

SM text 1: sampling methods

- Soil sampling

One composite soil sample was taken per field. This sample resulted from the combination of 5 subsamples randomly collected across the field, using an auger. Sample depth varied among case study sites (CSS): 0–5 cm depth in CSS with permanent crops and 0–20 cm depth in CSS with arable land with tillage. The composite soil samples were stored at -20°C until shipment to the RECETOX laboratory for pesticide analysis.

- Crop sampling

One crop class was sampled per CSS, and one plant species per field. CSS 1, 2, 3, 6, 7, and 9 samples covered a single plant species; CSS 4, 5, 8, and 10 multiple plant species. At least 5 plants, spread over the area of the field, were used for the preparation of the field sample. The part of the plant to sample was crop-specific, depending on what part of the plant is used as harvesting product (leaves, roots/tuber, grains, grapes, fruits, spikes, seeds, etc). Plant samples were cooled or frozen (in case of having high water content and quick decomposition) until pesticide analyses at RECETOX laboratories.

- Outdoor air sampling

TIEM passive air sampling devices (Kruse-Platz et al., 2021) were used to sample outdoor air. These devices were in the field for approximately 2 months; they were installed one month before the middle of the growing season, overlapping then with the sampling time of most of the other environmental matrices. Two TIEM devices were installed per CSS: one at the edge of an organic field, and one at the edge of a conventional field. Fields were randomly selected from the pool of sampled fields, but preference was given to fields with no clear barriers, or on top of hills, to account also for long-distance transport. The TIEM head collector, installed at 1.5 meters from the soil surface, uses a polyurethane foam (PUF) disk positioned in a sheltered surrounding (enclosed dome open to outside air), to absorb (semi) volatile pesticides from the gas phase, and polyester filters (PEF), positioned underneath the dome, i.e. openly exposed to air, to collect particulate matter to analyze glyphosate and AMPA. After approximately 2 months in the field, the PEF and the PUF disks were removed from each device and sent to the KWALIS laboratory for pesticide analysis.

- Indoor dust sampling

Farmers were requested to replace their vacuum cleaner bag with a new one month prior to the field campaign and proceed with their normal house cleaning. During the field campaign, the contents of that bag were transferred to a Ziplock bag, which was then wrapped in aluminum foil and stored at -20°C prior to pesticide analysis by CIEMAT laboratories.

- Water sampling

Water samples were collected from small rivers, stream channels, small lakes, or ponds in the CSS regions. A minimum of 3 and a maximum of 8 water bodies were agreed to be sampled per CSS (due to limited lab capacity, and higher availability of monitoring data for such matrix). Riverine ecosystems located in the vicinity of organic and/or conventional agricultural fields were preferred. One composite water sample of ~2L was collected per water body. Water was collected sub-superficially, at three points of the water body (distancing at least 10 meters from each other), into a plastic bottle. After sampling, the water samples were deep-frozen and sent to the CIEMAT laboratory for pesticide analysis.

- Sediment sampling

Riverbed sediments were collected at the same locations as water samples. Sediments were not possible to collect at all the CSS, due to waterbodies features (i.e. too deep, or artificial channel substrate). A composite sediment sample was collected per water body. This composite sample resulted from a combination of 3 sub-samples, collected using a core sampler, stainless-steel scoop, or an auger, at the sediment surface. After thorough mixing of sub-samples, 1kg of composite sediment sample was placed in a polypropylene container and stored at -20°C until pesticide analysis at Wageningen University laboratories.

SM text 2: Analytical methods for pesticide determination

Complete details on SPRINT analytical work (method development and validation, and quality control measures) will be presented in matrix-specific publications. Here is a brief overview of what the lab work entailed.

- Soil

The soil samples were thawed and homogenized (until a visual homogeneous sample was obtained) and divided into two portions: one of 2 grams for the determination of glyphosate and its main metabolite AMPA, and one of 5 grams for analyses of multi-residue (LC-MS/MS & GC-MS/MS). The first aliquot was analyzed using the procedure described by Bento et al. (2016) and Yang et al. (2015), with KOH 0.6 M as extraction solvent, isotopically labeled standards, and FMOC-Cl as derivatization agent. The second aliquot was analyzed using a method similar to the one described in Silva et al. (2019), using the QuEChERS approach, acetonitrile as extraction solvent, and a PSA-based clean-up step, added to part of the extraction supernatant used for the GC analyses.

- Crop

The plant samples were also thawed, homogenized, and divided into two parts: one for the determination of glyphosate AMPA and one for the multi-residue. The former of 5g (2g in case of wet samples), the latter of 10 g (2 g in case of wet samples). Glyphosate and AMPA were determined using the

modified method described Yang et al. (2015), Goscinny et al. (2012) and Kaczyński et al. (2015) which involves isotopically labelled standards, 0.1% formic acid and dichloromethane-based extraction, and a FMOC-Cl based derivatization. The other pesticide residues were extracted using an adaptation of the QuEChERS approach, as described by Silva et al. (2019). Acetonitrile was used as extraction solvent, and dispersive sorbents for the extra clean-up step done before the GC analyses.

- Outdoor air

Lastly, the extraction of the TIEM-PUF disks was carried out with dichloromethane in a Soxhlet extractor. The samples were extracted for 24 h using at least 16 extraction cycles. After concentrating the extract to 1 ml under a stream of nitrogen, the extract was analysed by LC-MS/MS and GC-MS/MS. PEF filters: Glyphosate and AMPA were extracted from the PEF materials with 0.125 N hydrochloric acid and then derivatized with fluorenylmethoxycarbonyl. The analysis was conducted by means of LC-MS/MS.

- Indoor dust

Each indoor dust sample was analysed by three methods, for the determination of multi-residue analysis of pesticides, glyphosate & AMPA, and organochlorinated pesticides. Multi-residues were analysed via the QuEChERS approach, which involved labeled standards, acetonitrile containing 1% (v/v) of acetic acid as extraction solvent, and dSPE cleaning of the GC extract, followed by a solvent exchange to isooctane. Glyphosate and AMPA were analyzed as described in Mendez et al. (2017), also using labelled standards, but $\text{KH}_2\text{PO}_4/\text{Na}_2\text{B}_4\text{O}_7$ as extraction solvent, and a FMOC-based derivatization. Finally, organochlorinated pesticides were measured as described in de la Torre et al., (2020). Here the extraction was done with a hexane:acetone solution (3:1), and extract purification by a 1 g florisil column eluted with 16 mL of hexane.

- Water

The water samples thawed, filtered and spiked with the proper labeled standards were analysed by three different methods, for the determination of multi-residue analysis of pesticides, glyphosate & AMPA, and organochlorinated pesticides. Multi-residue analysis was based on Oasis HLB Solid-Phase Extraction (SPE) cartridges, conditioned with methanol and ultrapure water and eluted with methanol. The extract was evaporated under a stream of nitrogen and divided into two aliquots, for HPLC-MS/MS and GC-MS/MS. To analyse glyphosate and AMPA, water samples were treated with 0.1 M KH_2PO_4 and 0.1 M $\text{K}_3\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$ and derivatized with 6.5 mM FMOC-Cl. Then the extract was passed through Oasis HLB cartridge, eluted with methanol and finally analysed by HPLC-MS/MS. Organochlorine pesticides were extracted by liquid-liquid extraction with dichloromethane and analysed by GC-HRMS.

- Sediment

The sediment samples were analyzed as soil samples, with two minor adaptations. Sample preparation involved a centrifugation step at 3500 rpm for 5 min, the solid portion was decanted, and water was discarded. The aliquot for determination of glyphosate and AMPA and the aliquot for multi-residues both had 2 grams. The methods used were those described by Bento et al. (2016) and Yang et al. (2015), and Silva et al. (2019).

SM text 3: Sensitivity analysis by left-censored data imputation

Imputation was performed at matrix level, for pesticide residues detected in at least 40% of the samples. Residues present in all samples (100% detection) do not require imputation analyses. The medians of truncated distributions (i.e. non-imputed data, where only values $\geq$ limit of detection are accounted for) were compared to the medians of full distributions (non-imputed + imputed data) using the Mann Whitney U test. Names marked in **red** indicate pesticide residues where median from imputed data is significantly different (p-value < 0.05) from the non-imputed data.

Adding to what we described in the manuscript, from the 'survival' package we used survreg: Regression for a Parametric Survival Model. Here, we defined the parameter `dist="lognormal"`. It is important to stress that the data can be fitted to other types of distribution, such as Weibull, exponential and so on...However, based on the body of evidence on pesticides the overall trend of the distributions is lognormal. In the setting of the survival function the user can define the intervals of the expected censored data (see <https://www.rdocumentation.org/packages/survival/versions/2.11-4/topics/Surv>). Here, we define the lower boundary as a small number (e.g. $1E-9$) allowing for flexibility in the imputation and the upper-boundary as equal to LOD, meaning the imputed values can vary between LOD and lower boundary. Finally, an initial set of values is attributed to the left-censored data and the model ran X amount of times (iterations) to achieve the best fit. Here we used 100 iterations, although usually with 10 iterations the model already fully converges. The pesticides X,Y,Z,... concentrations are used as predictors of pesticide K concentrations. This means that if 80 out of 100 pesticides were detected in sample A and 20 out of 100 pesticides were detected in sample B, there is a higher likelihood for the non-detects (left-censored data) to be closer to LOD in sample A vs sample B.

- Soil

2 pesticide residues imputed -> 1 of them differ significantly after imputation

- | | |
|---|-------------------|
| 1 | DDE.p,p' |
| 2 | Hexachlorobenzene |

- Crop

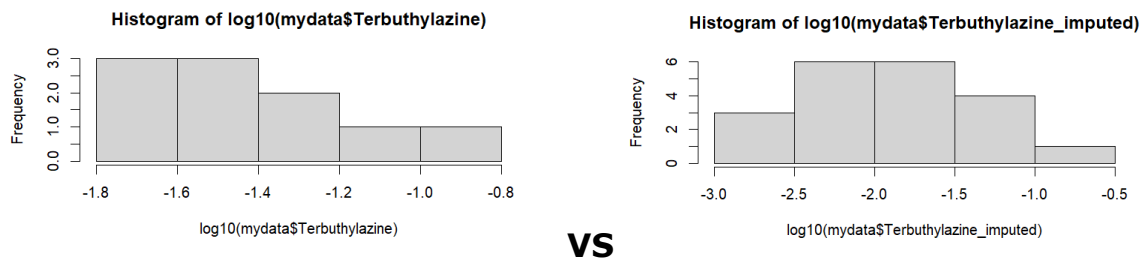
0 pesticides imputed

- Outdoor air
9 pesticide residues imputed -> 7 of them differ significantly after imputation

~~———— Glyphosate (100% detection);~~

- 1 AMPA
- 2 Folpet
- 3 Folpet_PHI
- 4 Metolachlor_S
- 5 Pendimethalin
- 6 Prosulfocarb
- 7 Prothioconazole_desthio
- 8 Terbutylazine
- 9 Terbutylazine_desethyl

Example for Terbutylazine (left non-imputed, right is imputed)



- Indoor dust
59 pesticides imputed -> 36 of them differ significantly after imputation

- 1 Glyphosate
- 2 **D2_4**
- 3 Acetamiprid
- 4 **Ametoctradin**
~~Azoxystrobin (100% detection)~~
- 5 **Bentazone**
- 6 **Boscalid**
- 7 **Bromoxynil**
~~Carbendazim (100% detection)~~
- 8 Chloridazon
- 9 Chlorothalonil.4.hydroxy
- 10 **Chlorpropham**
- 11 **Chlorpyrifos**
- 12 Chlorpyrifos..methyl..TCPy
- 13 **Clomazone**
- 14 **Cymoxanil**
- 15 **Cypermethrin**
- 16 **DDD.o.p.**
- 17 **DDD.p.p.**
- 18 DDE.p.p.
- 19 **Deltamethrin**
- 20 Dieldrin
- 21 Difenoconazole
- 22 **Diflufenican**
- 23 Dimethomorph
- 24 Diuron
~~Fipronil (100% detection)~~
~~Fipronil.sulfone (100% detection)~~

25 Flonicamid
26 Florasulam
Fludioxonil (100% detection)
27 Fluopicolide
28 Fluopyram
29 Flupyradifurone
30 Fluroxypyr
31 Fluxapyroxad
Hexachlorobenzene (100% detection)
32 Imazalil
Imidacloprid (100% detection)
33 Imidacloprid..desnitro
34 Isoproturon
Lindane (100% detection)
35 Mandipropamid
36 MCPA
37 Mecoprop..P.
38 Metalaxyl_M
39 Metolachlor_S
40 Metrafenone
41 Pendimethalin
42 Permethrin
Piperonyl.butoxide (100% detection)
43 Pirimicarb
44 Pirimiphos.methyl.DEAMPY
45 Prochloraz.BTS.44595
46 Propamocarb..hydrochloride
47 Propiconazole
48 Propoxur
49 Prosulfocarb
50 Prothioconazole_desthio
51 Pyraclostrobin
52 Pyrimethanil
Tebuconazole (100% detection)
53 Terbutylazine
54 Terbutryn
55 Thiabendazole
56 Thiacloprid
57 Thiamethoxam
58 Tolyfluanid.metabolite.DMST
59 Trifloxystrobin

- **Water**

15 pesticides imputed -> 11 of them differ significantly after imputation

- 1 Glyphosate
- 2 AMPA
- 3 DDD.o.p.
- 4 DDD.p.p.
- 5 DDE.p.p.
- 6 DDE.o.p.
- 7 DDT.o.p.
- 8 DDT.p.p.
- 9 Dieldrin
- 10 Piperonyl.butoxide
- 11 Tebuconazole
- 12 Terbutylazine
- 13 Terbutylazine_desethyl
- 14 Terbutryn
- 15 Trifloxystrobin_CGA321113

- **Sediment**

3 pesticides imputed -> 2 of them differ significantly after imputation

- 1 Glyphosate
- 2 AMPA
- 3 DDE.p.p

SM text 4: Sensitivity analysis by assigning the worst/best case score to the “no data” hazard entries

Human:

With this worst case sensitivity analysis, a hazard score of 2-8 was attributed to the 'No data' entries depending on the endpoint of concern. Regarding the best case sensitivity analysis, a hazard score of 0 was assigned, the same as “no effect”. The PPI was then calculated for all compounds.

Ecosystem

With regards to the worst case sensitivity analysis, a hazard score of 3 was attributed to “No data” entries as if they are all of high hazard, and a 0 was assigned in case of the best case sensitivity analysis. Then we calculated PPI for all compounds using the median concentration for comparison.

The new matrix specific curves, the PPI curve for human and ecosystem, and the top-10 compounds of each matrix for comparison are shown below.

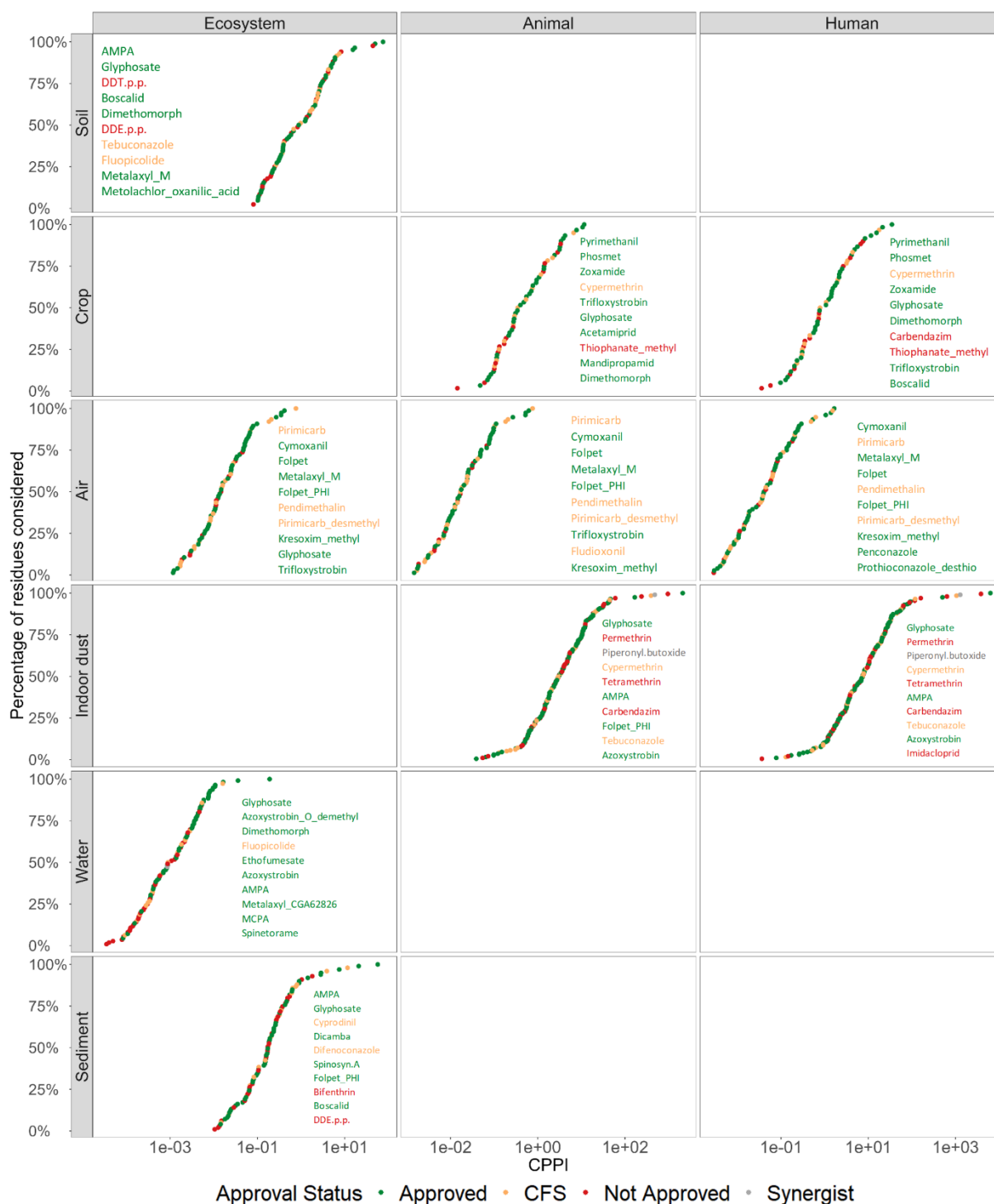


Figure 4-1 The worst case cumulative Pesticide Prioritization Indicator (CPPI) for each matrix where 'no data' entries are treated with the highest hazard score regarding the endpoints of concern. The red dots indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The names of the 10 compounds with the highest PPI values per panel are marked at the top left or bottom right corners.



Figure 4-2 The best case cumulative Pesticide Prioritization Indicator (CPPI) for each matrix where 'no data' entries are treated with 0. The red dots indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The names of the 10 compounds with the highest PPI values per panel are marked at the top left or bottom right corners.

Table 4-1 Top 10 compounds for each panel as the result of the sensitivity analysis in comparison with the original hazard score

Original	No data = highest score	No data = 0
Ecosystem - Soil		
AMPA	AMPA	AMPA
Glyphosate	Glyphosate	Glyphosate
Boscalid	DDT.p.p.	Boscalid
Dimethomorph	Boscalid	Dimethomorph
DDT.p.p.	Dimethomorph	DDE.p.p.
DDE.p.p.	DDE.p.p.	Tebuconazole
Tebuconazole	Tebuconazole	Fluopicolide
Fluopicolide	Fluopicolide	Metalaxyl_M
Metalaxyl_M	Metalaxyl_M	Metolachlor_oxanilic_acid
Metolachlor_oxanilic_acid	Metolachlor_oxanilic_acid	Iprovalicarb
Ecosystem - Air		
Pirimicarb	Pirimicarb	Pirimicarb
Cymoxanil	Cymoxanil	Cymoxanil
Folpet	Folpet	Folpet
Metalaxyl_M	Metalaxyl_M	Metalaxyl_M
Pendimethalin	Folpet_PHI	Pendimethalin
Kresoxim_methyl	Pendimethalin	Kresoxim_methyl
Folpet_PHI	Pirimicarb_desmethyl	Glyphosate
Glyphosate	Kresoxim_methyl	Trifloxystrobin
Trifloxystrobin	Glyphosate	Fludioxonil
Fludioxonil	Trifloxystrobin	Flutolanil
Ecosystem - Water		
Glyphosate	Glyphosate	Glyphosate
Dimethomorph	Azoxystrobin_O_demethyl	Dimethomorph
Fluopicolide	Dimethomorph	Fluopicolide
Azoxystrobin_O_demethyl	Fluopicolide	Ethofumesate
Ethofumesate	Ethofumesate	Azoxystrobin
Azoxystrobin	Azoxystrobin	MCPA
MCPA	AMPA	Spinetorame
Spinetorame	Metalaxyl_CGA62826	Dimethenamid_P
Dimethenamid_P	MCPA	Terbutylazine_desethyl
Terbutylazine_desethyl	Spinetorame	Metalaxyl_M
Ecosystem - Sediment		
Glyphosate	AMPA	Glyphosate
AMPA	Glyphosate	Cyprodinil
Cyprodinil	Cyprodinil	Difenoconazole
Difenoconazole	Dicamba	Bifenthrin
Dicamba	Difenoconazole	Boscalid
Bifenthrin	Spinosyn.A	Spiroxamine
Boscalid	Folpet_PHI	Deltamethrin
Spinosyn.A	Bifenthrin	Tebuconazole
Folpet_PHI	Boscalid	Cypermethrin
Spiroxamine	DDE.p.p.	Fluopicolide
Animal - Crop		
Pyrimethanil	Pyrimethanil	Pyrimethanil
Phosmet	Phosmet	Phosmet
Zoxamide	Zoxamide	Zoxamide
Cypermethrin	Cypermethrin	Cypermethrin
Trifloxystrobin	Trifloxystrobin	Trifloxystrobin
Glyphosate	Glyphosate	Glyphosate
Acetamiprid	Acetamiprid	Acetamiprid
Thiophanate_methyl	Thiophanate_methyl	Thiophanate_methyl
Mandipropamid	Mandipropamid	Mandipropamid
Dimethomorph	Dimethomorph	Dimethomorph

Animal - Air		
Pirimicarb	Pirimicarb	Pirimicarb
Cymoxanil	Cymoxanil	Cymoxanil
Folpet	Folpet	Folpet
Metalaxyl_M	Metalaxyl_M	Metalaxyl_M
Pendimethalin	Folpet_PHI	Pendimethalin
Pirimicarb_desmethyl	Pendimethalin	Pirimicarb_desmethyl
Trifloxystrobin	Pirimicarb_desmethyl	Trifloxystrobin
Fludioxonil	Trifloxystrobin	Fludioxonil
Kresoxim_methyl	Fludioxonil	Kresoxim_methyl
Penconazole	Kresoxim_methyl	Penconazole
Animal - Indoor dust		
Glyphosate	Glyphosate	Glyphosate
Permethrin	Permethrin	Permethrin
Piperonyl.butoxide	Piperonyl.butoxide	Piperonyl.butoxide
Cypermethrin	Cypermethrin	Cypermethrin
AMPA	Tetramethrin	AMPA
Tetramethrin	AMPA	Carbendazim
Carbendazim	Carbendazim	Tebuconazole
Tebuconazole	Folpet_PHI	Azoxystrobin
Azoxystrobin	Tebuconazole	Imidacloprid
Imidacloprid	Azoxystrobin	MCPA
Human - Crop		
Pyrimethanil	Pyrimethanil	Pyrimethanil
Phosmet	Phosmet	Phosmet
Cypermethrin	Cypermethrin	Cypermethrin
Zoxamide	Zoxamide	Zoxamide
Glyphosate	Glyphosate	Glyphosate
Carbendazim	Dimethomorph	Carbendazim
Thiophanate_methyl	Carbendazim	Thiophanate_methyl
Trifloxystrobin	Thiophanate_methyl	Trifloxystrobin
Boscalid	Trifloxystrobin	Boscalid
Pirimicarb	Boscalid	Pirimicarb
Human - Air		
Pirimicarb	Cymoxanil	Pirimicarb
Folpet	Pirimicarb	Folpet
Cymoxanil	Metalaxyl_M	Cymoxanil
Pendimethalin	Folpet	Pendimethalin
Folpet_PHI	Pendimethalin	Folpet_PHI
Metalaxyl_M	Folpet_PHI	Metalaxyl_M
Kresoxim_methyl	Pirimicarb_desmethyl	Kresoxim_methyl
Penconazole	Kresoxim_methyl	Penconazole
Prothioconazole_desthio	Penconazole	Prothioconazole_desthio
Glyphosate	Prothioconazole_desthio	Glyphosate
Human - Indoor dust		
Glyphosate	Glyphosate	Glyphosate
Permethrin	Permethrin	Permethrin
Piperonyl.butoxide	Piperonyl.butoxide	Piperonyl.butoxide
Cypermethrin	Cypermethrin	Cypermethrin
Tetramethrin	Tetramethrin	Tetramethrin
AMPA	AMPA	AMPA
Carbendazim	Carbendazim	Carbendazim
Tebuconazole	Tebuconazole	Tebuconazole
Folpet_PHI	Azoxystrobin	Folpet_PHI
Propiconazole	Imidacloprid	Propiconazole

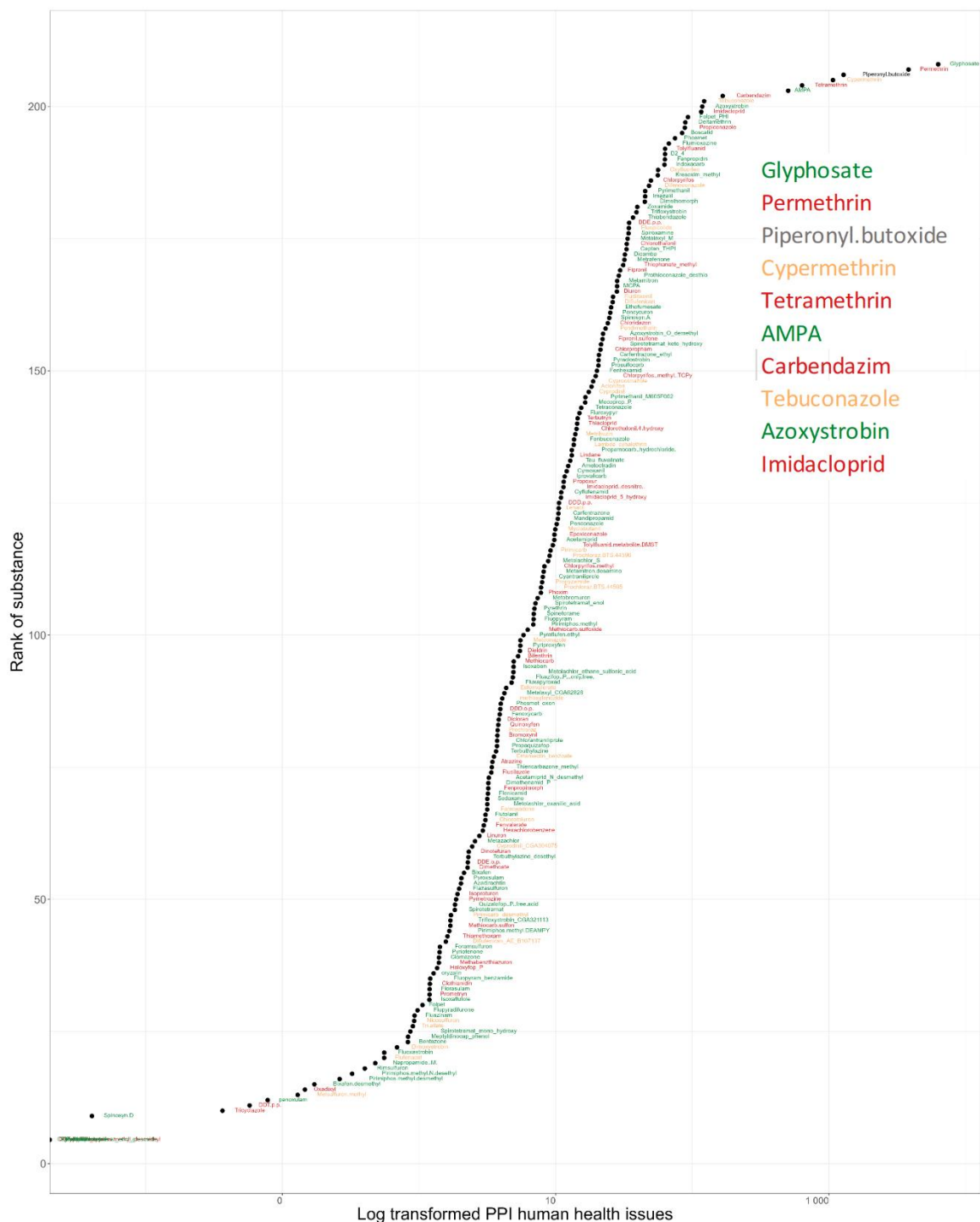


Figure 4-3 The worst case pesticide Prioritization Indicator (PPI) for humans where 'no data' entries are treated with the highest hazard score regarding the endpoints of concern. Residues are sorted from lowest to highest PPI values. The name of pesticide residues in red indicates not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The list on the top right corner corresponds to the 10 compounds with the highest PPI values.

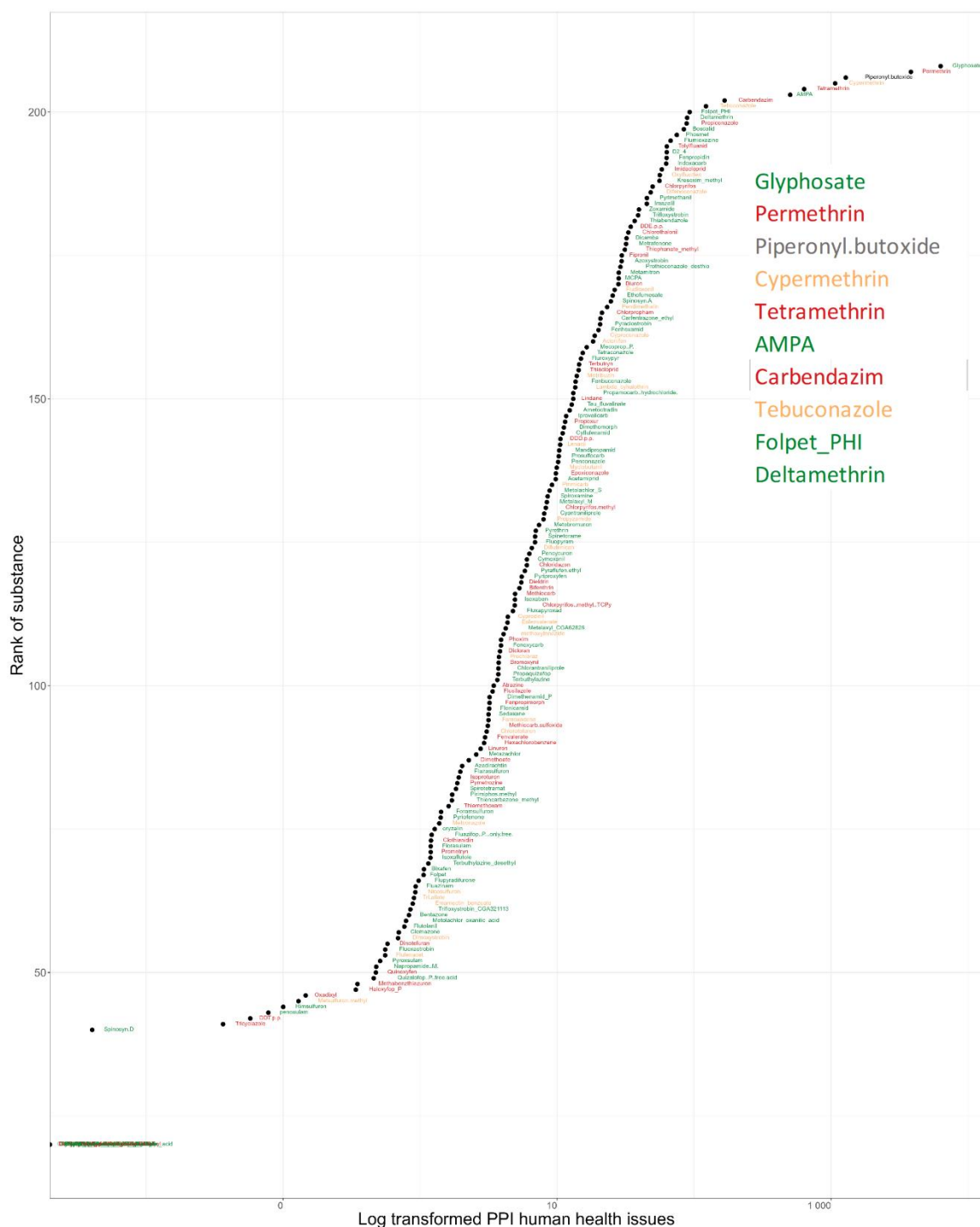


Figure 4-4 The best case pesticide Prioritization Indicator (PPI) for humans where 'no data' entries are treated with 0. Residues are sorted from lowest to highest PPI values. The name of pesticide residues in red indicates not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The list on the top right corner corresponds to the 10 compounds with the highest PPI values.

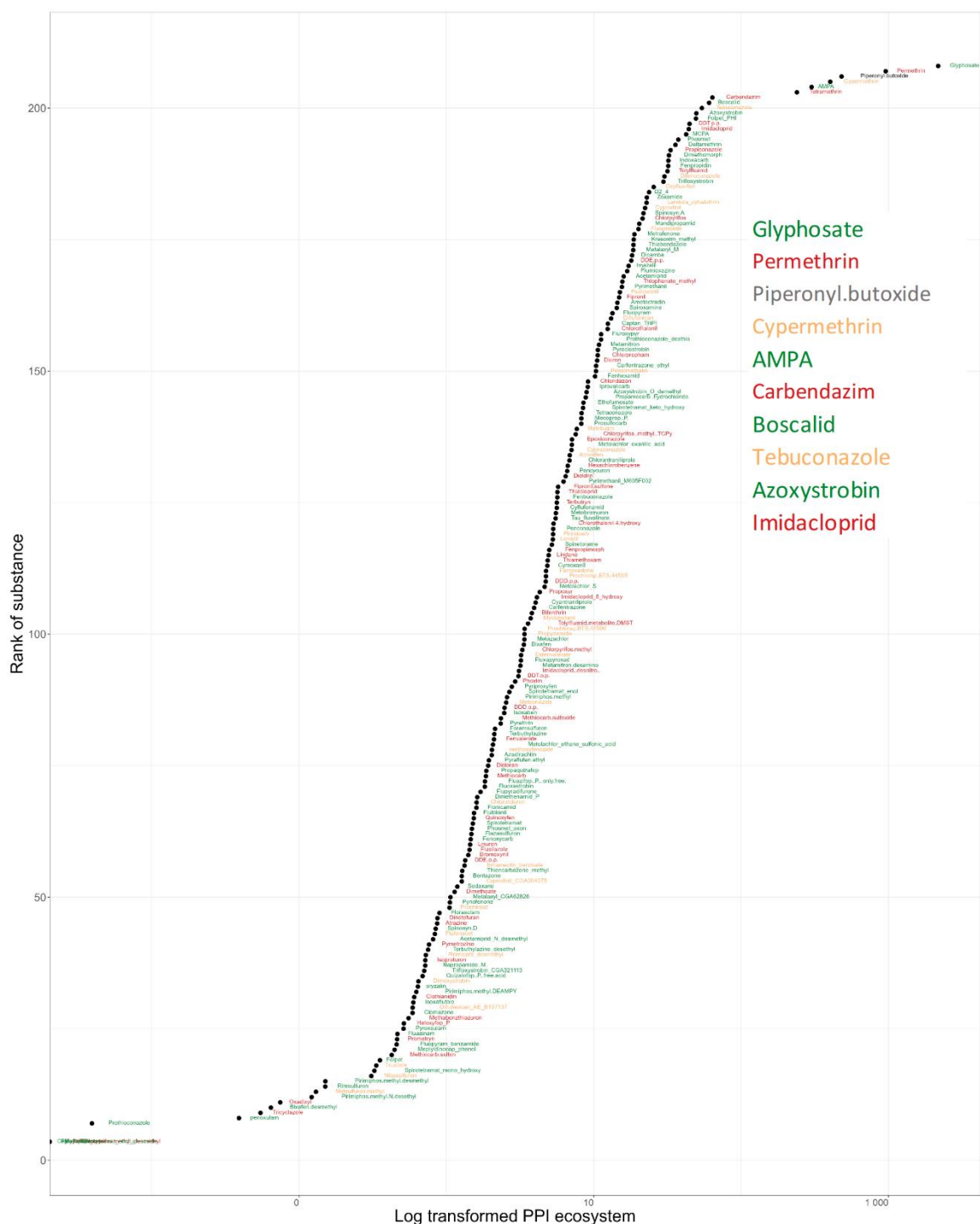


Figure 4-5. The worst case Pesticide Prioritization Indicator (PPI) for ecosystems where 'no data' entries are treated with 0. Residues are sorted from lowest to highest PPI values. The name of pesticide residues in red indicates not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The list on the top right corner corresponds to the 10 compounds with the highest PPI values.

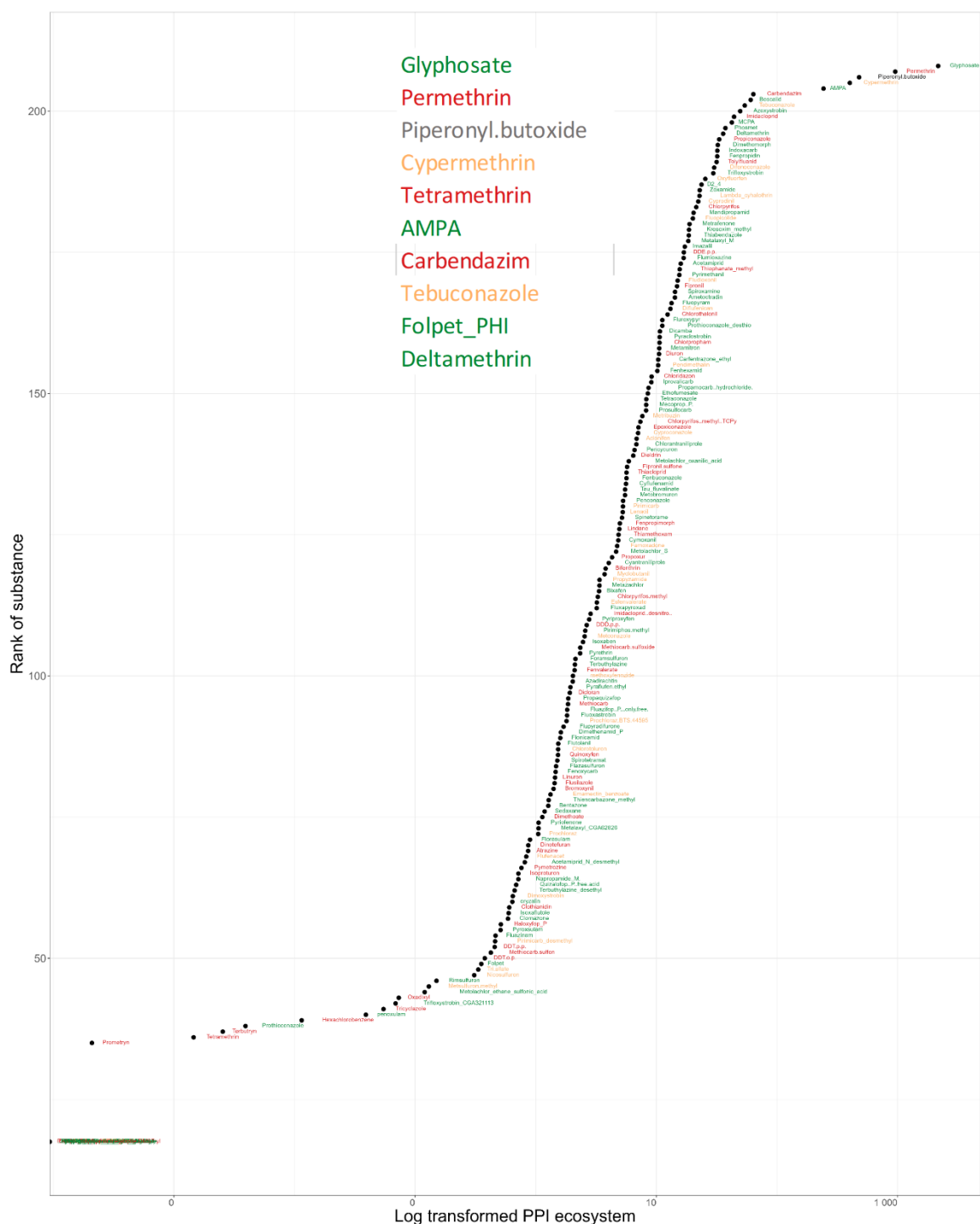


Figure 4-6. The best case Pesticide Prioritization Indicator (PPI) for ecosystems where 'no data' entries are treated with 3 (meaning the highest hazard score). Residues are sorted from lowest to highest PPI values. The name of pesticide residues in red indicates not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The list on the top right corner corresponds to the 10 compounds with the highest PPI values.

Table 4-2 Top 10 compounds for ecosystem and human health issues as the result of the sensitivity analysis in comparison with the original PPI.

Original	No data = highest score	No data = 0
Ecosystem overall PPI		
Glyphosate	Glyphosate	Glyphosate
Permethrin	Permethrin	Permethrin
Piperonyl.butoxide	Piperonyl.butoxide	Piperonyl.butoxide
Cypermethrin	Cypermethrin	Cypermethrin
AMPA	AMPA	AMPA
Tetramethrin	Tetramethrin	Carbendazim
Carbendazim	Carbendazim	Boscalid
Boscalid	Boscalid	Tebuconazole
Tebuconazole	Tebuconazole	Azoxystrobin
Azoxystrobin	Azoxystrobin	Imidacloprid
Human overall PPI		
Glyphosate	Glyphosate	Glyphosate
Permethrin	Permethrin	Permethrin
Piperonyl.butoxide	Piperonyl.butoxide	Piperonyl.butoxide
Cypermethrin	Cypermethrin	Cypermethrin
Tetramethrin	Tetramethrin	Tetramethrin
AMPA	AMPA	AMPA
Carbendazim	Carbendazim	Carbendazim
Tebuconazole	Tebuconazole	Tebuconazole
Folpet_PHI	Azoxystrobin	Folpet_PHI
Deltamethrin	Imidacloprid	Deltamethrin

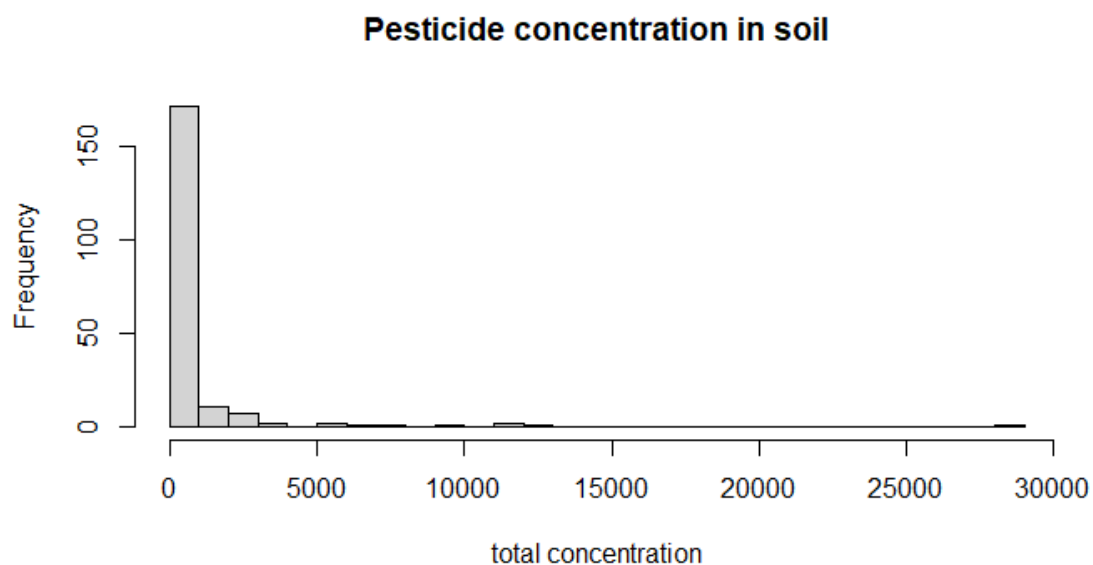
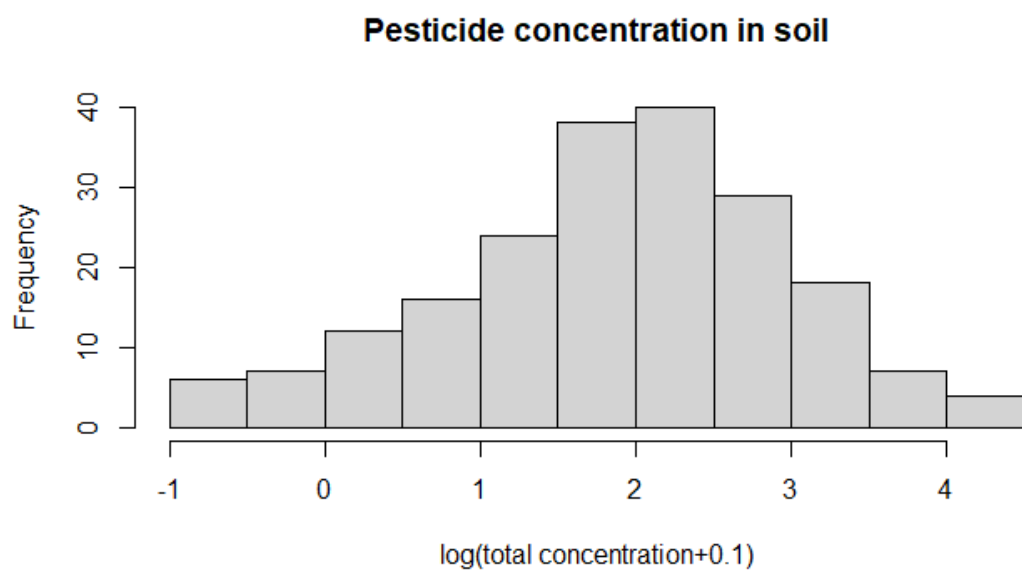
SM text 5: Complete statistical report on comparisons of number and total pesticide residues between conventional and organic farming

- Soil

Response: total concentration of pesticide residues in soils

Aim: comparison of conventional and organic farms

Method: LMM for $\log(\text{total_concentration} + 0.1)$



```
## Linear mixed model fit by REML. t-tests use Satterthwaite's method [  
## lmerModLmerTest]
```

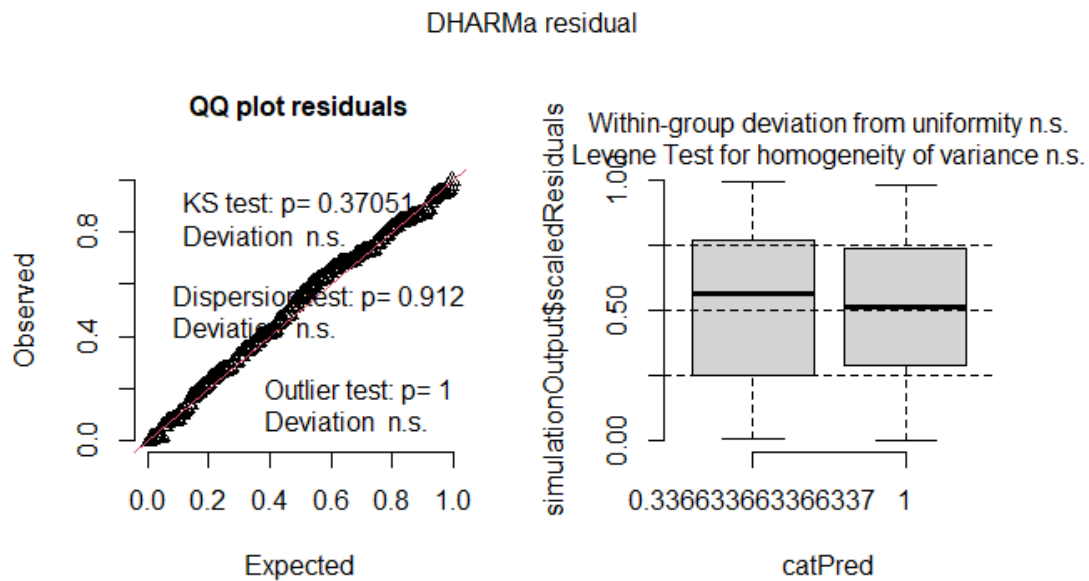
```
## Formula:
## log10(Total_concentration + 0.1) ~ farmtype + (1 | Country/farmtype
/Sample)
##
## REML criterion at convergence: 506
##
## Scaled residuals:
##      Min       1Q   Median       3Q      Max
## -1.3794 -0.2357  0.0274  0.2727  0.8939
##
## Random effects:
##      Groups                Name         Variance Std.Dev.
## Sample:(farmtype:Country) (Intercept) 0.4927   0.7019
## farmtype:Country          (Intercept) 0.3140   0.5604
## Country                    (Intercept) 0.0910   0.3017
## Residual                    0.1137   0.3372
##
## Fixed effects:
##              Estimate Std. Error    df t value Pr(>|t|)
## (Intercept)    2.3831    0.2161 17.6370 11.026 2.43e-09
## farmtypeOrganic -1.0531    0.2744  9.0336 -3.838 0.00395
##
## Correlation of Fixed Effects: (Intr)
## frmtypOrgnc -0.634

## Type II Analysis of Variance Table with Kenward-Roger's method
##              Sum Sq Mean Sq NumDF  DenDF F value  Pr(>F)
## farmtype 1.6738  1.6738    1 8.9961 14.725 0.003986

## farmtype    emmean    SE  df lower.CL upper.CL
## Conventional  2.38 0.216 17.3   1.928    2.84
## Organic       1.33 0.216 17.4   0.874    1.79
##
## Degrees-of-freedom method: kenward-roger
## Results are given on the log10(mu + 0.1) (not the response) scale.
## Confidence level used: 0.95
##
## $contrasts
##      contrast      estimate    SE df t.ratio p.value
## Conventional - Organic    1.05 0.274  9  3.837 0.0040
##
## Degrees-of-freedom method: kenward-roger
## Results are given on the log10 (not the response) scale.

## farmtype    response    SE  df lower.CL upper.CL
## Conventional 241.5 120.3 17.3  84.57   689
## Organic      21.3  10.7 17.4   7.39    61
##
```

```
## Degrees-of-freedom method: kenward-roger
## Confidence level used: 0.95
## Intervals are back-transformed from the log10(mu + 0.1) scale
```

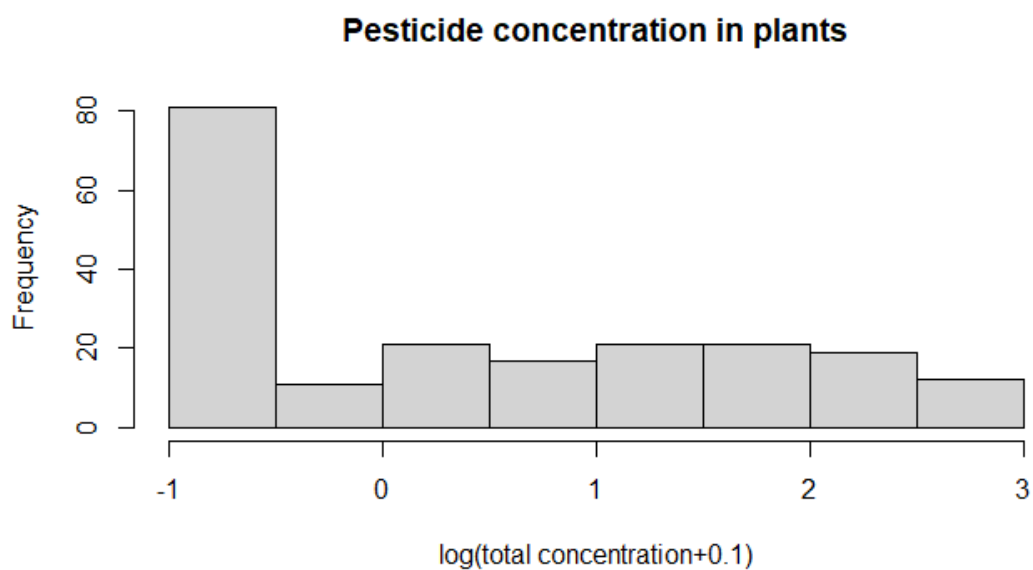


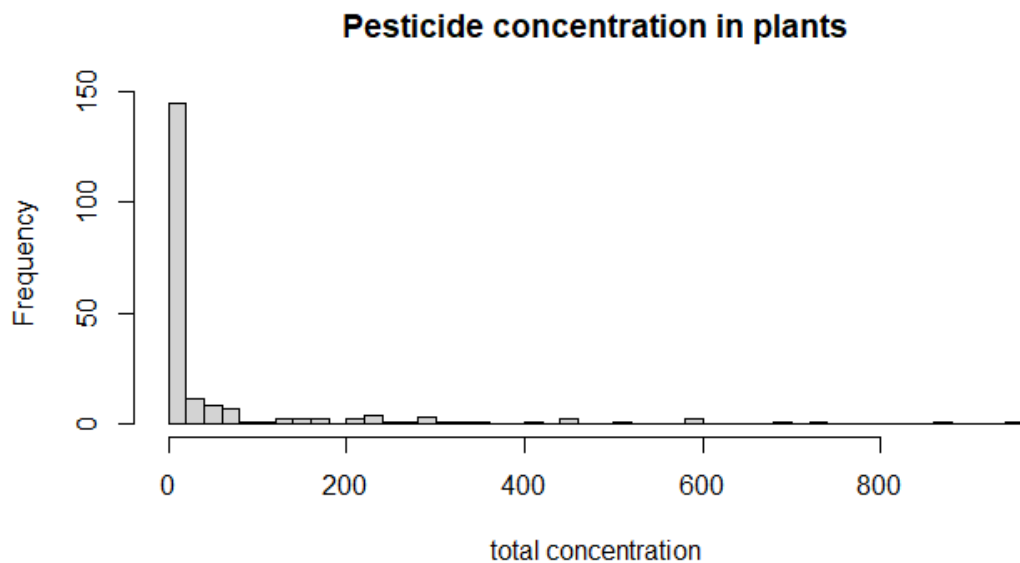
- Crop

Response: total concentration of pesticide residues in plants

Aim: comparison of conventional and organic farms

Method: GLMM with Tweedie distribution and log-link





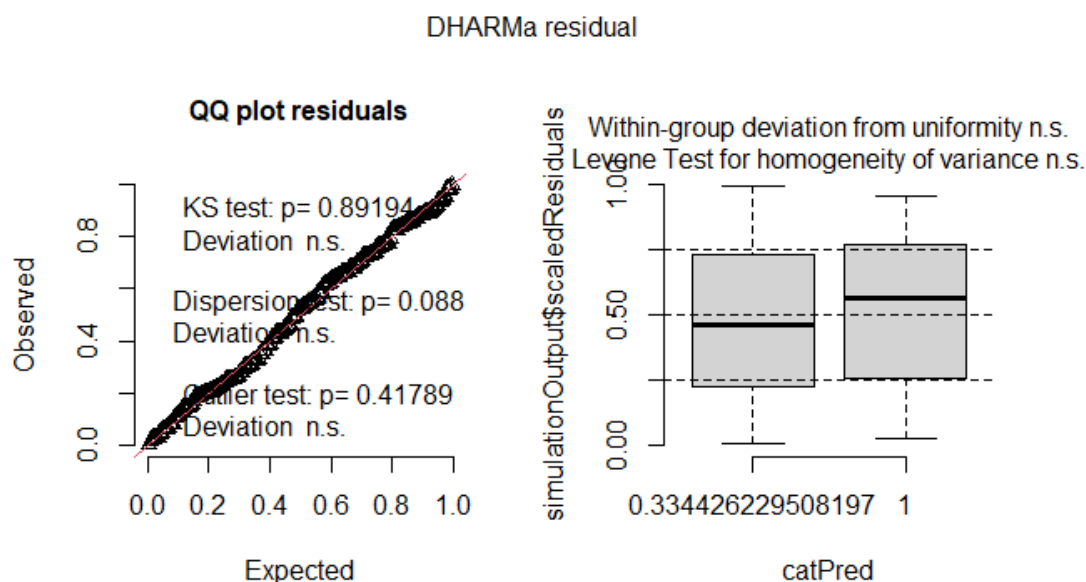
```
## Family: tweedie ( log )
## Formula:
## Total_concentration ~ farmtype + (1 | Country/farmtype/Sample)
##
##      AIC      BIC  logLik deviance df.resid
## 1386.1 1409.3 -686.0 1372.1    196
##
## Random effects:
##
## Conditional model:
## Groups              Name      Variance Std.Dev.
## Sample:farmtype:Country (Intercept) 3.667  1.9149
## farmtype:Country      (Intercept) 0.841  0.9171
## Country                (Intercept) 3.613  1.9009
##
## Dispersion parameter for tweedie family (): 1.92
##
## Conditional model:
##              Estimate Std. Error z value Pr(>|z|)
## (Intercept)   2.6772    0.7105  3.768 0.000164
## farmtypeOrganic -3.9800    0.5778 -6.888 5.64e-12

## Analysis of Deviance Table (Type II Wald chisquare tests)
##
## Response: Total_concentration
##      Chisq Df Pr(>Chisq)
## farmtype 47.45 1 5.642e-12

## $emmeans
## farmtype  emmean  SE df lower.CL upper.CL
## Conventional 2.68 0.710 196 1.28 4.08
```

```
## Organic      -1.30 0.782 196   -2.85   0.24
##
## Results are given on the log (not the response) scale.
## Confidence level used: 0.95
##
## $contrasts
## contrast      estimate    SE df t.ratio p.value
## Conventional - Organic    3.98 0.578 196   6.888 <.0001
##
## Results are given on the log (not the response) scale.

## farmtype    response    SE df lower.CL upper.CL
## Conventional 14.545 10.334 196   3.5826   59.05
## Organic      0.272  0.213 196   0.0581    1.27
##
## Confidence level used: 0.95
## Intervals are back-transformed from the log scale
```



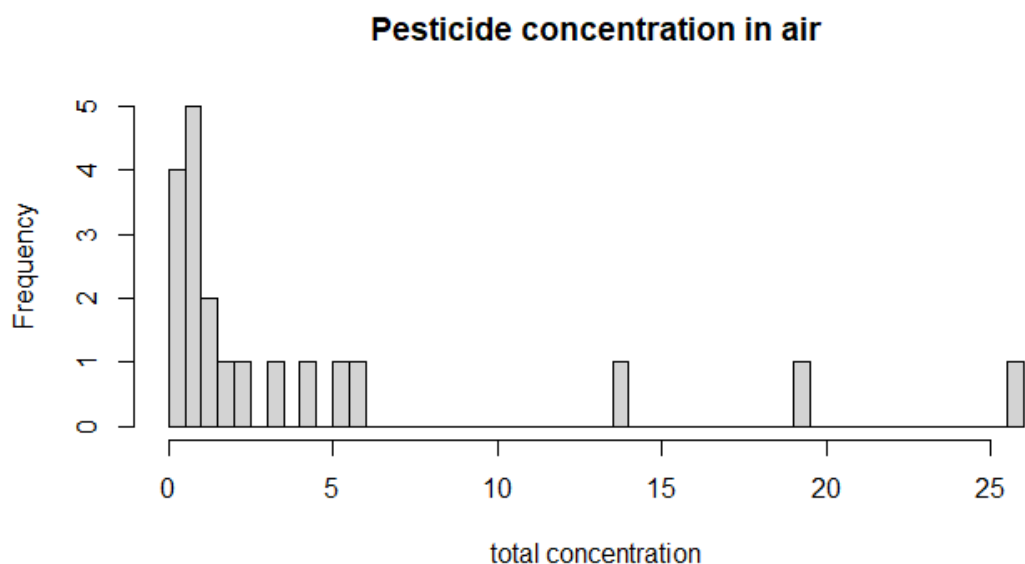
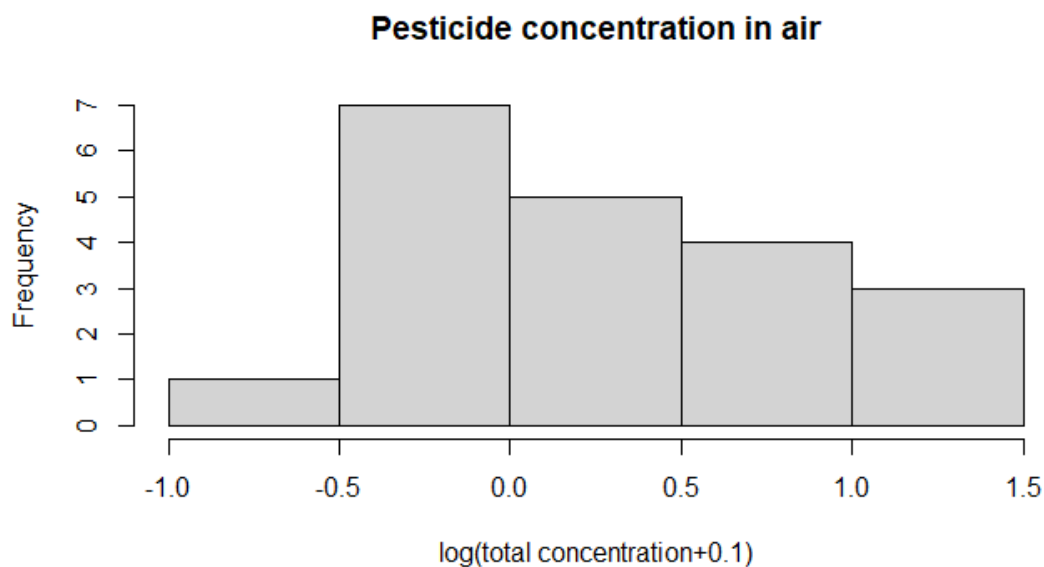
- Outdoor air

Response: total concentration of pesticide residues in air

Aim: comparison of conventional and organic farms

Method: LMM for $\log(\text{total_concentration} + 0.1)$

Analyze as randomized block design, with random blocks (=country/crop) effects. For air it is not possible to specify random country:farmtype interaction, because there no replicates of farmtype within sample (=country/crop).



```
## Linear mixed model fit by REML. t-tests use Satterthwaite's method [
## lmerModLmerTest]
## Formula: log10(Total_concentration + 0.1) ~ farmtype + (1 | Country
## )
##
## REML criterion at convergence: 25.5
##
## Scaled residuals:
##      Min       1Q   Median       3Q      Max
## -1.41755 -0.49824 -0.05376  0.72043  1.01776
##
## Random effects:
##  Groups   Name      Variance Std.Dev.
```



```

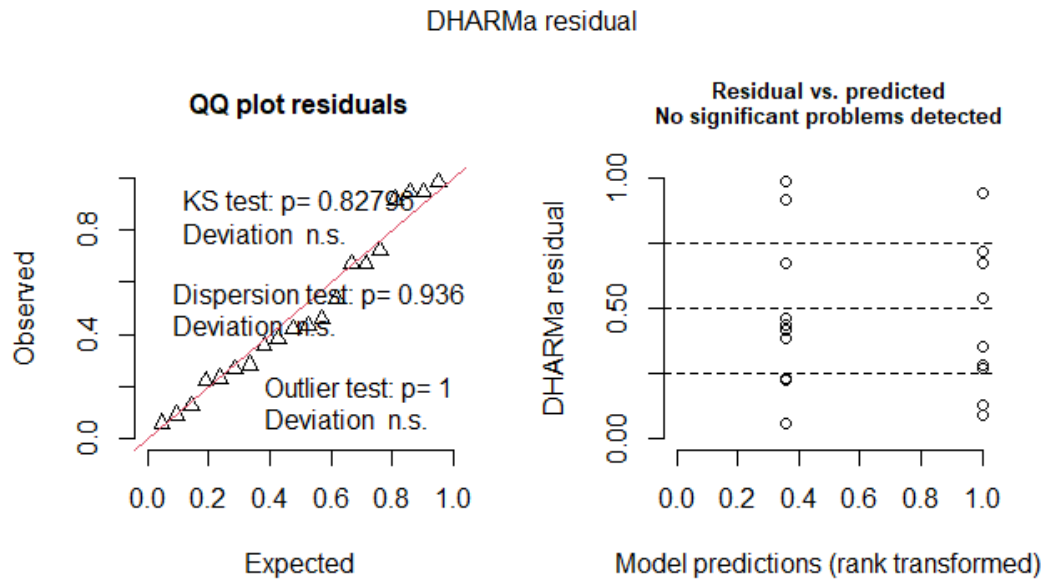
## Country (Intercept) 0.20728 0.4553
## Residual          0.07152 0.2674
##
## Fixed effects:
##           Estimate Std. Error    df t value Pr(>|t|)
## (Intercept)    0.5052    0.1670 11.5924   3.026 0.01093
## farmtypeOrganic -0.4184    0.1196  9.0000  -3.498 0.00675
##
## Correlation of Fixed Effects:
##           (Intr)
## frmtypOrgnc -0.358

## Type II Analysis of Variance Table with Kenward-Roger's method
##           Sum Sq Mean Sq NumDF DenDF F value  Pr(>F)
## farmtype 0.87511 0.87511    1    9 12.236 0.006746

## $emmeans
## farmtype  emmean  SE  df lower.CL upper.CL
## Conventional 0.5052 0.167 11.6  0.140  0.870
## Organic      0.0868 0.167 11.6 -0.278  0.452
##
## Degrees-of-freedom method: kenward-roger
## Results are given on the log10(mu + 0.1) (not the response) scale.
## Confidence level used: 0.95
##
## $contrasts
## contrast          estimate  SE df t.ratio p.value
## Conventional - Organic  0.418 0.12 9   3.498 0.0067
##
## Degrees-of-freedom method: kenward-roger
## Results are given on the log10 (not the response) scale.

## farmtype  response  SE  df lower.CL upper.CL
## Conventional  3.10 1.23 11.6  1.280  7.32
## Organic       1.12 0.47 11.6  0.427  2.73
##
## Degrees-of-freedom method: kenward-roger
## Confidence level used: 0.95
## Intervals are back-transformed from the log10(mu + 0.1) scale

```

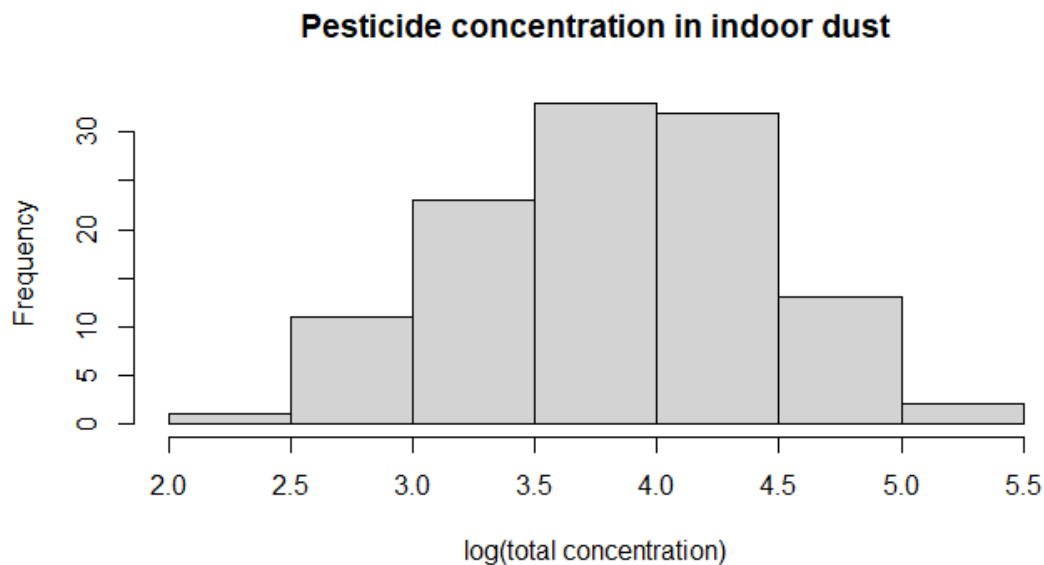


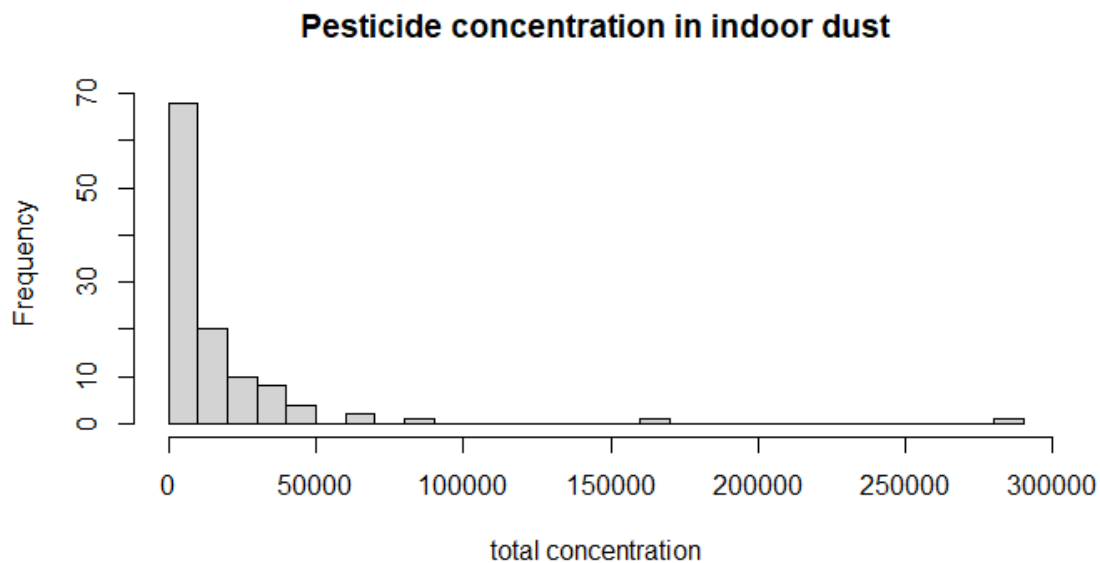
- Indoor dust

Response: total concentration of pesticide residues in indoor dust

Aim: comparison of conventional and organic farms

Method: LMM for $\log(\text{total_concentration})$

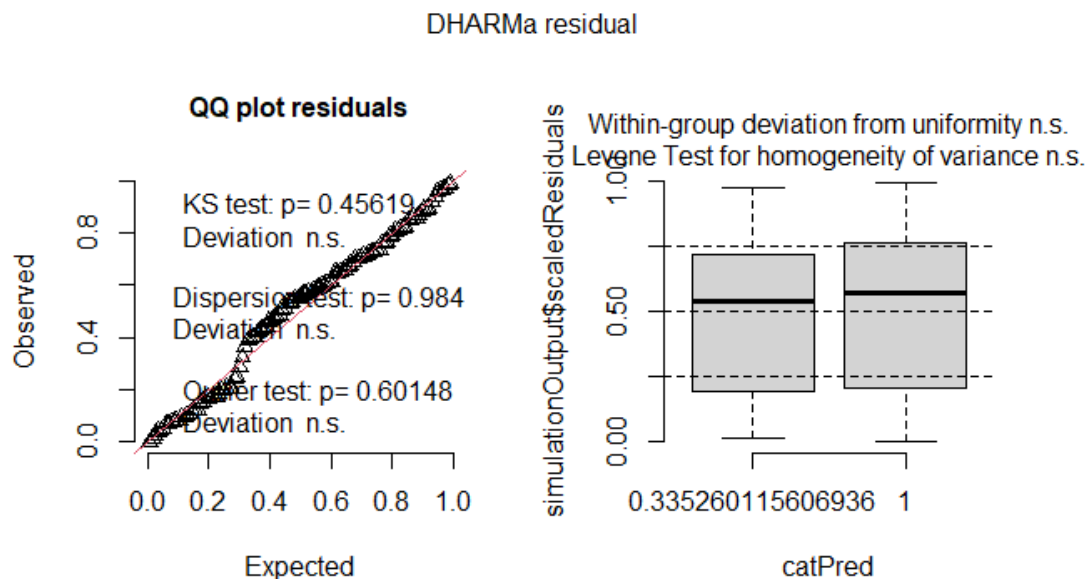




```
## Linear mixed model fit by REML. t-tests use Satterthwaite's method [
## lmerModLmerTest]
## Formula: log10(Total_concentration + 0.1) ~ farmtype + (1 | Country
/farmtype)
##
## REML criterion at convergence: 182.2
##
## Scaled residuals:
##   Min     1Q  Median     3Q      Max
## -3.7547 -0.5958  0.0546  0.5787  2.3058
##
## Random effects:
##   Groups             Name             Variance Std.Dev.
## farmtype:Country (Intercept) 0.04112  0.2028
## Country              (Intercept) 0.03377  0.1838
## Residual                                0.23434  0.4841
##
## Fixed effects:
##              Estimate Std. Error    df t value Pr(>|t|)
## (Intercept)    4.0581    0.1090 15.6699 37.217 <2e-16
## farmtypeOrganic -0.4920    0.1308  5.5488 -3.762  0.0108
##
## Correlation of Fixed Effects:
##              (Intr)
## frmtypOrgnc -0.594
##
## Type II Analysis of Variance Table with Kenward-Roger's method
##              Sum Sq Mean Sq NumDF  DenDF F value  Pr(>F)
## farmtype  3.2571  3.2571     1  8.5321  13.899 0.005175
```

```
## $emmeans
## farmtype    emmean    SE    df lower.CL upper.CL
## Conventional  4.06 0.110 16.1    3.83    4.29
## Organic       3.57 0.111 16.0    3.33    3.80
##
## Degrees-of-freedom method: kenward-roger
## Results are given on the log10(mu + 0.1) (not the response) scale.
## Confidence level used: 0.95
##
## $contrasts
## contrast          estimate    SE    df t.ratio p.value
## Conventional - Organic  0.492 0.132 8.53   3.728  0.0052
##
## Degrees-of-freedom method: kenward-roger
## Results are given on the log10 (not the response) scale.

## farmtype    response    SE    df lower.CL upper.CL
## Conventional 11432 2885 16.1    6697 19515
## Organic      3682  938 16.0    2146  6318
##
## Degrees-of-freedom method: kenward-roger
## Confidence level used: 0.95
## Intervals are back-transformed from the log10(mu + 0.1) scale
```

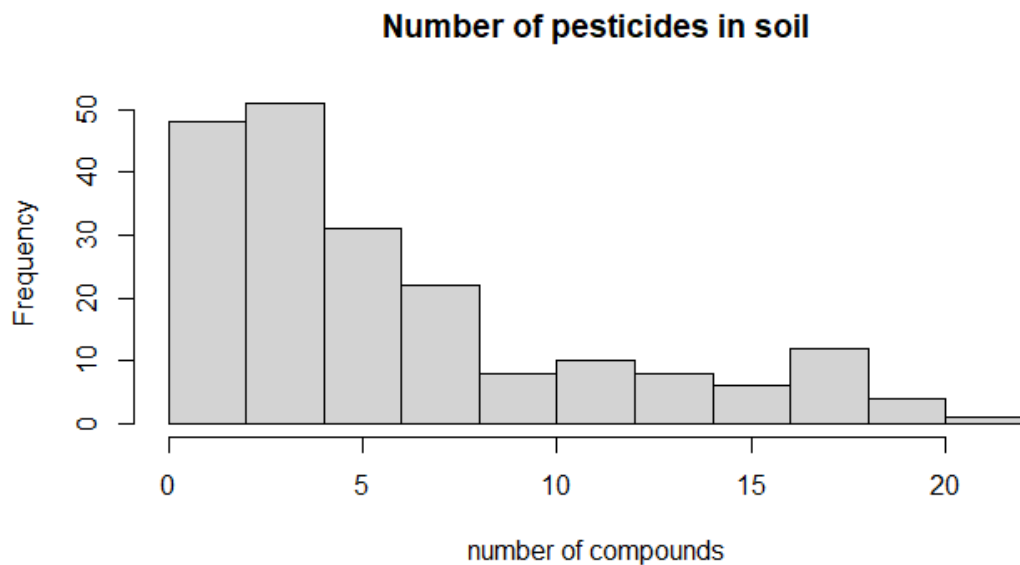


- Soil

Response: number of pesticides in soils

Aim: comparison of conventional and organic farms

Method: GLMM with Poisson distribution and log link



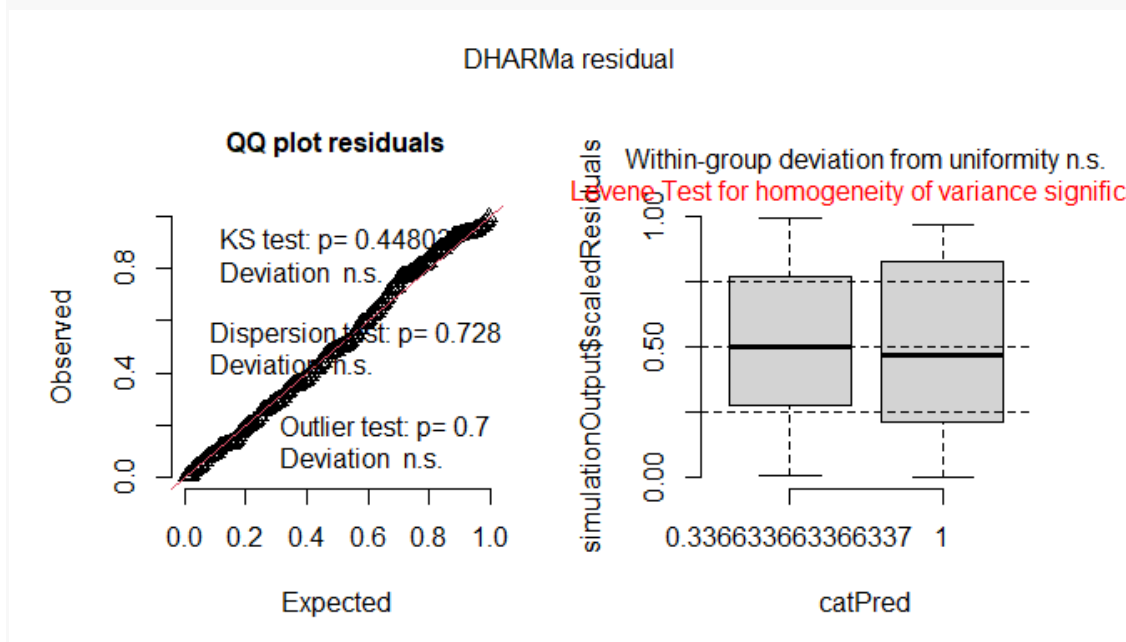
```
## Family: poisson ( log )
## Formula:      Amount_PPP ~ farmtype + (1 | Country/farmtype/Sa
mple)
##
##      AIC      BIC  logLik deviance df.resid
##  985.4   1001.9  -487.7   975.4     196
##
## Random effects:
##
## Conditional model:
## Groups              Name      Variance Std.Dev.
## Sample:farmtype:Country (Intercept) 0.04624  0.2150
## farmtype:Country        (Intercept) 0.08284  0.2878
## Country                 (Intercept) 0.09528  0.3087

## Conditional model:
##              Estimate Std. Error z value Pr(>|z|)
## (Intercept)    2.0956    0.1405  14.916 < 2e-16
## farmtypeOrganic -0.9929    0.1492  -6.656 2.81e-11

## Analysis of Deviance Table (Type II Wald chisquare tests)
##
## Response: Amount_PPP
##              Chisq Df Pr(>Chisq)
## farmtype 44.304  1 2.812e-11

## $emmeans
## farmtype    emmean    SE df lower.CL upper.CL
## Conventional  2.1 0.140 196  1.819    2.37
## Organic       1.1 0.148 196  0.811    1.39
```

```
##
## Results are given on the log (not the response) scale.
## Confidence level used: 0.95
##
## $contrasts
## contrast      estimate    SE df t.ratio p.value
## Conventional - Organic  0.993 0.149 196  6.656 <.0001
##
## Results are given on the log (not the response) scale.
## farmtype    rate    SE df lower.CL upper.CL
## Conventional 8.13 1.142 196   6.16  10.73
## Organic      3.01 0.445 196   2.25   4.03
##
## Confidence level used: 0.95
## Intervals are back-transformed from the log scale
```

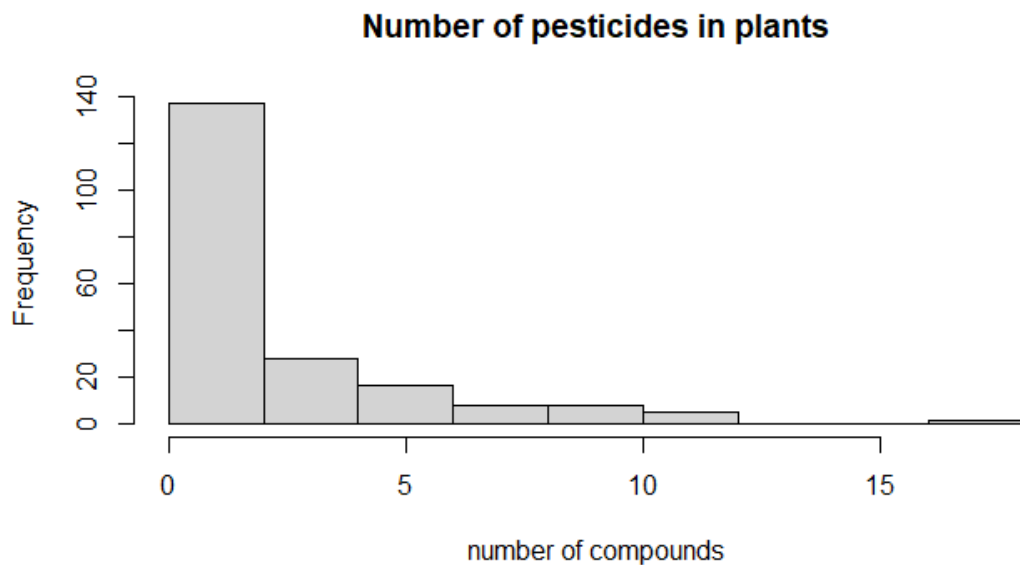


- Crop

Response: number of pesticides in plants

Aim: comparison of conventional and organic farms

Method: GLMM with Poisson distribution and log link



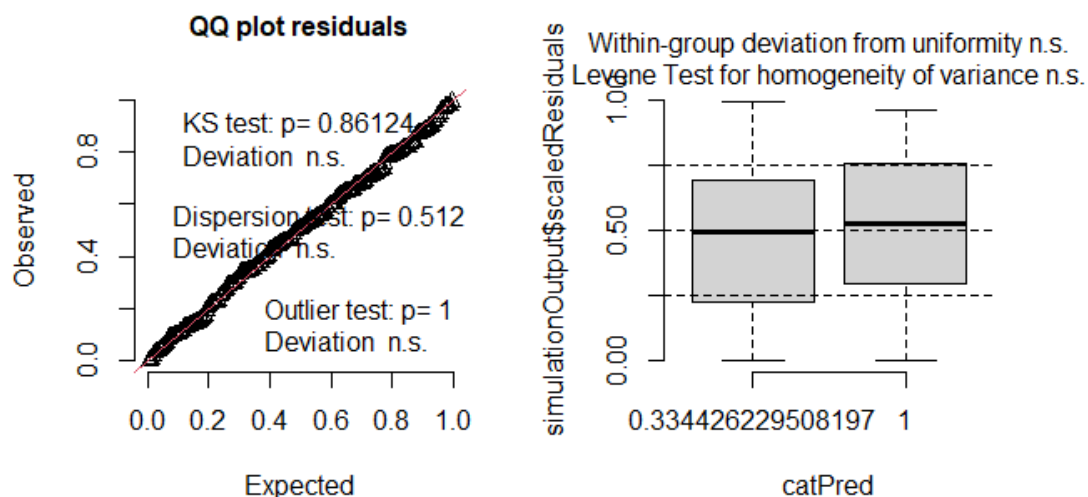
```
## Family: poisson ( log )
## Formula:      Amount_PPP ~ farmtype + (1 | Country/farmtype/Sa
mple)
##
##      AIC      BIC  logLik deviance df.resid
##  672.6    689.1  -331.3   662.6     198
##
## Random effects:
##
## Conditional model:
## Groups              Name      Variance Std.Dev.
## Sample:farmtype:Country (Intercept) 0.2067  0.4547
## farmtype:Country      (Intercept) 0.1551  0.3938
## Country                (Intercept) 0.8790  0.9375
##
## Conditional model:
##              Estimate Std. Error z value Pr(>|z|)
## (Intercept)    0.9433    0.3361  2.807  0.005
## farmtypeOrganic -1.8262    0.2525 -7.232 4.75e-13
##
## Analysis of Deviance Table (Type II Wald chisquare tests)
##
## Response: Amount_PPP
```

```
##          Chisq Df Pr(>Chisq)
## farmtype 52.306  1  4.748e-13

## $emmeans
## farmtype   emmean   SE df lower.CL upper.CL
## Conventional 0.943 0.336 198  0.281  1.606
## Organic     -0.883 0.368 198 -1.608 -0.158
##
## Results are given on the log (not the response) scale.
## Confidence level used: 0.95
##
## $contrasts
## contrast          estimate   SE df t.ratio p.value
## Conventional - Organic    1.83 0.253 198  7.232 <.0001
##
## Results are given on the log (not the response) scale.

## farmtype   rate   SE df lower.CL upper.CL
## Conventional 2.568 0.863 198  1.32  4.983
## Organic      0.414 0.152 198  0.20  0.854
##
## Confidence level used: 0.95
## Intervals are back-transformed from the log scale
```

DHARMA residual

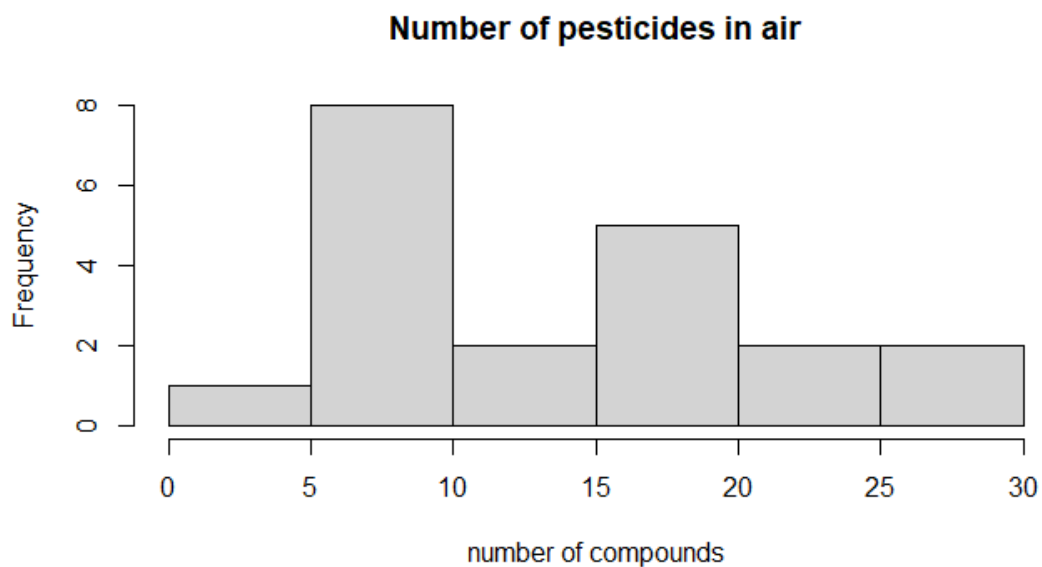


- Outdoor air

Response: number of pesticides in the air

Aim: comparison of conventional and organic farms

Method: GLMM with Poisson distribution and log link



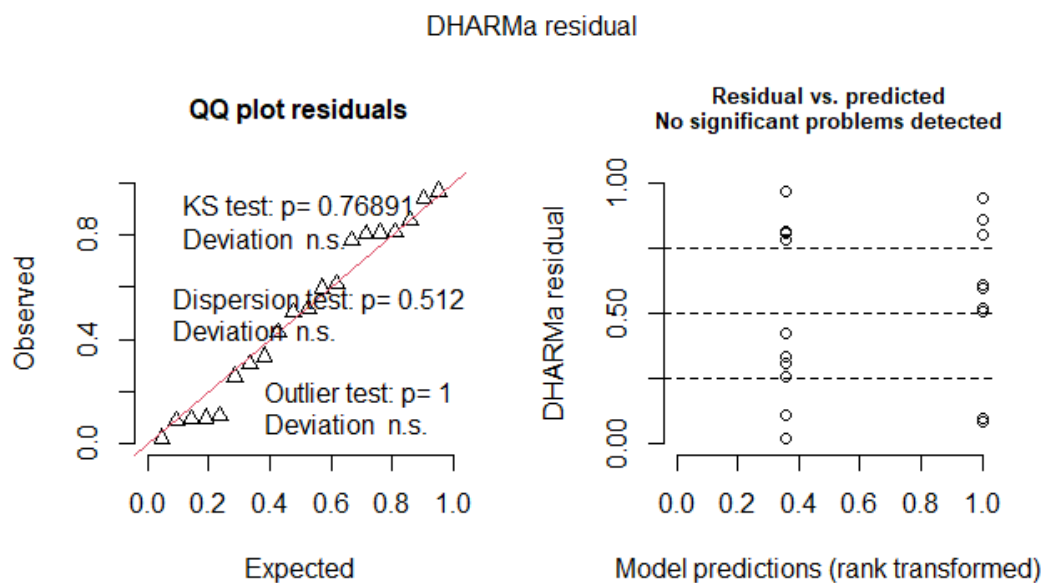
```
## Family: poisson ( log )
## Formula:      Amount_PPP ~ farmtype + (1 | Country)
##
##    AIC    BIC logLik deviance df.resid
##  134.8  137.8  -64.4  128.8     17
##
## Random effects:
##
## Conditional model:
## Groups Name      Variance Std.Dev.
## Country (Intercept) 0.1106  0.3325
##
## Conditional model:
##              Estimate Std. Error z value Pr(>|z|)
## (Intercept)    2.6677    0.1347  19.803  <2e-16
## farmtypeOrganic -0.2364    0.1221  -1.936  0.0529

## Analysis of Deviance Table (Type II Wald chisquare tests)
##
## Response: Amount_PPP
##           Chisq Df Pr(>Chisq)
## farmtype  3.7472  1    0.0529

## $emmeans
## farmtype   emmean   SE df lower.CL upper.CL
## Conventional  2.67 0.135 17    2.38    2.95
## Organic      2.43 0.141 17    2.13    2.73
##
## Results are given on the log (not the response) scale.
## Confidence level used: 0.95
##
```

```
## $contrasts
## contrast      estimate    SE df t.ratio p.value
## Conventional - Organic  0.236 0.122 17  1.936  0.0697
##
## Results are given on the log (not the response) scale.

## farmtype    rate  SE df lower.CL upper.CL
## Conventional 14.4 1.94 17  10.84   19.1
## Organic      11.4 1.60 17   8.45   15.3
##
## Confidence level used: 0.95
## Intervals are back-transformed from the log scale
```

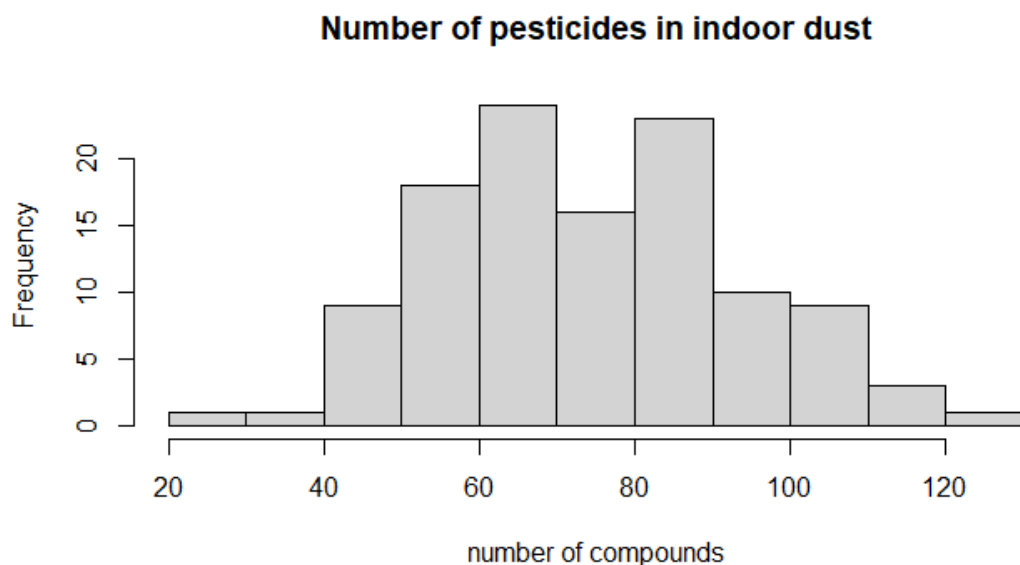


- Indoor dust

Response: number of pesticides in indoor dust

Aim: comparison of conventional and organic farms

Method: GLMM with Poisson distribution and log link



```
## Family: poisson ( log )
## Formula:      Amount_PPP ~ farmtype + (1 | Country/farmtype)
##
##    AIC    BIC logLik deviance df.resid
##  989.1 1000.1 -490.5  981.1    111
##
## Random effects:
##
## Conditional model:
## Groups      Name      Variance Std.Dev.
## farmtype:Country (Intercept) 0.008135 0.09019
## Country      (Intercept) 0.028248 0.16807
##
## Conditional model:
##              Estimate Std. Error z value Pr(>|z|)
## (Intercept)    4.38270   0.06237  70.27 < 2e-16
## farmtypeOrganic -0.18472   0.04697  -3.93 8.39e-05

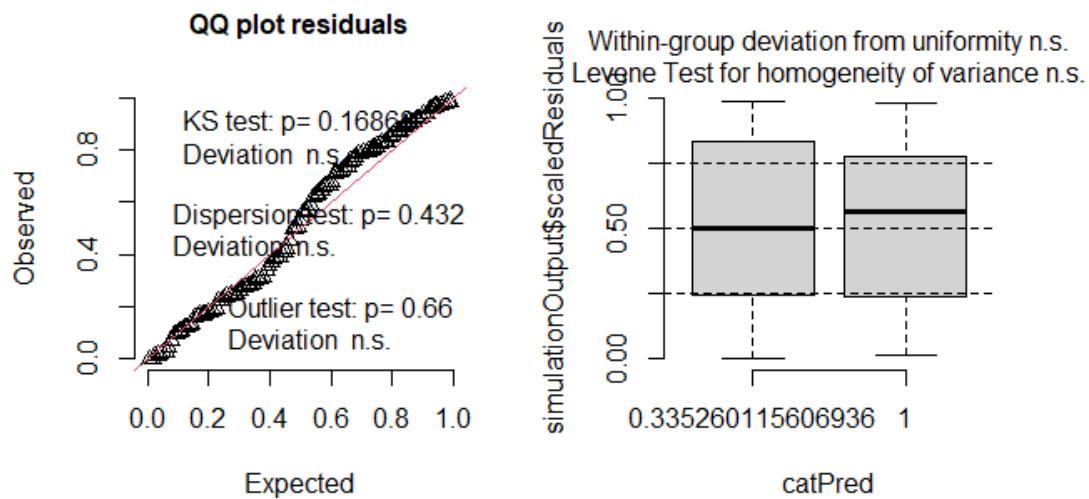
## Analysis of Deviance Table (Type II Wald chisquare tests)
##
## Response: Amount_PPP
##      Chisq Df Pr(>Chisq)
## farmtype 15.467  1 8.395e-05

## $emmeans
## farmtype  emmean   SE df lower.CL upper.CL
## Conventional  4.38 0.0624 111    4.26    4.51
## Organic      4.20 0.0631 111    4.07    4.32
##
## Results are given on the log (not the response) scale.
## Confidence level used: 0.95
```

```
##
## $contrasts
## contrast      estimate    SE df t.ratio p.value
## Conventional - Organic  0.185 0.047 111  3.933  0.0001
##
## Results are given on the log (not the response) scale.

## farmtype    rate  SE df lower.CL upper.CL
## Conventional 80.1 4.99 111   70.7   90.6
## Organic      66.6 4.20 111   58.7   75.4
##
## Confidence level used: 0.95
## Intervals are back-transformed from the log scale
```

DHARMA residual



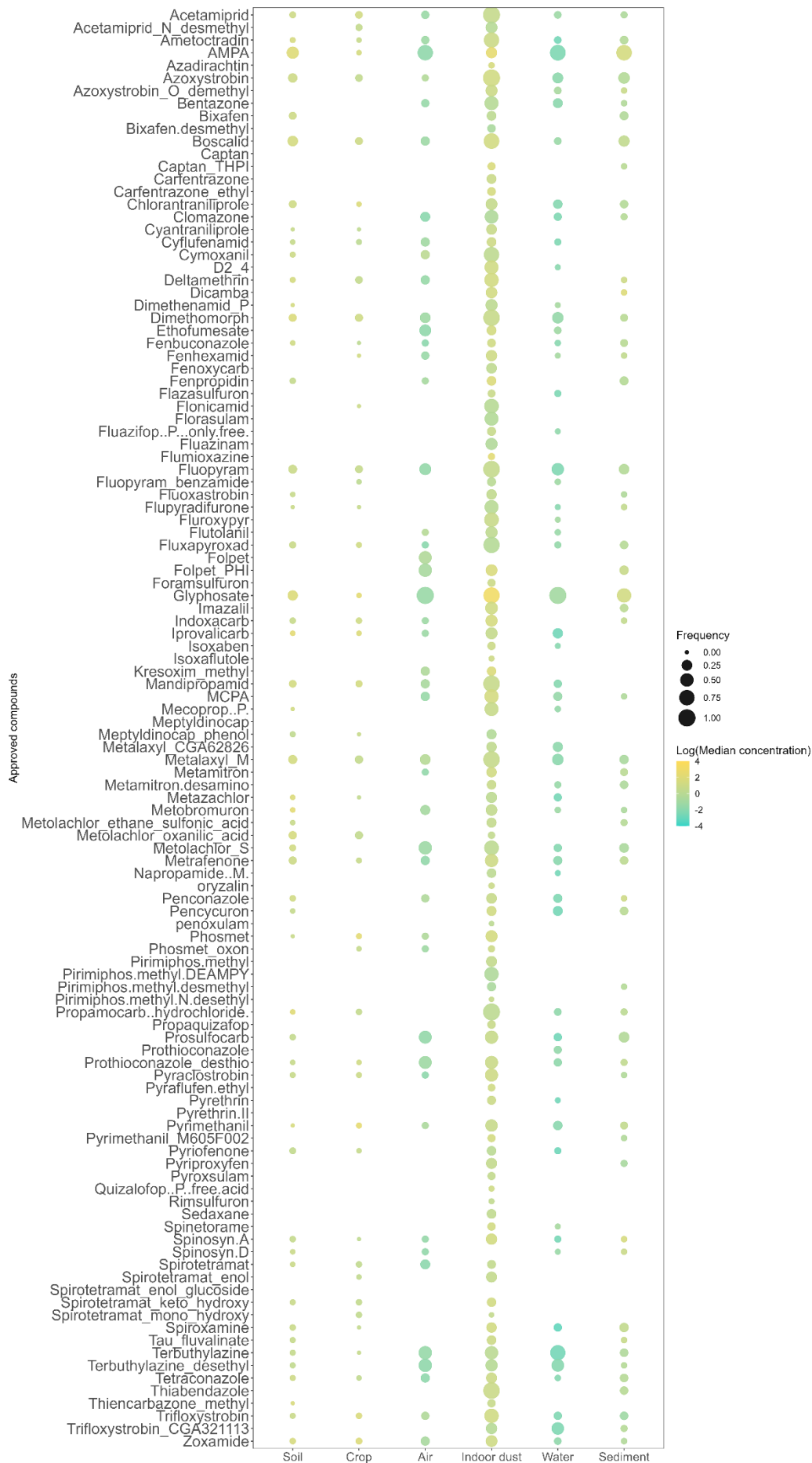


Figure SM1 - Overview of frequency and median levels of approved compounds across soil, crop, water, sediment, outdoor air, and indoor dust samples.

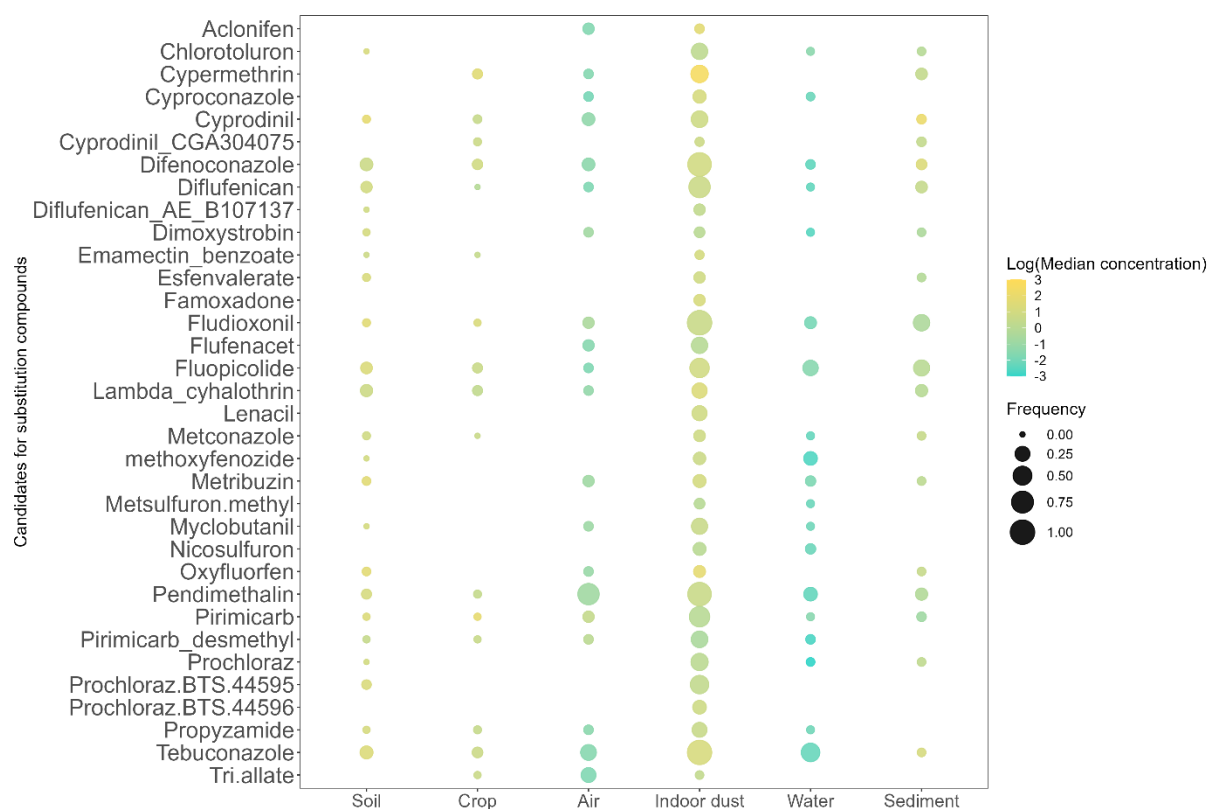


Figure SM2 - Overview of frequency and median levels of candidates for substitution across soil, crop, water, sediment, outdoor air, and indoor dust samples.

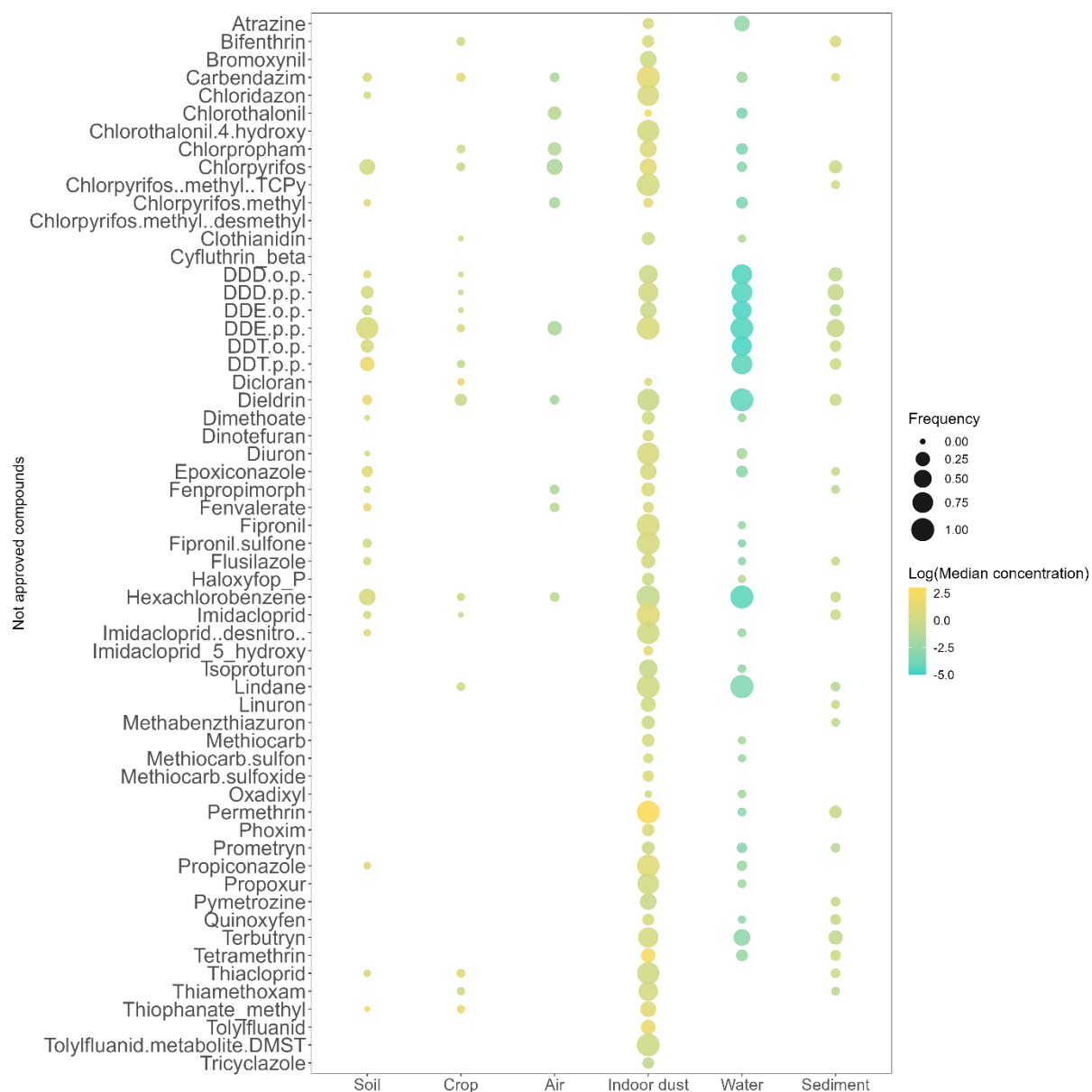


Figure SM3- Overview of frequency and median levels of not approved compounds across soil, crop, water, sediment, outdoor air, and indoor dust samples.

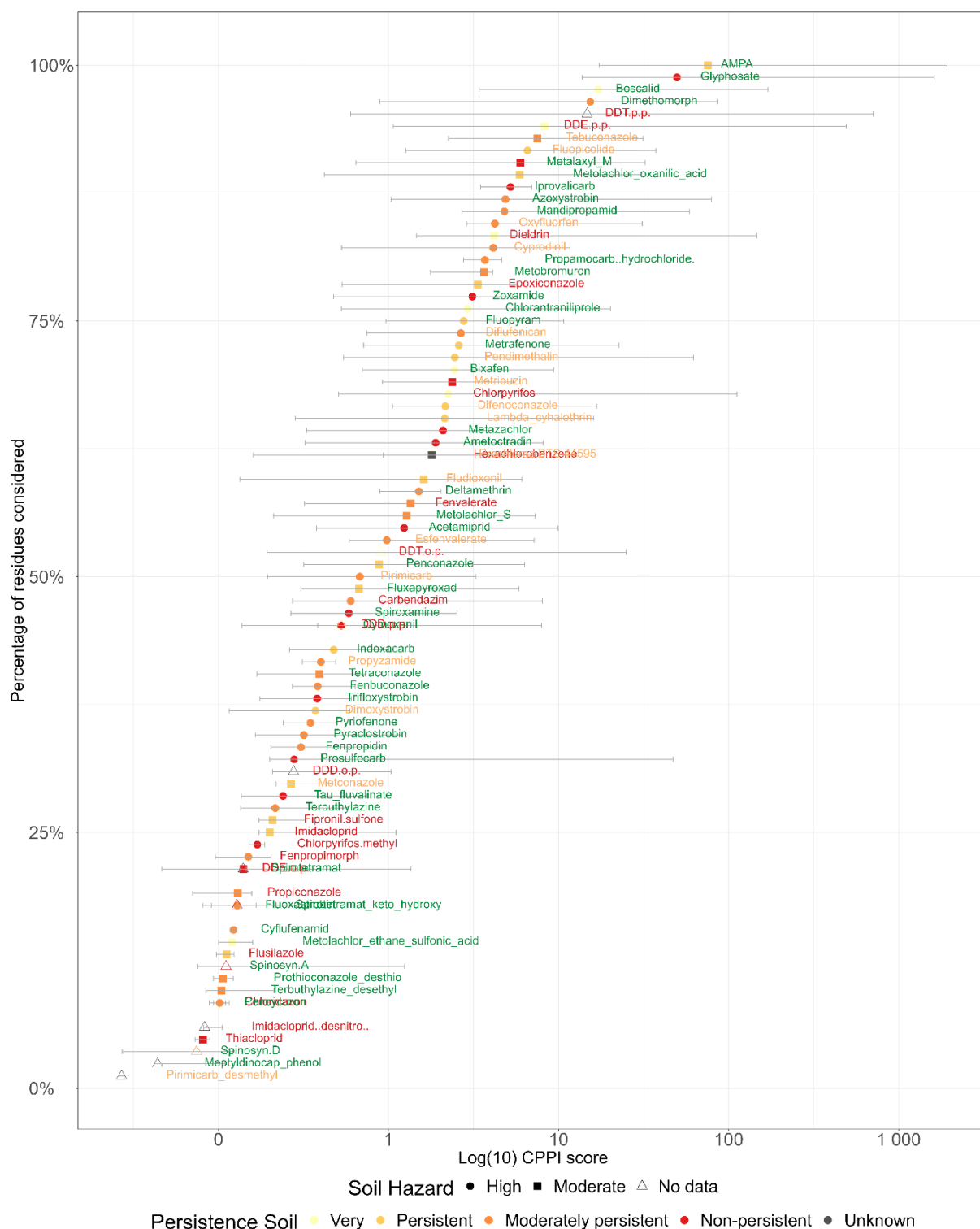
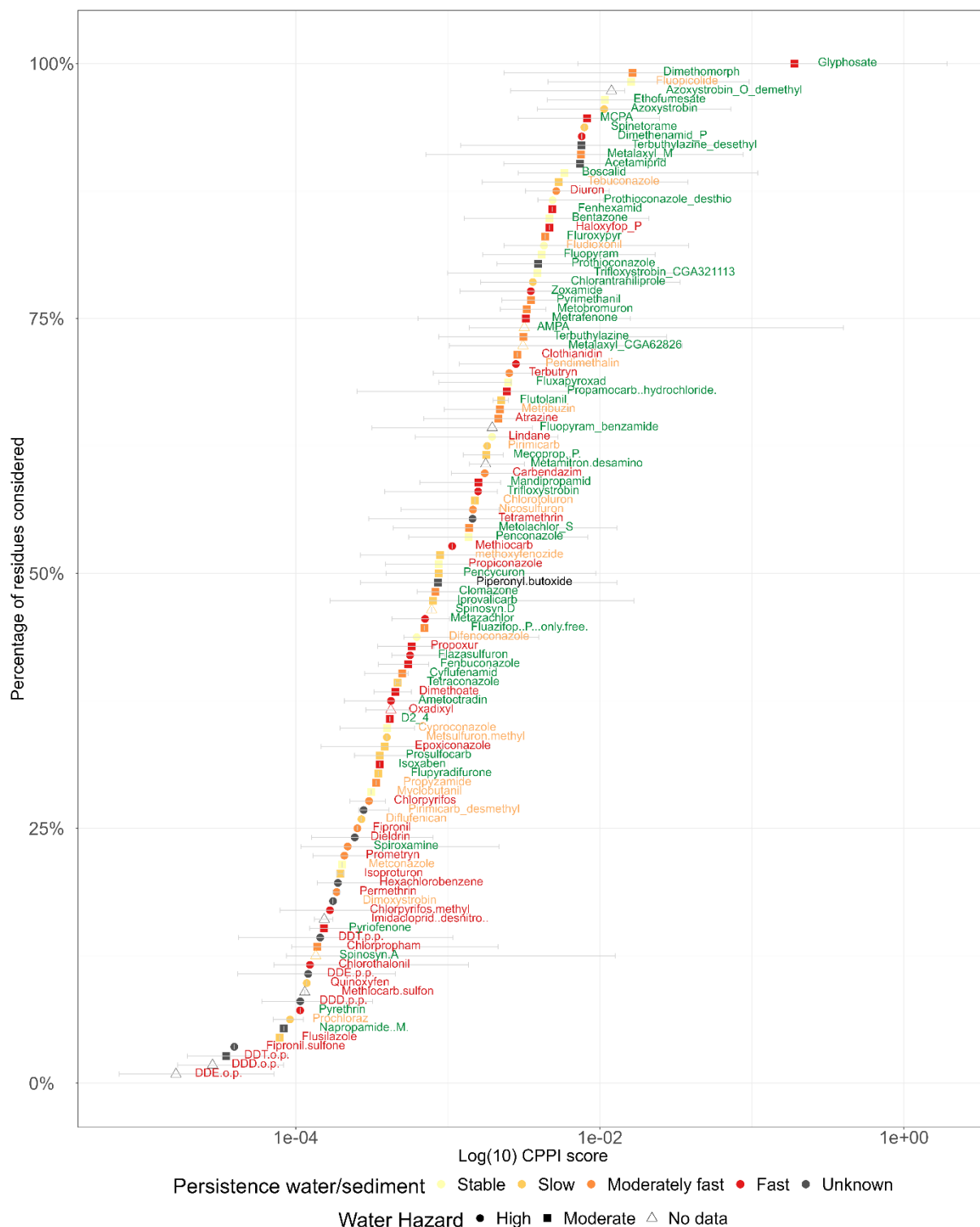
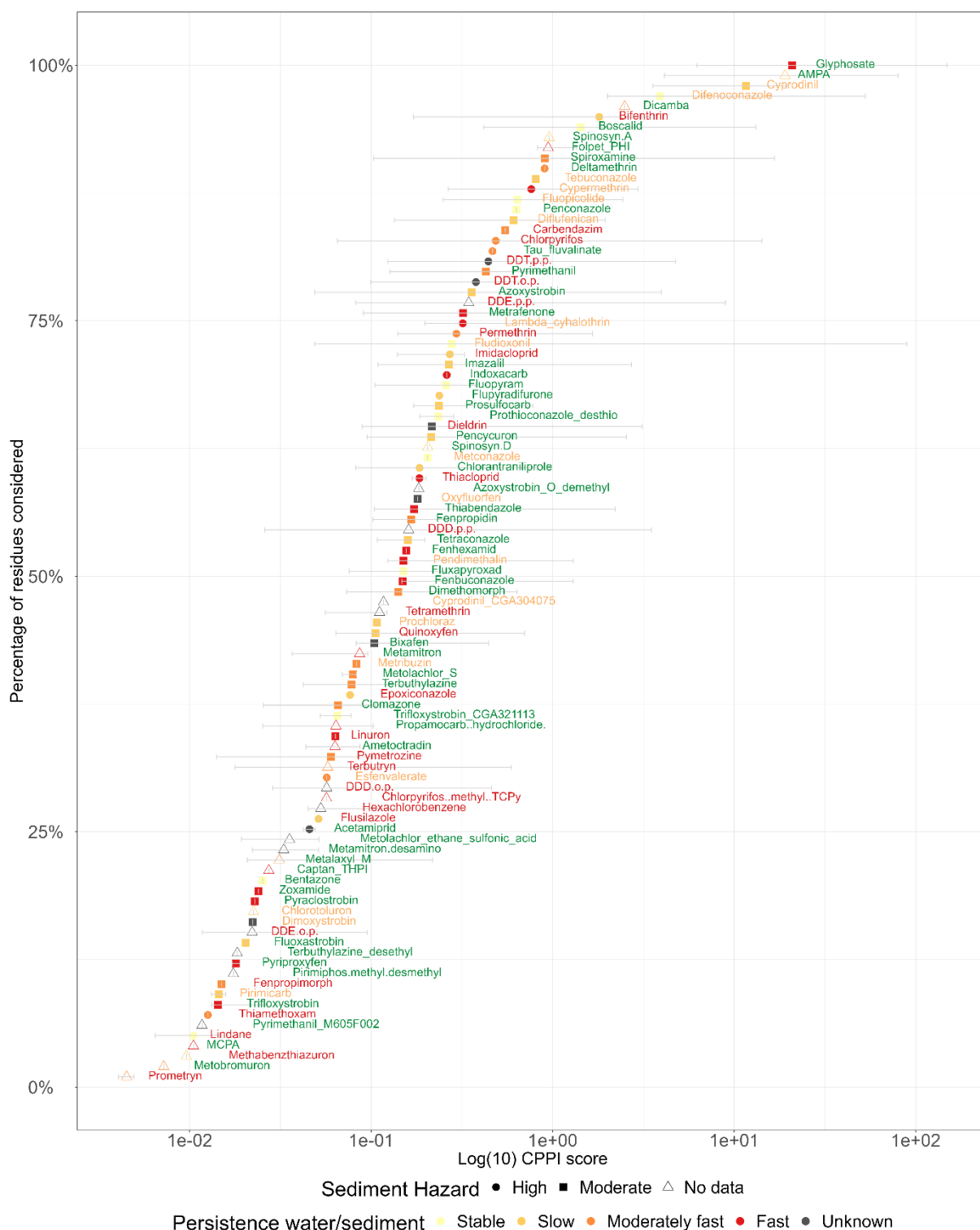


Figure SM4 - Cumulative Pesticide Prioritization Indicator (CPPI) for soil. This curve accounts for the frequency and levels of different pesticide residues in soil and their hazard to soil organisms. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The hazard to soil organisms are shown in different symbol shapes and the persistence of compounds in soils are shown in different colours. The “no data” situation in soil hazard and the “Non-persistent” and “Unknown” situation in soil persistence post a bigger uncertainty to the PPI score.





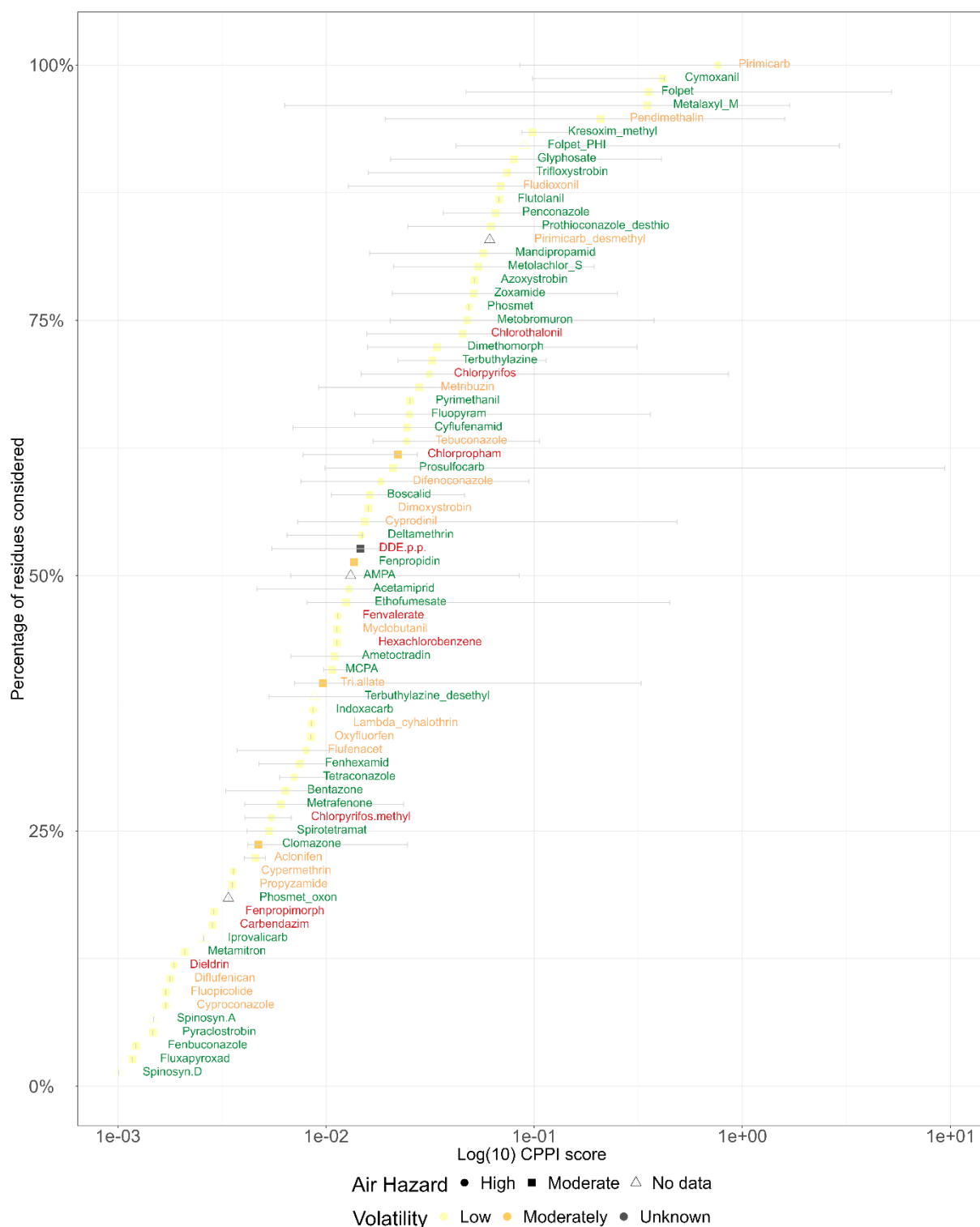


Figure SM7 - Cumulative Pesticide Prioritization Indicator (CPPI) for air and outdoor dust. This curve accounts for the frequency and levels of different pesticide residues in air and outdoor dust and their hazard to aerial organisms. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The hazard to aerial organisms are shown in different symbol shapes and the volatility of compounds are shown in different colours. The “no data” situation in the hazard and the “Unknown” situation in volatility post a bigger uncertainty to the PPI score.

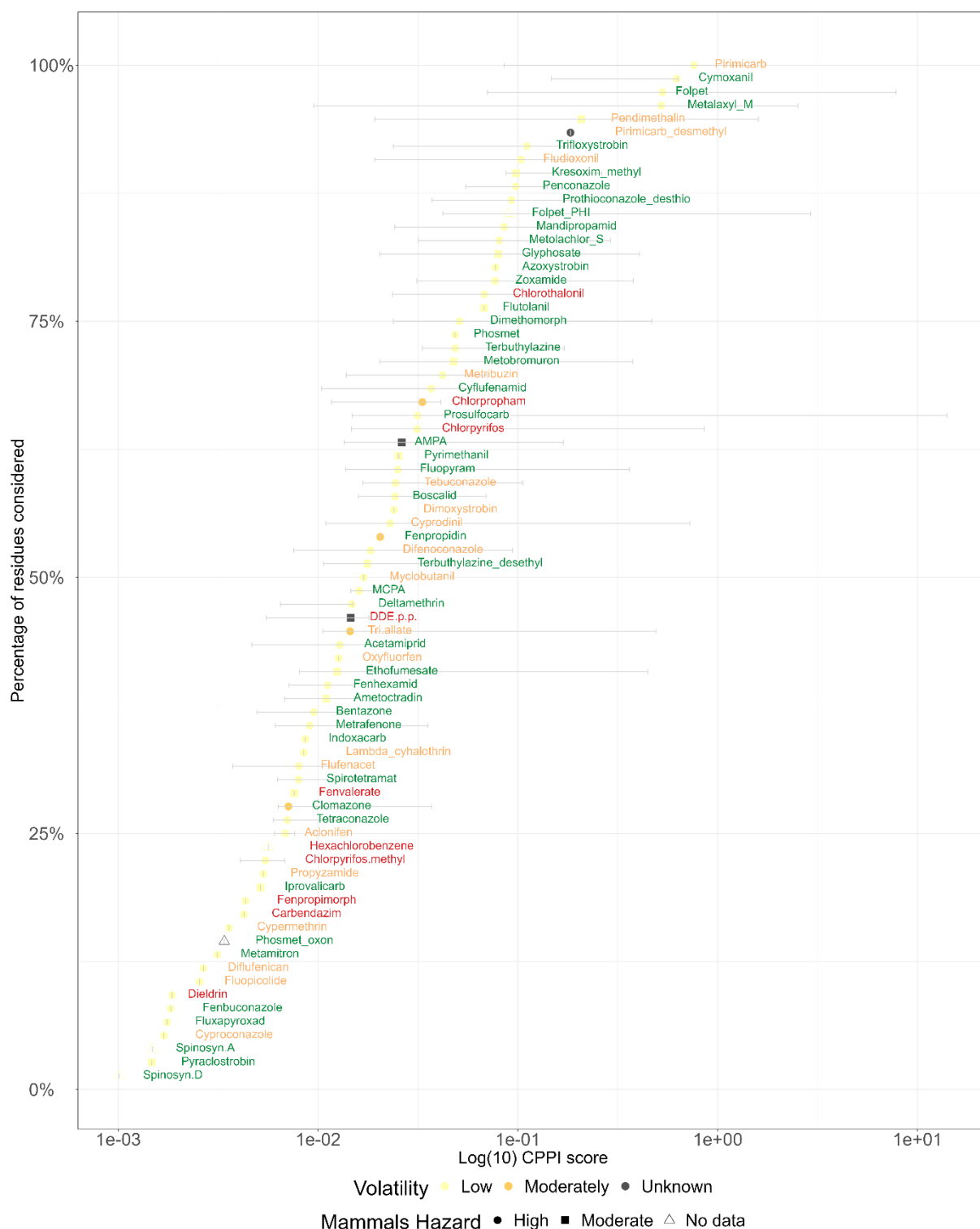
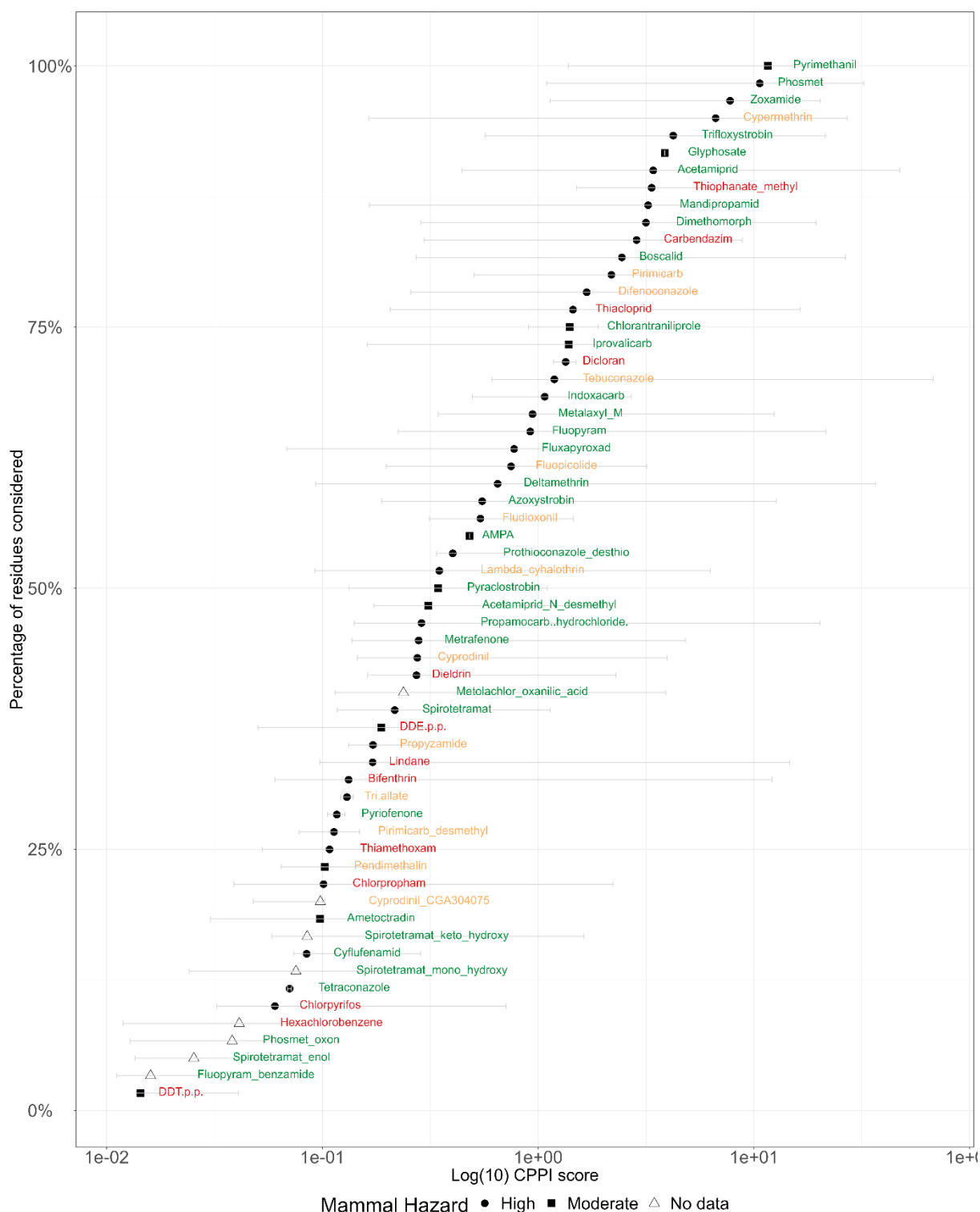


Figure SM8 - Cumulative Pesticide Prioritization Indicator (CPPI) for air and outdoor dust. This curve accounts for the frequency and levels of different pesticide residues in air and outdoor dust and their hazard to mammals. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The hazard to mammals are shown in different symbol shapes and the volatility of compounds are shown in different colours. The “no data” situation in the hazard and the “Unknown” situation in volatility post a bigger uncertainty to the PPI score.



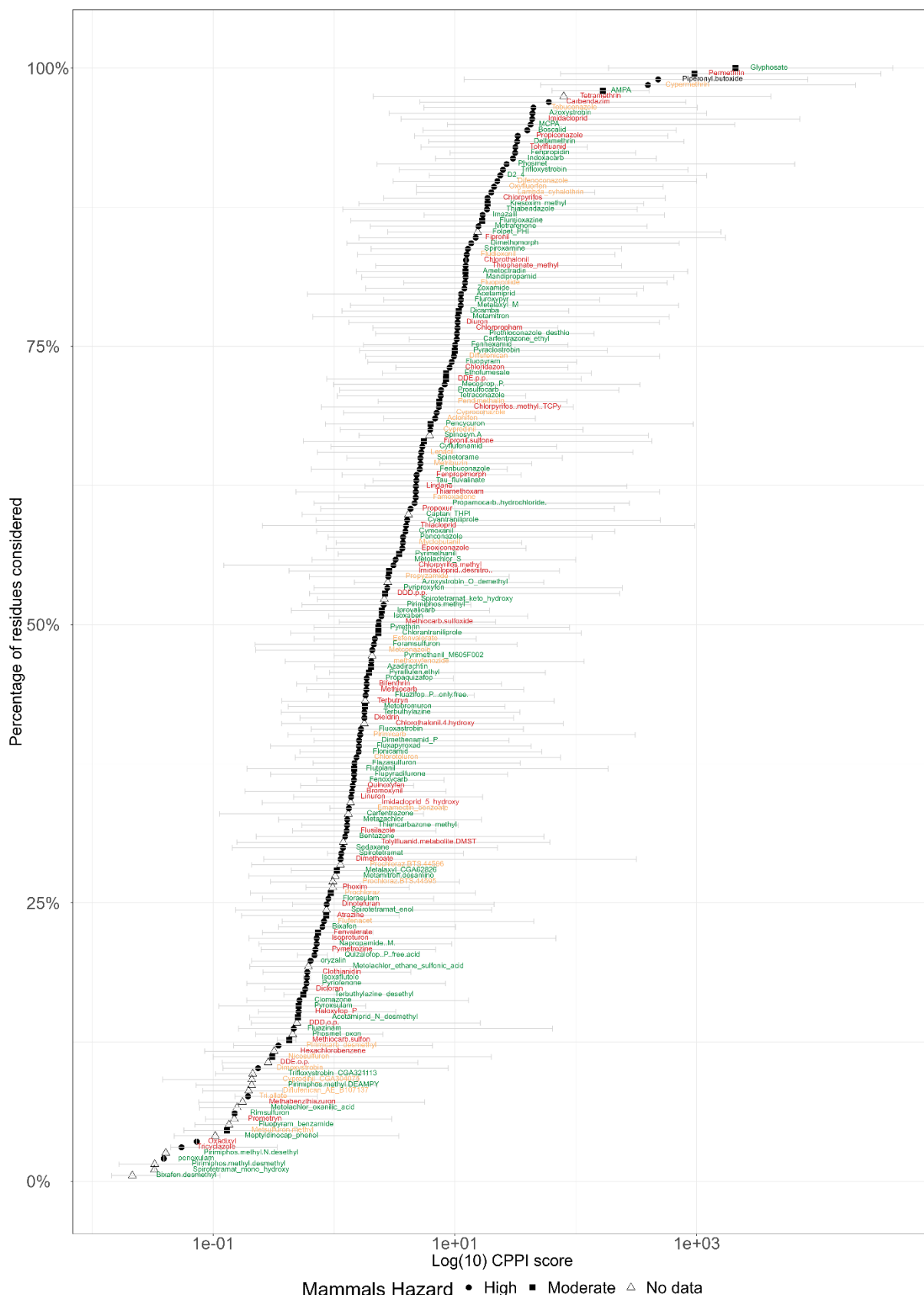


Figure SM10 - Cumulative Pesticide Prioritization Indicator (CPPI) for indoor dust. This curve accounts for the frequency and levels of different pesticide residues in indoor dust and their hazard to mammals. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange denotes

candidates for substitution and green indicates approved compounds. The hazard to mammals are shown in different symbol shapes where the “no data” situation post a bigger uncertainty to the PPI score.

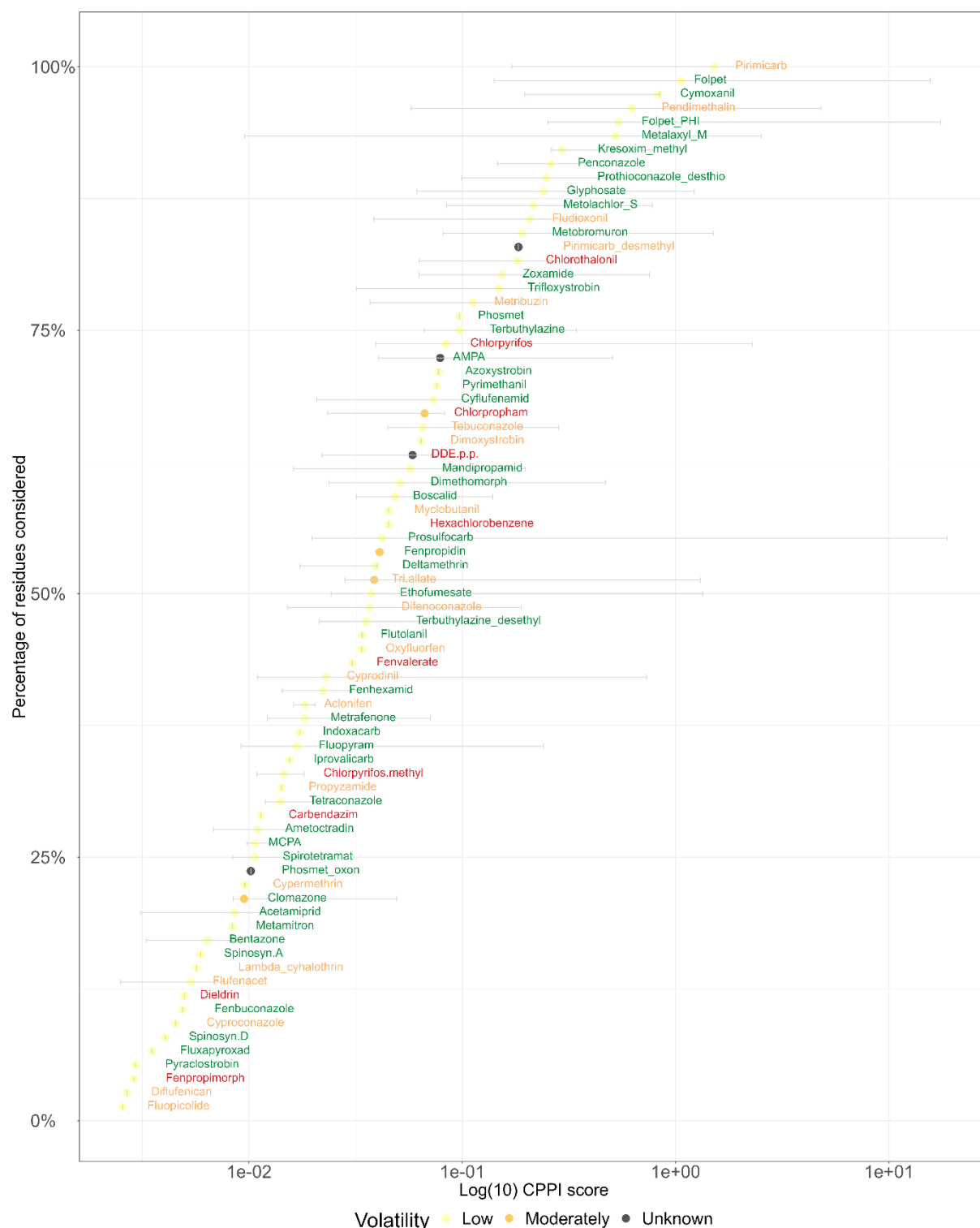


Figure SM11 - Cumulative Pesticide Prioritization Indicator (CPPI) for air and outdoor dust. This curve accounts for the frequency and levels of different pesticide residues in air and outdoor dust and their hazard to human health issues. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The hazard to human health issues are shown in different symbol shapes and the volatility of compounds are shown in different colours. The “no data” situation in the hazard and the “Unknown” situation in volatility post a bigger uncertainty to the PPI score.

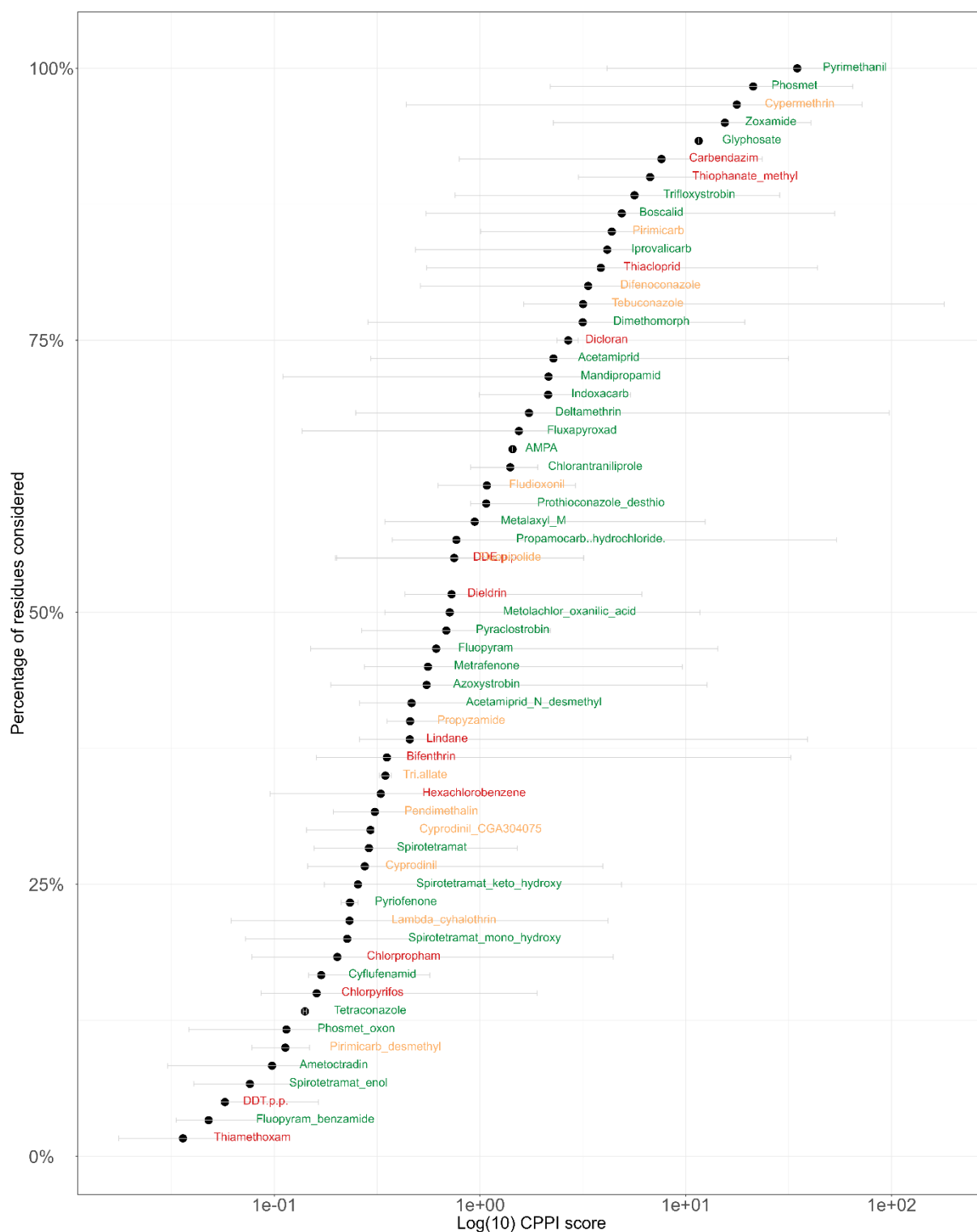


Figure SM12 - Cumulative Pesticide Prioritization Indicator (CPPI) for crops. This curve accounts for the frequency and levels of different pesticide residues in crops and their hazard to human health issues. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange indicates candidates for substitution and green indicates approved compounds. The hazard to human health issues are shown in different symbol shapes where the “no data” situation post a bigger uncertainty to the PPI score.

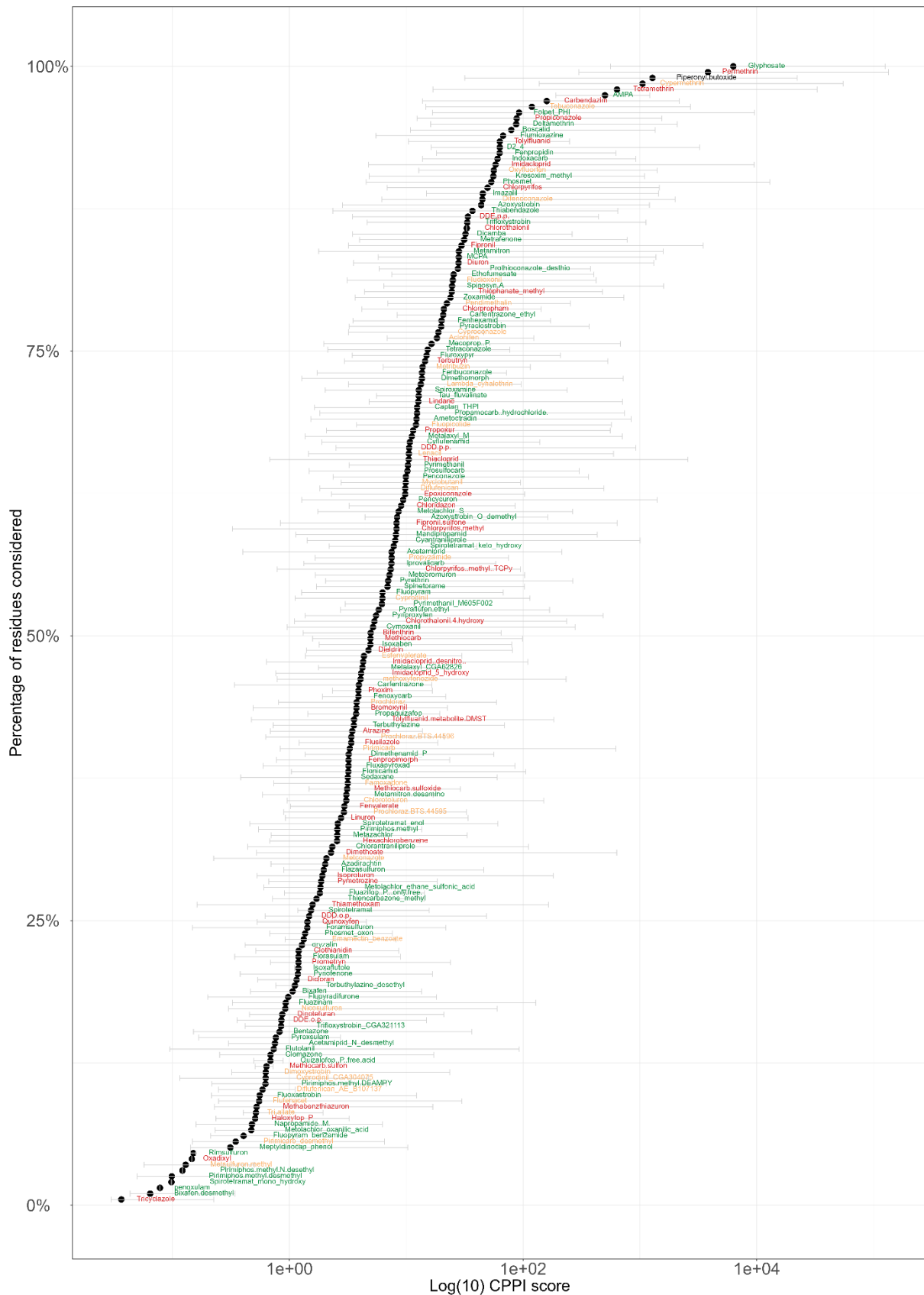


Figure SM13 - Cumulative Pesticide Prioritization Indicator (CPPI) for indoor dust. This curve accounts for the frequency and levels of different pesticide residues in indoor dust and their hazard to human health issues. The grey bars denote uncertainty in the PPI value; the bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration. The name of pesticide residues are presented in different colors according to respective approval status: red indicate not approved compounds, orange

denotes candidates for substitution and green indicates approved compounds. The hazard to human health issues are shown in different symbol shapes where the “no data” situation post a bigger uncertainty to the PPI score.

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