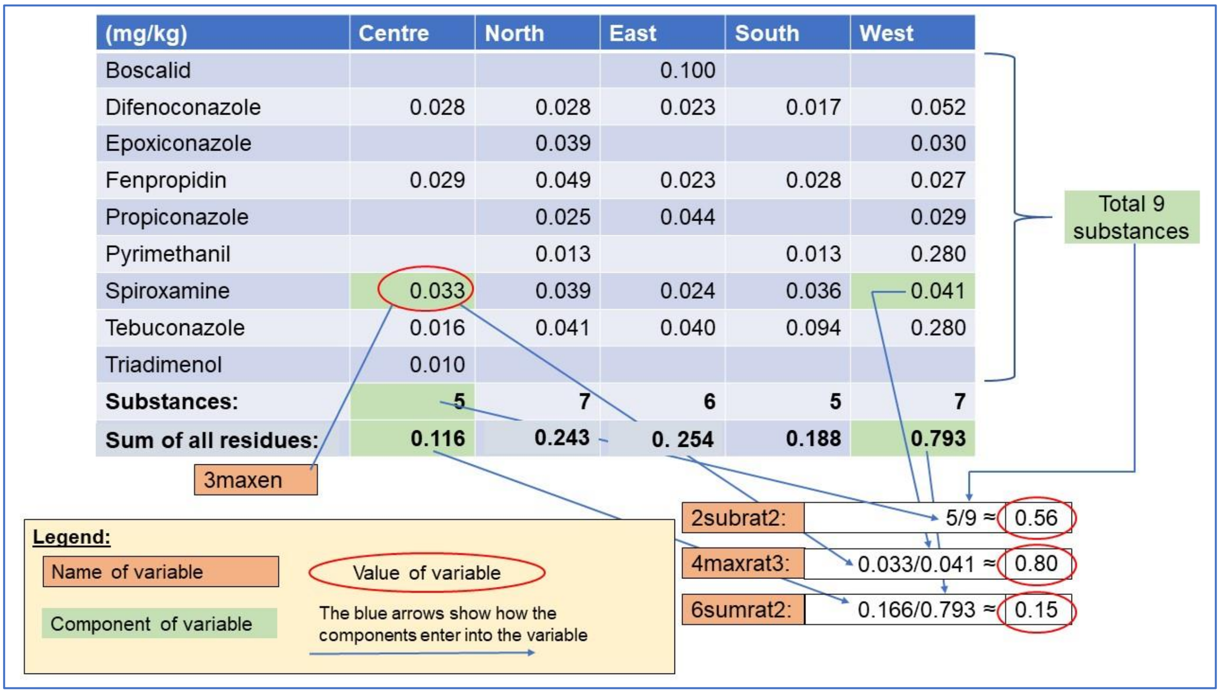


Figure 4: Residues of different fungicides found in leaf samples from three banana farms in Ecuador: (a) is a clear "application" case, (b) is a clear "drift" case. Also (c) is a "drift" case, but more complex because of the small size of the farm and the many different substances involved. Interestingly, the drift in (c) comes from the West (which is also the main direction of wind), where another organic banana farm (not certified by CERES) is located ("org.ban.", "conv.ban." etc. refer to organic banana, conventional banana, cocoa and plantain farms as neighbours on each side; N, E, S, W to the cardinal points). (a) and (b) are cases from 2017, and are therefore not included in the statistical evaluation, while (c) represents case N° 49 (see also Figure 5). (d) Sampling banana leaves.Photo by L.Guamán.

3.2.2 Statistical Approach

In a first approach, the discriminant analysis identified six variables as the most promising ones based on a cross-validated stepwise selection procedure (1subcen, 2subrat2, 3max-cen, 4maxrat3, 5sumcen and 6sumrat2, see Figure 5). The one-way ANOVA also indicated that the six selected variables are significantly different between the drift and the application group (Supplementary Table 7). The biplot based on the first two principal components using these selected variables explains 60 and 17% of the total variation, respectively (Figure 6b).



(mg/kg)	Centre	North	East	South	West
Boscalid			0.100		
Difenoconazole	0.028	0.028	0.023	0.017	0.052
Epoxiconazole		0.039			0.030
Fenpropidin	0.029	0.049	0.023	0.028	0.027
Propiconazole		0.025	0.044		0.029
Pyrimethanil		0.013		0.013	0.280
Spiroxamine	0.033	0.039	0.024	0.036	0.041
Tebuconazole	0.016	0.041	0.040	0.094	0.280
Triadimenol	0.010				
Substances:	5	7	6	5	7
Sum of all residues:	**0.116**	**0.243**	**0. 254**	**0.188**	**0.793**

Figure 5: Raw data for case 49 (see also Figure 4c), including an explanation of the four selected variables. A total of 5 fungicides were found in the centre, while in total 9 different fungicides were found in all samples, therefore the ratio of the two (called "2subrat2") is 597≈0.56. The value 0.033 mg/kg for spiroxamine is the highest figure out of the five residues found in the centre (here called "3maxcen"). We compare this to the highest value for spiroxamine among all samples, which is 0.041 mg/kg. The ratio of the two (called "4maxrat3") is 0.033/0.041≈0.80. The sum of all residues from the centre is 0.116 mg/kg, whereas the highest sum of residues from all samples is 0.793 mg/kg. The ratio of both, here called "6sumrat2", is 0.116/0.793≈0.15.

Farms previously considered as having been subject to drift mostly clustered around zero while application farms scattered on the left side of plot with two exceptions clustering around zero. The farms considered unclear are distributed throughout. The raw data for the six variables were visualized using a heatmap (Figure 6a). For this, each variable was standardized to a mean of zero and unit variance. The clustering of farms is visualized using a dendrogram based on the Unweighted Pair Group Method with Arithmetic means (UPGMA). The heatmap shows that “application” farms tend to be elevated in all six variables, even though there is non-negligible heterogeneity within groups, confirming the one-way ANOVA results. Application farms are clustered in the top rows, showing that two farms that had been considered subject to drift grouped clearly with the "applicants", whereas three supposed applicants grouped with the spray drift group. Four of the five unclear cases grouped in the application group or at the edge towards the drift group while one unclear case fell in the drift group. Interestingly, farm 61 that groups in Figure 6b with the applicants, falls into the drift group in Figure 6a. Taking a closer look at the initial raw data and sampling record for those farms, which visually in both heatmap and biplot appeared to be misclassified as "drift" (cases 42 and 48), revealed that these cases should not have been included in the analysis because the field samples had been taken in a wrong way, and that the cases 5, 8 and 14 should have been classified as “drift”. Thus, we subsequently re-classified the latter three cases as “drift” and the former two as “unclear”, leaving a training dataset containing the 60 farms for which the class had been assigned as either “drift” or “application”. The test dataset contains the five cases originally classified as “unclear” (cases 58, 59, 60, 61, 62) and the two cases subsequently removed from the test set (cases 42 & 48). The linear discriminant function performed best in terms of accuracy with the four variables 2subrat2, 3maxcen, 4maxrat3 and 6sumrat2 (Figure 5 and Figure 7).

This linear discriminant function was evaluated by cross validation and found to correctly classify a farm as either "drift" or "application" with an accuracy of 93.3%. The leave-one-out cross validation method was used to evaluate the accuracy of the model. In this method, each sample farm was dropped from the test data and then the class of that farm was predicted using the discriminant model. The misclassification rate in this cross validation of a “drift” as an “application” farm was 2.1%. This means that for a farm that is truly a “drift farm" there is an estimated probability of 2.1% that this is erroneously classified as an “application” farm. The misclassification rate of an “application” farm as a “drift” farm was estimated at 25% (Figure 7). Thus, for an “application farm” there is an estimated 1 in 4 chance of being

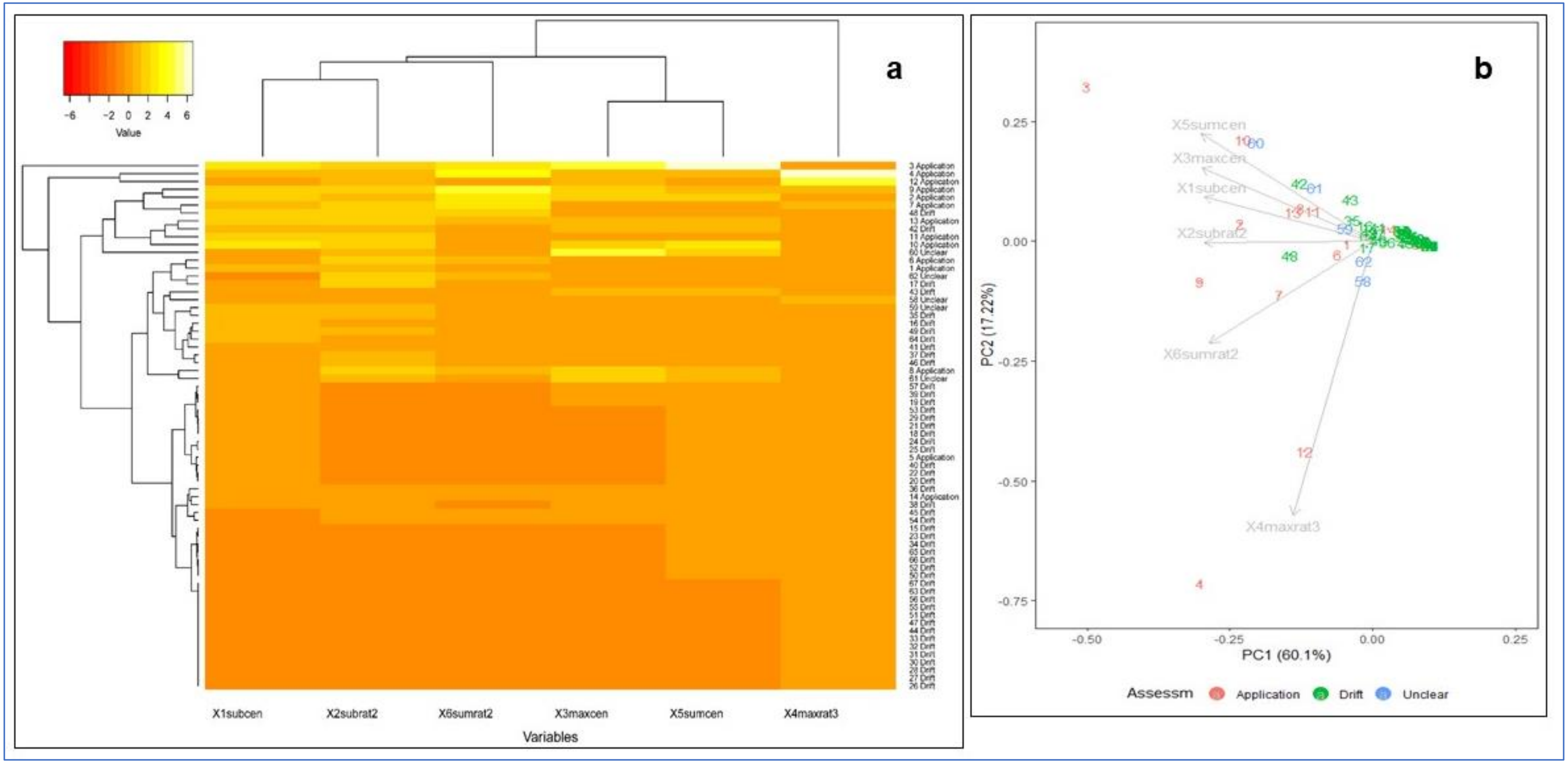


Figure 6: (a) Heatmap of six variables from 67 farms. The samples appear on the heatmap according to the original classification. The "application" farms are grouped at the top of plot, the "drift" farms below. The clustering of farms is visualized using a dendrogram based on the Unweighted Pair Group Method with Arithmetic means (UPGMA). (b) PCA biplot of 67 farm samples for six variables. The samples are coloured by the initial classification. The "drift" farms are clustered around (0, 0) while "application" sample farms are spread on left of the plot, and the "unclear" cases in between.

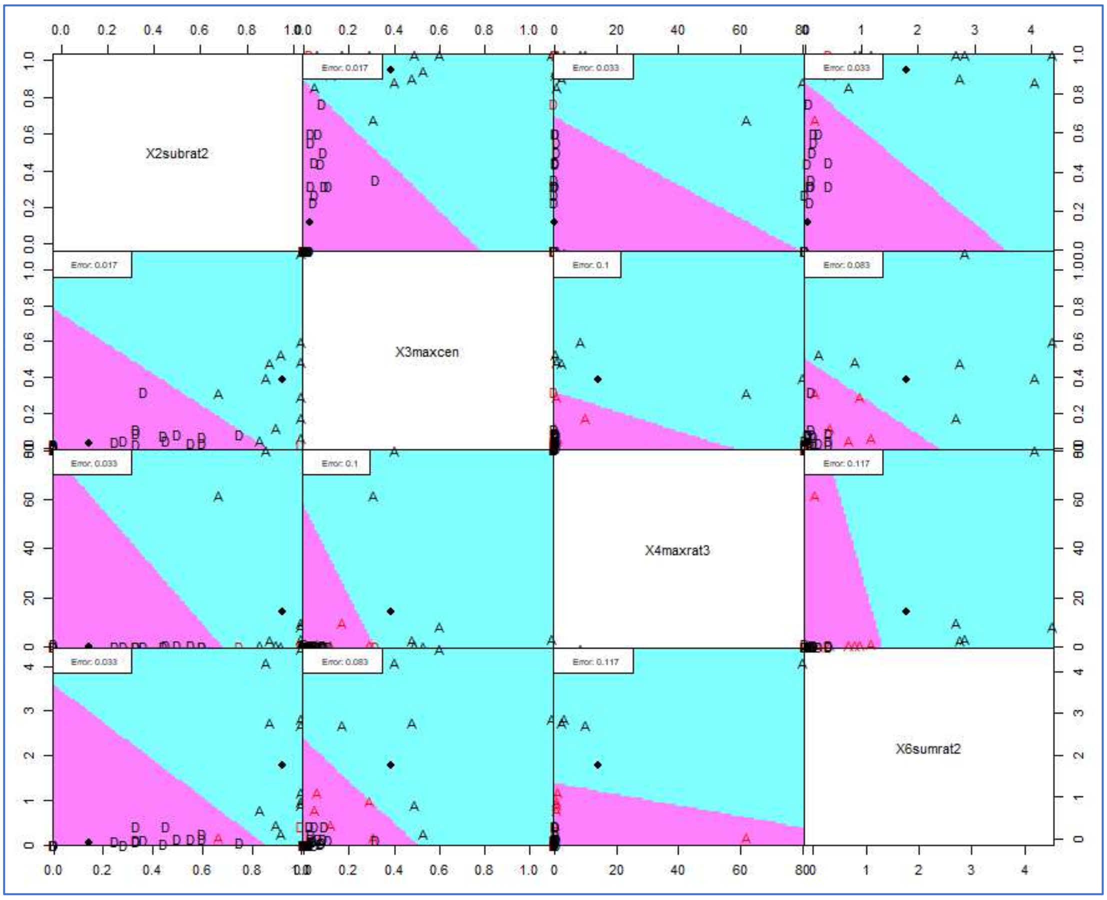


Figure 7: Linear discriminant function for the four selected variables. The D represents the true "drift" cases while A represents the true "application" cases. The two colours represent the decision rule: cases falling into the magenta region are classified as "application", cases falling into the turquoise region are classified as "drift" cases. Misclassified farms are plotted in red, correctly classified ones in black.

falsely classified as a "drift" farm. Of course, these estimated error rates are themselves subject to estimation error, and it is desirable to accumulate data from more farms to stabilize these estimates, as well as the estimates of the discriminant function. It also needs to be taken into account that, as we have explained here, there was some uncertainty regarding correct group membership for some farms that was only revealed by closer scrutiny of the initial statistical analysis. This may mean that the error rates we obtained in cross-validation of the final analysis presented here are on the optimistic side. The continuation of the present work, and especially the accumulation of data from more farms, will help to avoid such wrong assessments in the future. Three out of the five initially "unclear" cases turned out to belong to the "application" group, two to the "drift" group.

The linear discriminant function in our analysis is (See Figure 5 and Supplementary Table 2 for explanation of variables):

$$Application = -13.63421 + 16.17447(2subrat2) + 12.20420(3maxcen) + 0.11764(4maxrat3 + 1.5647(6sumrat2) \quad (2)$$

$$Drift = -0.42757 + 2.76201(2subrat2) + 0.91776(3maxcen) + 0.00942(4maxrat3) - 0.16929(6sumrat2) \quad (3)$$

$$If\ Application > Drift \rightarrow Application$$

$$If\ Application < Drift \rightarrow Drift$$

The linear discriminant function is also depicted in Figure 7 for the four selected variables. For each pair of variables, the plot shows the separation of the two groups by two different colours, and the placement of individual samples represents the rate of correct classification.

4 Conclusions

1. In most cases, comparing pesticide residues in leaf samples from field margins close to a possible source of spray-drift, to samples from the centre of the organic field, allows to distinguish the effects of spray-drift from deliberate pesticide use by the organic farm. The method works even in regions with extremely intensive pesticide use and aerial spraying by conventional neighbours.
2. The distinction is also possible when it comes to very small fields, where the distance between border and centre is short – provided that manual knapsack (as is normally the case in smallholder setups) or tractor boom sprayers are used. It becomes difficult to impossible on such small fields, when neighbours use air-blast or aerial spraying.
3. When residues below approximately 0.03 mg/kg are found evenly spread over the field, it becomes difficult to distinguish long-range drift (from evaporation or wind erosion) from the results of deliberate use several weeks before sampling. In such cases, the test results alone do not allow to prove fraudulent practices, as long as other evidence (pesticide containers in the farm house, residues in rinse water in the sprayer, records, etc.) do not exist.
4. Following strict sampling protocols and keeping detailed records are the key for using this

method in a meaningful way. Sampling must be planned in a way that allows for clear interpretation of results. Taking only one sample from a field, often leads to useless results. Sampling residues in spraying equipment, cross-checking with book-keeping records and other inspection methods, should be used as complementary methods.

5. A weak point of our survey of the 67 banana farms is that some of the centre samples were not taken at sufficient distance from the edges. This has meanwhile been corrected through improved work instructions (Supplementary Fig. 5). Another correction of the procedure, which is currently being tested, is reduction of the "action level" for separate testing of centre and border samples from 0.1 to 0.06 mg/kg. Once a substantial number of test results under this new protocol have been obtained, other variables from Supplementary Table 2 might perform better, e.g. N° 7 through 15.
6. When a reference sample from the field margin is not available, and residues are high in the central part of the organic farm, comparing the test results to expected values from standard deposition curves, can be enough to distinguish drift from application.
7. For the specific conditions of aerial fungicide spraying in the banana industry, the variables explained in Figure 5 and a linear discriminant function such as the one outlined above can be used tentatively for differentiating drift from application. We suspect that the same method can be used for other crops exposed to heavy drift pressure (e.g., fruit orchards and vineyards), but this is yet to be confirmed.

Authors' Contributions: A. Benzing wrote most of the article. H.-P. Piepho and W.A. Malik contributed the statistical part. M.R. Finckh conducted a thorough review and improved the article's consistency. M. Mittelhammer and D. Strempel selected some of the case studies and made the initial decision for the banana farms. J. Jaschik prepared the organic vs. conventional test results from the Eurofins database. J. Neuendorff contributed data from the GfRS archive. J. Mancheno, L. Melo, L. Guamán, R. Cangahuamín, J.C. Ullauri and O. Pavón were in charge of the sampling from the banana farms.

Additional Information

The authors declare no competing interests.

Acknowledgement: Thanks to the inspectors in different countries for thorough sampling and keeping sample records, to Kasey Moctezuma and Anton H. Scholtz for proof-reading and improving the English, and to Bernhard Schulz (CERES) for starting the gradient oriented sampling program for the banana farms many years ago.

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Legends

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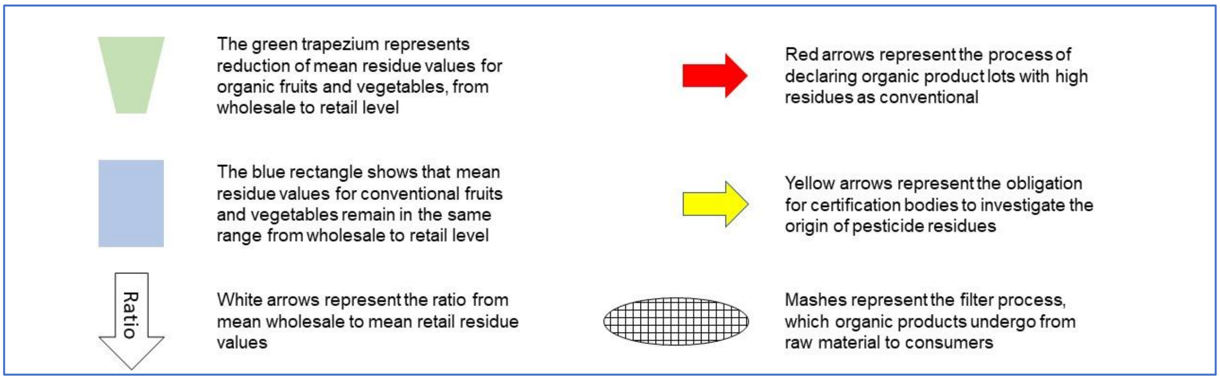

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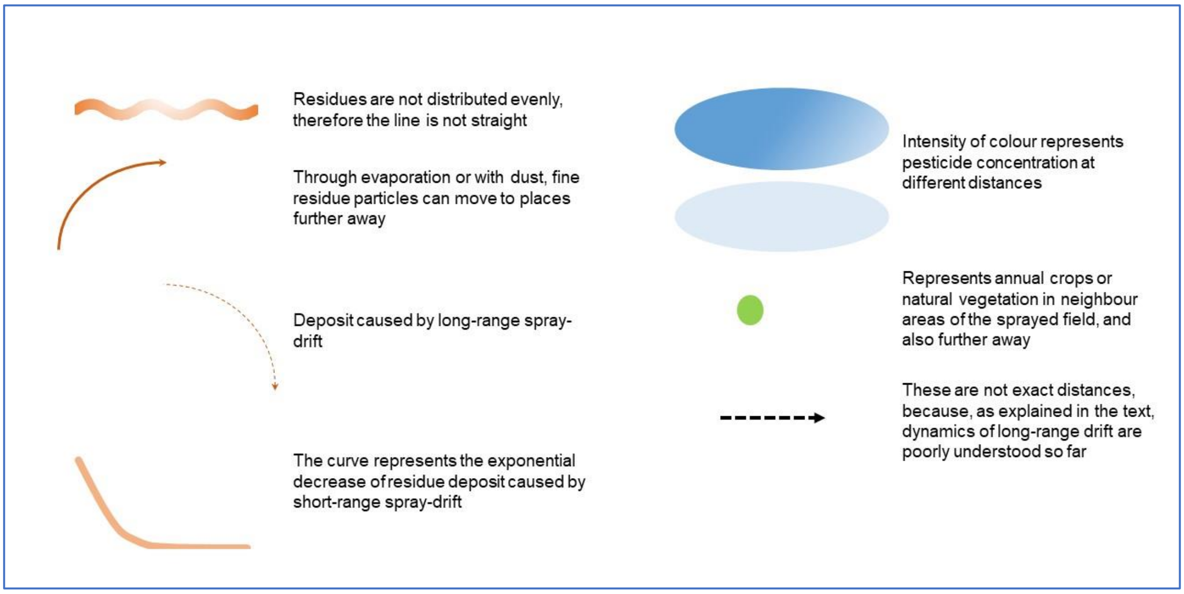

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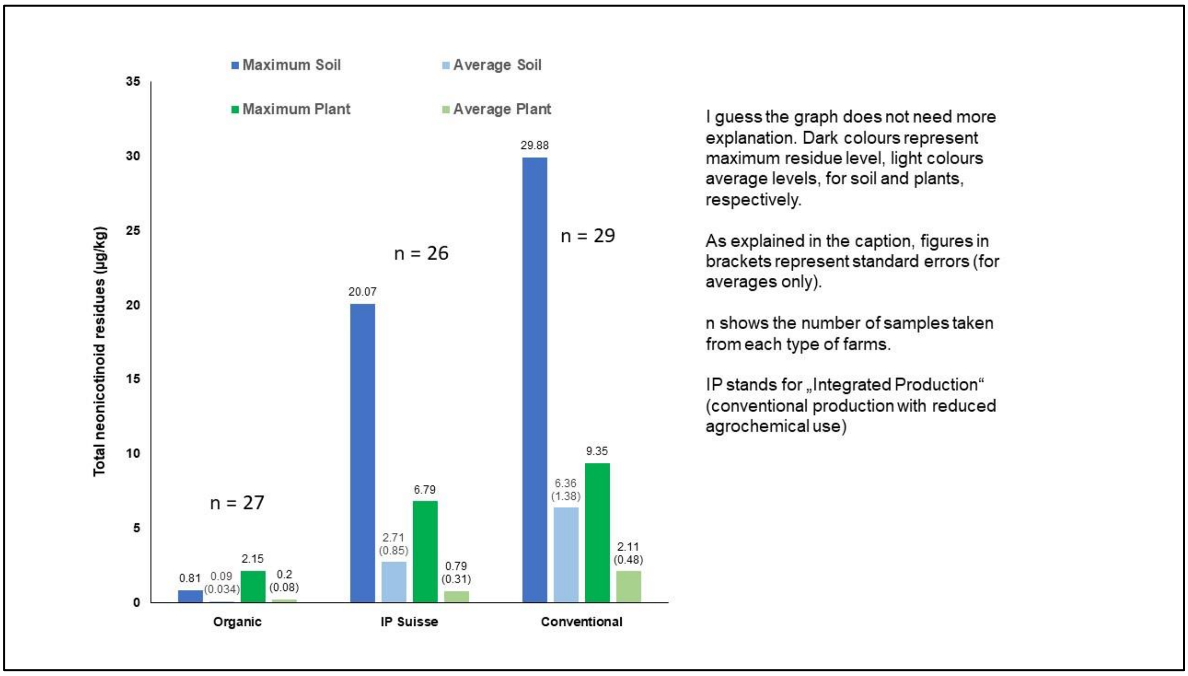

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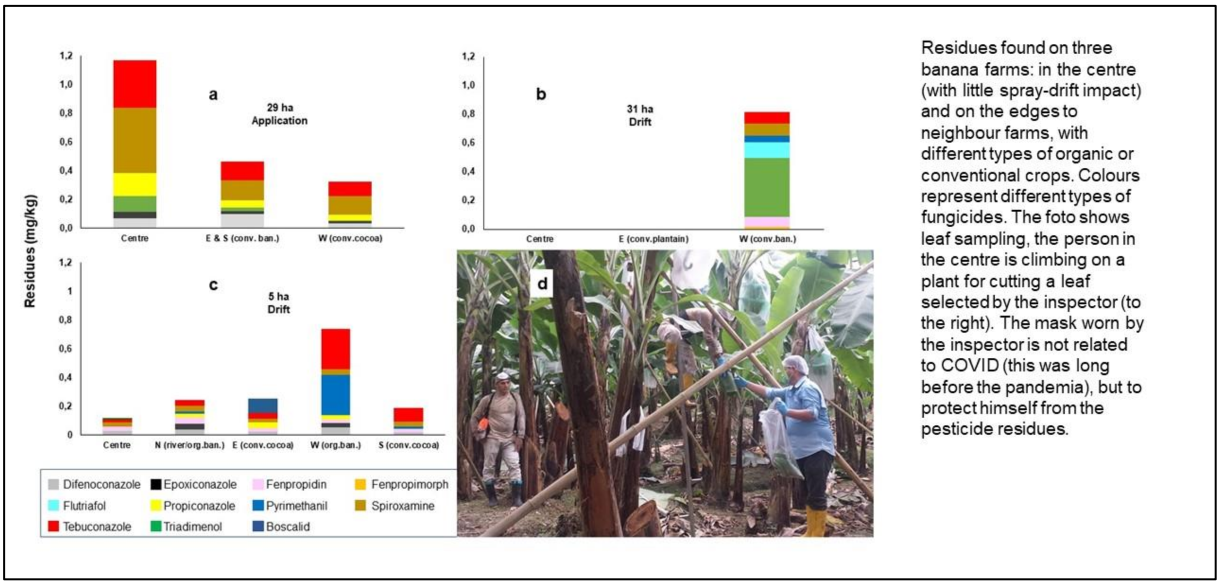

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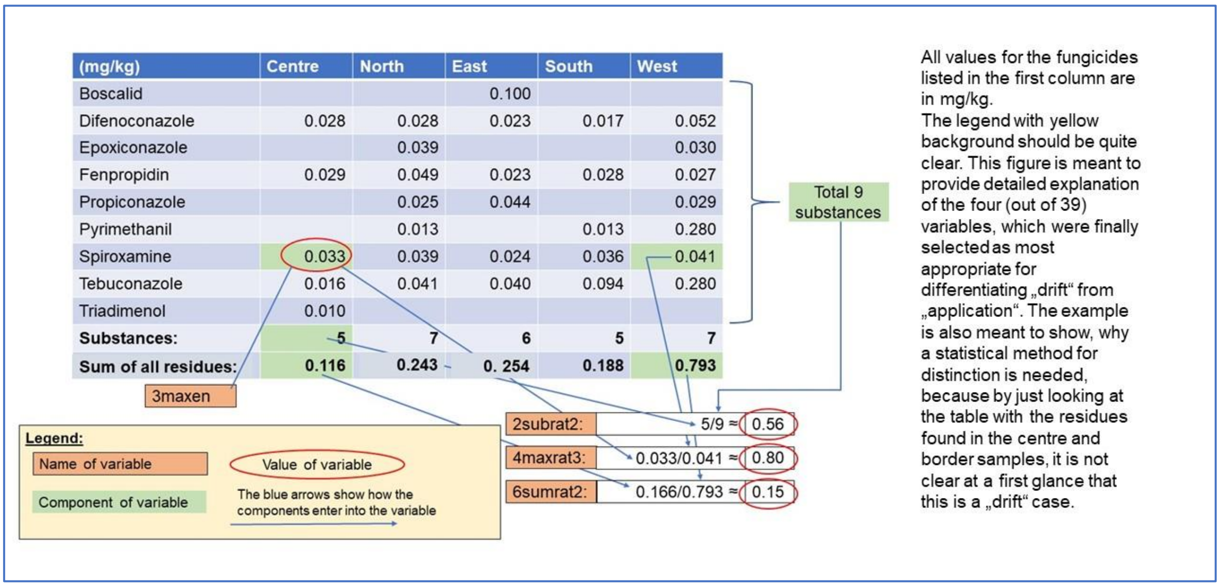

(mg/kg)	Centre	North	East	South	West
Boscalid			0.100		
Difenoconazole	0.028	0.028	0.023	0.017	0.052
Epoxiconazole		0.039			0.030
Fenpropidin	0.029	0.049	0.023	0.028	0.027
Propiconazole		0.025	0.044		0.029
Pyrimethanil		0.013		0.013	0.280
Spiroxamine	0.033	0.039	0.024	0.036	0.041
Tebuconazole	0.016	0.041	0.040	0.094	0.280
Triadimenol	0.010				
Substances:	**5**	**7**	**6**	**5**	**7**
Sum of all residues:	**0.116**	**0.243**	**0. 254**	**0.188**	**0.793**

Figure 6, 7 and 8:

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Figures

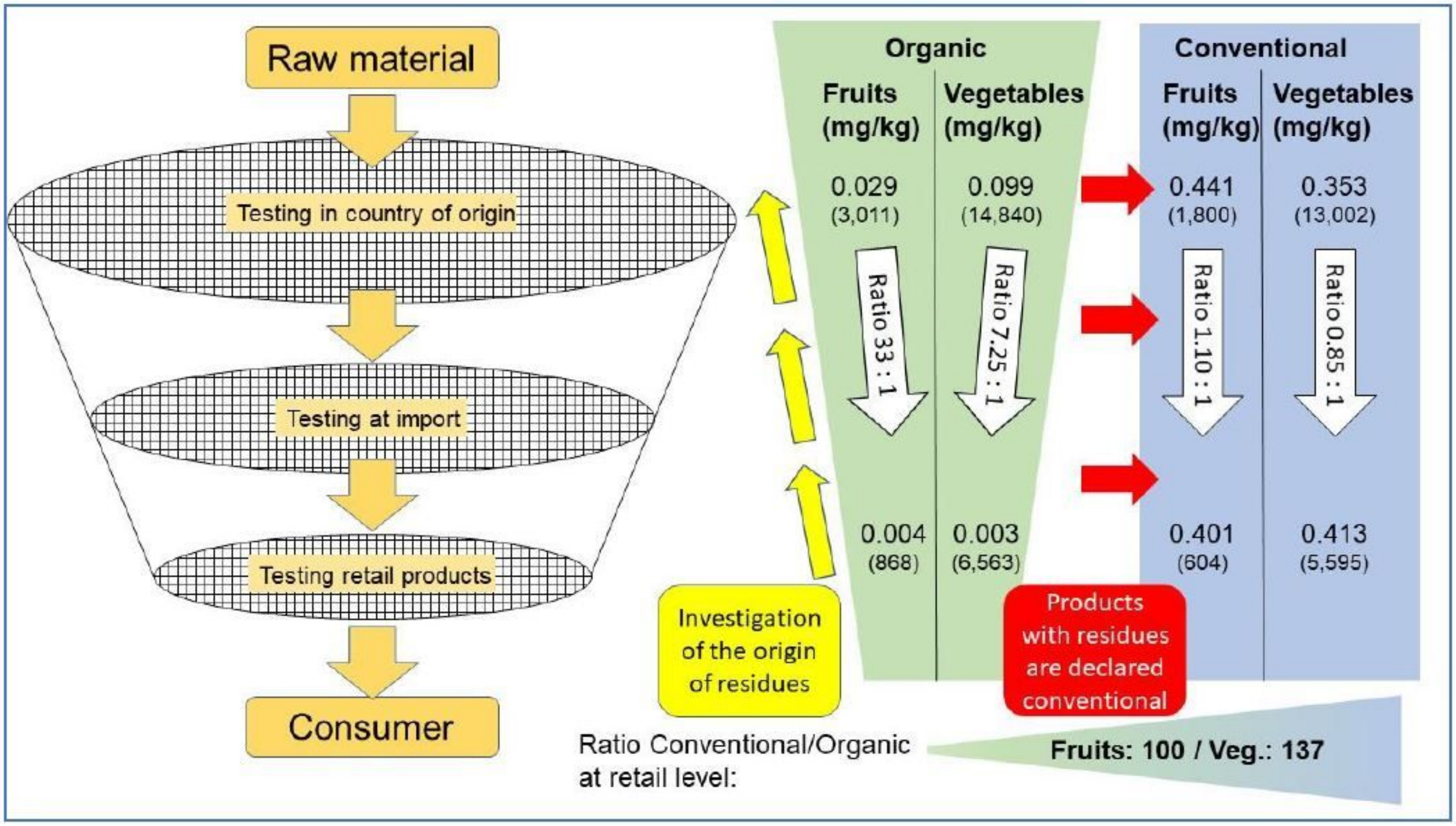

Figure 1

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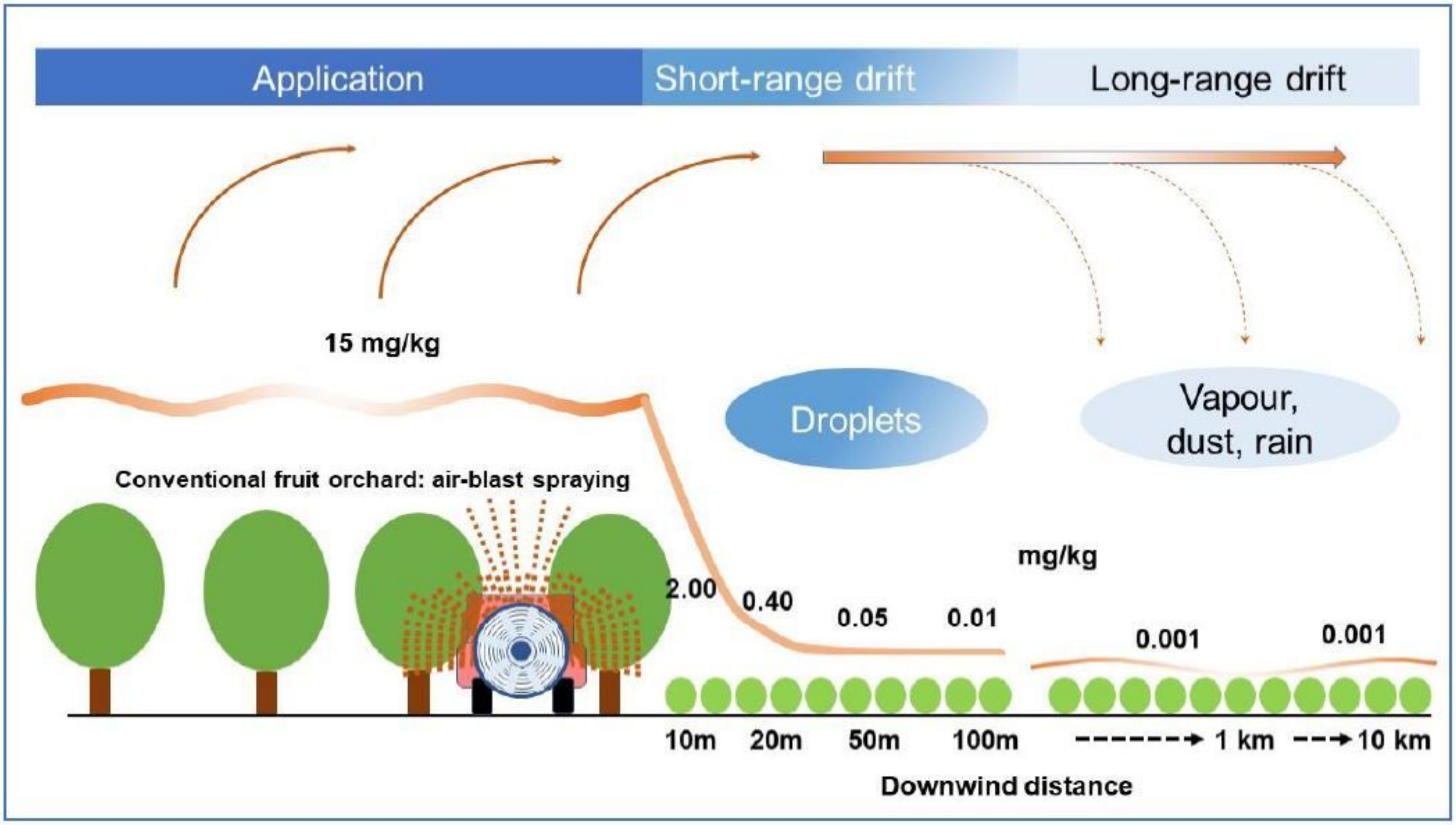

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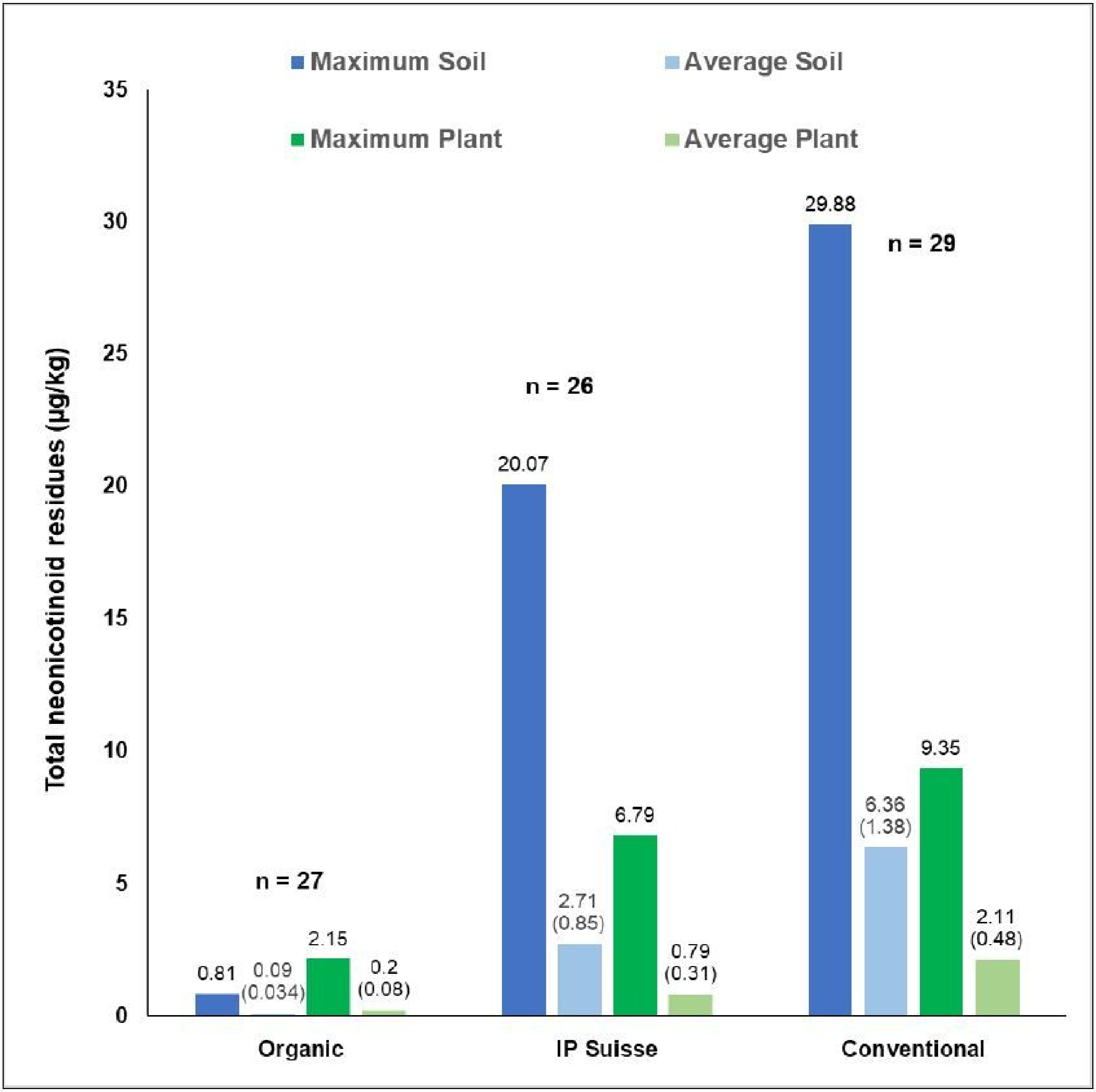

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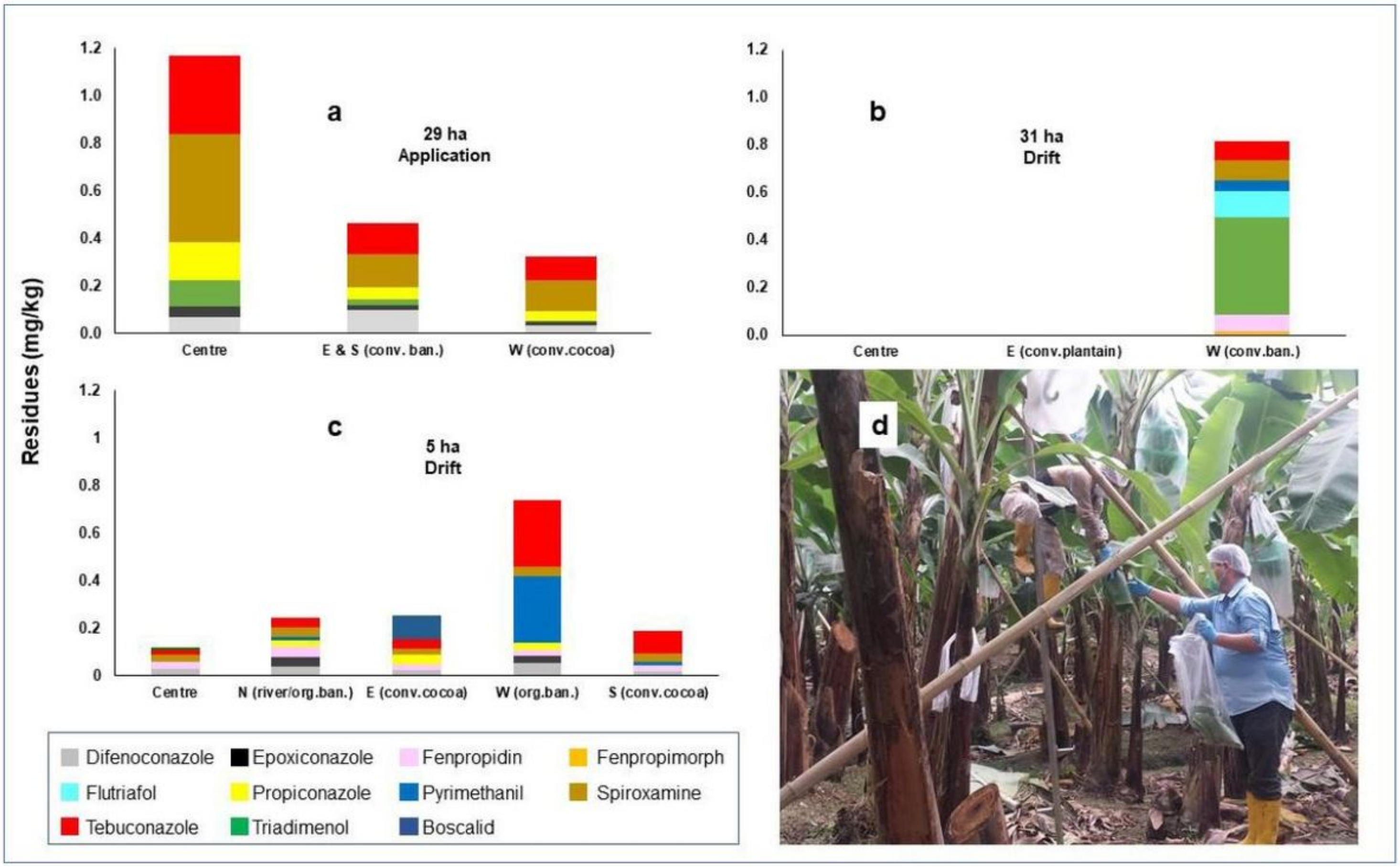
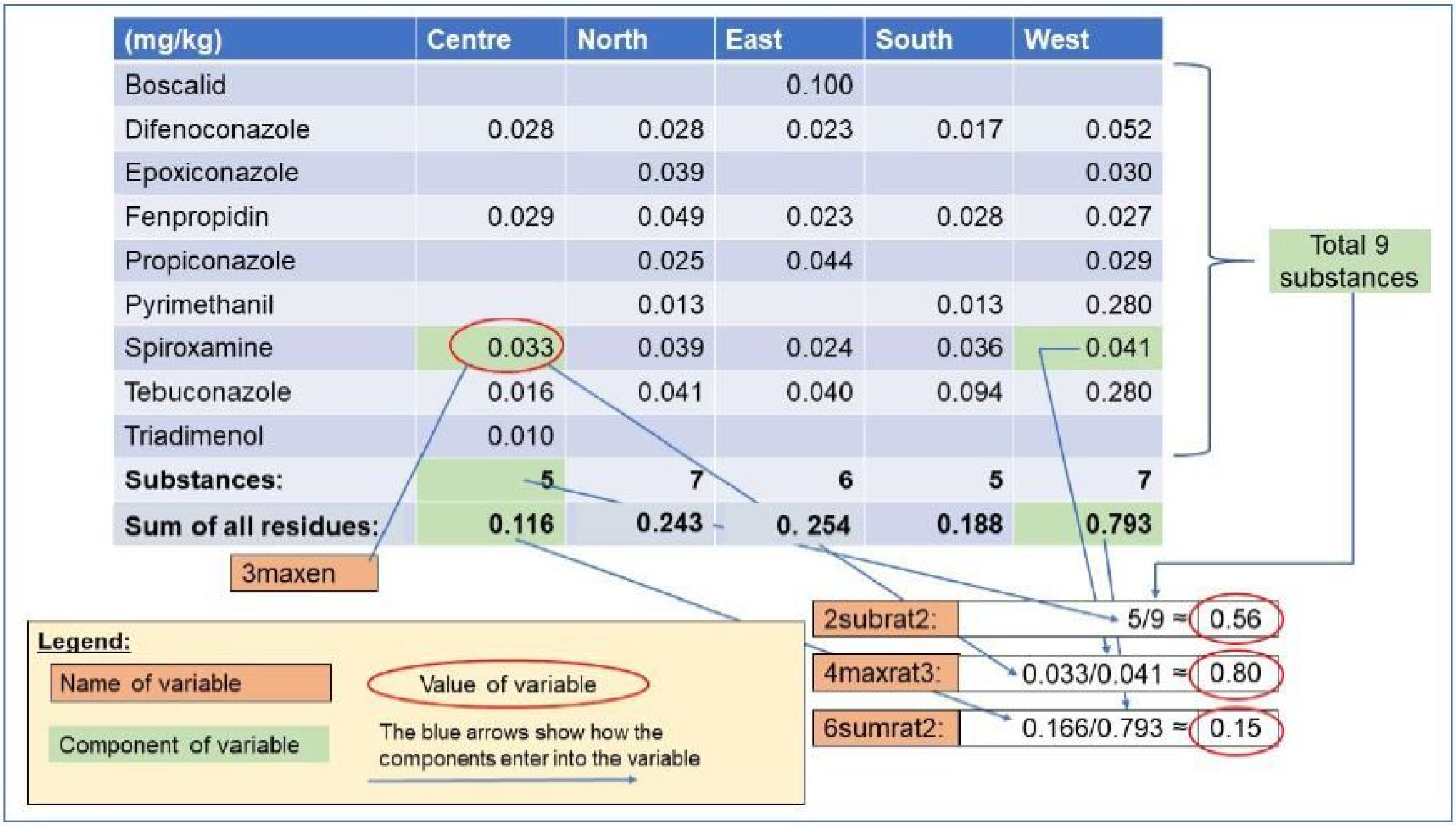


Figure 4

Due to technical limitations, the figure caption is only available in the manuscript file.



(mg/kg)	Centre	North	East	South	West
Boscalid			0.100		
Difenoconazole	0.028	0.028	0.023	0.017	0.052
Epoxiconazole		0.039			0.030
Fenpropidin	0.029	0.049	0.023	0.028	0.027
Propiconazole		0.025	0.044		0.029
Pyrimethanil		0.013		0.013	0.280
Spiroxamine	0.033	0.039	0.024	0.036	0.041
Tebuconazole	0.016	0.041	0.040	0.094	0.280
Triadimenol	0.010				
Substances:	**5**	**7**	**6**	**5**	**7**
Sum of all residues:	**0.116**	**0.243**	**0. 254**	**0.188**	**0.793**

Figure 5

Due to technical limitations, the figure caption is only available in the manuscript file.

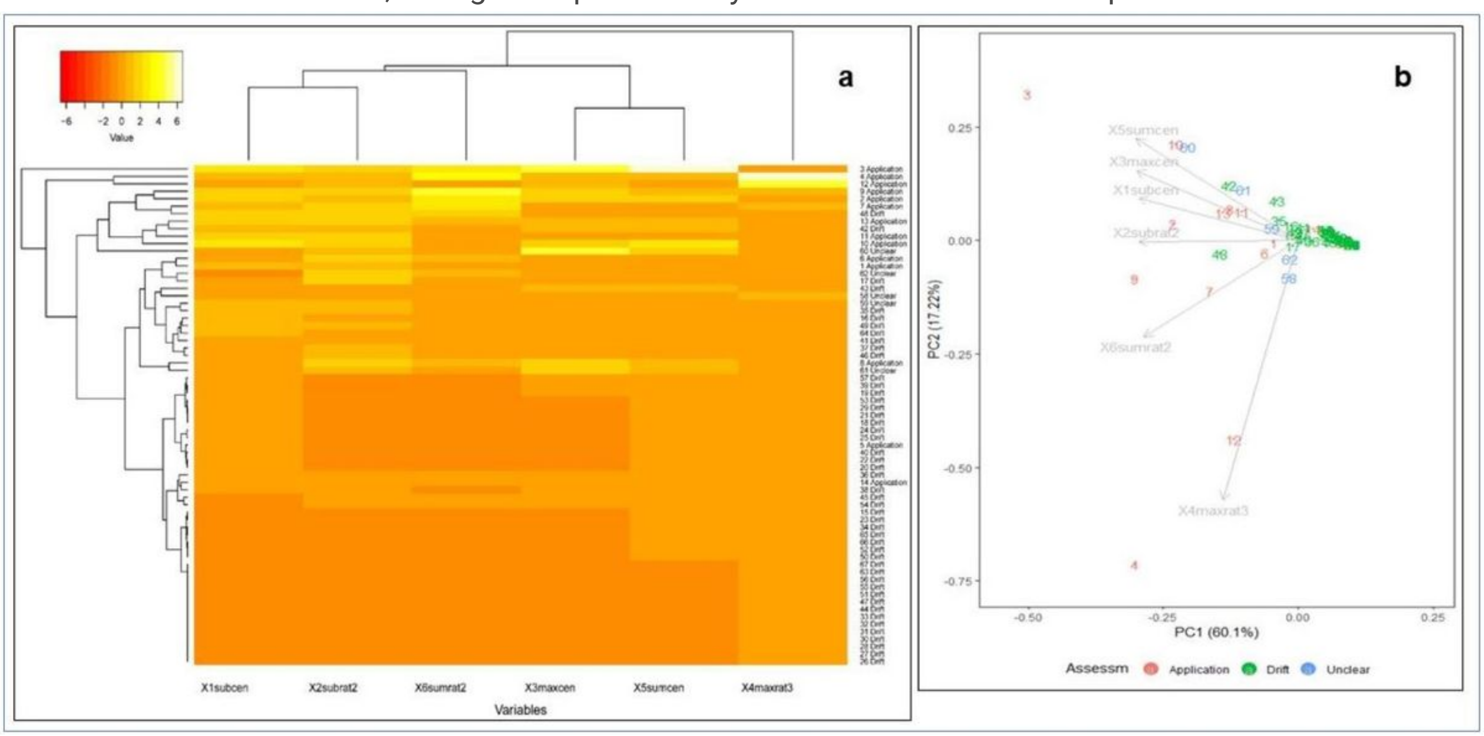


Figure 6

Due to technical limitations, the figure caption is only available in the manuscript file.

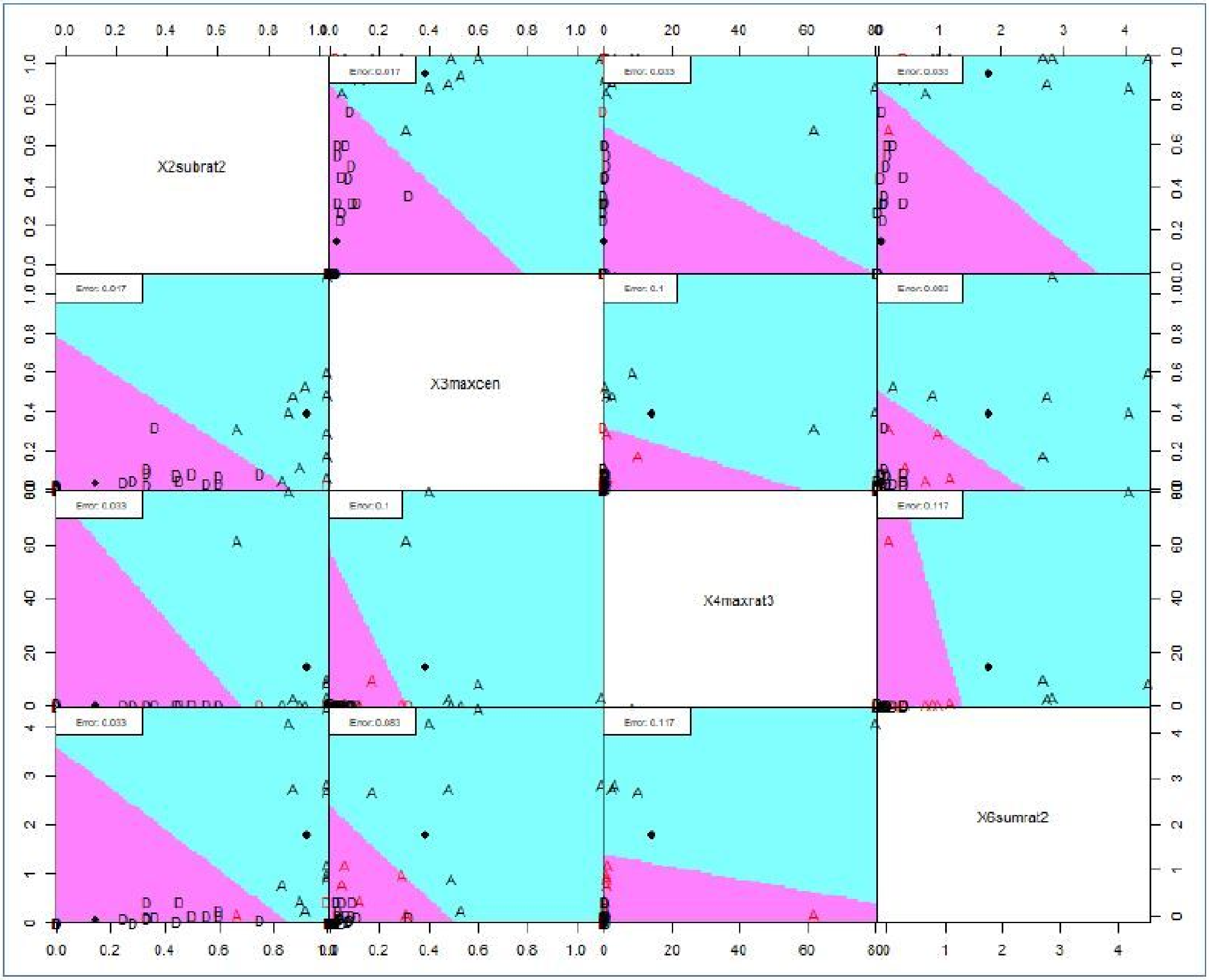


Figure 7

Due to technical limitations, the figure caption is only available in the manuscript file.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- SupplementaryMaterials.pdf
- LDA.sas
- LDA.sas
- SupplementaryDatasetSummaryFungicidesandVariablesperFarm.xlsx
- Rcodesuppli.r

- Rcodesuppli.r
- SupplementaryDatasetSummaryFungicidesandVariablesperFarm.xlsx
- SupplementaryDatasetVariablesonly.xlsx
- SupplementaryDatasetVariablesonly.xlsx
- SupplementaryMaterials.pdf