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Investigating the fate and transport behavior of pesticides in air via bioindicator plants and bulk deposition samplers in southeastern Germany

Frank Pompe[1], Marco U. Reitz[2], Björn Brumhard[2], Monica Waeber[1], Annelle A. Mendez Gutierrez[2], Jutta Köhler[3], Uwe Kunkel[4], Klaus Gehring[5,], Markus Scheithauer[6], and David Johnson[7,*]

[1]UMW Umweltmonitoring, Sauerlach, Germany
[2]Syngenta Agro GmbH, Frankfurt am Main, Germany
[3]Bayerisches Landesamt für Umwelt, Referat 76—Stofftestlabor, Umweltmonitoring, Wielenbach, Germany
[4]Bayerisches Landesamt für Umwelt, Referat 75—Spezielle Analytik für Umweltüberwachung, Augsburg, Germany
[5]Bayerische Landesanstalt für Landwirtschaft, Institut für Pflanzenschutz—Herbologie, Freising, Germany
[6]Bayerisches Landesamt für Umwelt, Augsburg, Germany
[7]Jealott's Hill International Research Centre, Syngenta Crop Protection AG, Bracknell, Berkshire, United Kingdom
Frank Pompe and Marco U. Reitz contributed equally to this work.
*Corresponding author: David Johnson, Email: Dave.Johnson@Syngenta.com

Abstract

Plant protection products (PPP) help growers minimize crop losses from pests. For their authorization in Europe, environmental fate and behavior must characterized, including possible transport via air, to assess the risk of potential impacts. Recent interest in airborne PPPs has significantly increased but there is a lack of standardization in measurements, and distinction between long-range transport and local off-target movement is often unclear in publications. In the present study, substance-deposition measurements from two bioindicator plants and bulk-deposition samplers, located at different sites in Bavaria, were used to elucidate aspects of different transport mechanisms. Bulk-deposition data, measured at an agricultural and a background site (≥3 km away from agricultural land) showed frequent pesticide detections. However, detection frequencies and measured concentrations in these samples (i.e., precipitation) were significantly lower at the background site compared with the agricultural site. At the background site, the highest measured deposition of a PPP substance corresponded to 0.007% of its maximum registered field-application rate. At the agricultural site, the highest measured deposition was attributed to local spray drift and represented 0.5% of the maximum field-application rate. Bioindicator plants showed that deposition was predominantly influenced by local agriculture, reflecting the extent of land use for the cultivation of different crops and, hence, the likely magnitude of local pesticide spraying. Observed peak depositions to bulk and bioindicator sampling systems corresponded in time with PPP application to crop fields. There was no clear correlation between the bulk- and bioindicator-plant deposition data sets. When investigating pesticide transport via air, special attention should therefore be placed in the selection of the measurement matrices and methodology to address specific research questions.

Keywords pesticide, volatilization, spray drift, bulk deposition, biomonitoring

Introduction

Plant protection products (PPPs), also referred to as pesticides, are used to repel, destroy, or control pests, weeds, or crop diseases, and they are an important element of integrated pest management, contributing to food security. The benefits of PPP use must be weighed against potential negative impacts for human and animal health and for the environment. In the European Union (EU), the main legal instrument ensuring PPPs on the market are safe is Regulation (EC) 1107/2009 (European Commission [EC], 2009). Within the framework of this regulation, risk assessments are performed considering the highest possible exposure resulting from PPP use according to good agricultural practice for all uses for which authorization is sought. Exposure to a PPP via air is addressed as follows. A toxicological risk assessment for farm workers, bystanders, and residents is performed considering potential inhalation exposure of PPP active substances (European Food Safety Authority [EFSA], 2022), assuming conservative default, or worst-case air concentrations. In addition, environmental risk assessment considers PPP entry

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to edge-of-field water bodies via spray drift for all substances and volatilization from soil and plant surfaces for substances with a vapor pressure (V_P) of 10^{-5} Pa (25 °C) or greater. Furthermore, according to EU Regulation, active substances in PPPs must be intrinsically degradable in the atmosphere, with atmospheric half-lives (gas-phase OH-radical-, or ozone-initiated photooxidation) less than 2 days (FOCUS, 2008; United Nations Environment Programme, 2001).

Off-field movement of PPPs in air can potentially occur: (a) during application via spray drift, or vapor drift following spray-droplet drying and/or (b) post-application via volatilization from surfaces or wind erosion of spray-exposed soil. Based on field study data, the magnitude of PPP deposited as spray is anticipated to be $<5\%$ of the field application rate at distances of 10 to 20 m, and $\leq 0.1\%$ at distances ≥ 100 m. The extent of spray drift is dependent on crop type and the spray method used (Holterman et al., 2017; Rautmann et al., 2001; Van de Zande et al., 2015). Spray drift can be minimized with low-drift spray nozzles, anti-drift technologies such as Pusle-Width-Modulation systems and nets, and by only spraying when weather conditions are appropriate. Substances deposited on soil and plant surfaces after application can volatilize and undergo further drift and deposition, the extent of which also decreases with increasing distance from the field. For example, wind-tunnel studies have shown that the amount of vapor deposited 20 m from the field is approximately 35% of that at 1 m, for substances with $V_P > 5 \times 10^{-3}$ Pa (Fent, 2004).

There is increasing interest in the presence of pesticides in air, as shown in a recent literature review, which identified 77 studies published in the scientific literature between 2002 and 2022, with more than 6.7 studies published per year since 2020 (Brüggermann et al., 2024). Pesticide monitoring in air commissioned by public institutions at the national or sub-national level within the EU has also increased in the last decade. The French network of Approved Air Quality Monitoring Associations has monitored pesticides in air since 2002 (Atmo France [Federation of Approved Air Quality Monitoring Associations], 2025). It performed a measurement campaign using a harmonized methodology from June 2018 to June 2019, targeting 75 active substances in airborne particulate matter and in the gas phase, measured on a weekly basis at 50 rural, urban, and semi-urban sites, located at least 200 m from agricultural plots (ANSES, 2020). The Walloon region of Belgium conducted their Exposition de la Population aux Pesticides Environnementaux (EXPOPESTEN) study from May 2015 to May 2016 focusing on 46 PPPs in the gas and particulate phase at 12 sites (including five near agricultural land, urban, and background sites; ISSEP, 2018). Another study was conducted in Belgium in 2018 to monitor PPP active substances in air (gas and particulate phase and drift deposits) during 2–48 hr after application at or near sties of PPP use (ISSEP, 2019). Risk assessments considering exposure via inhalation concluded that pesticide levels measured in the French and Belgian monitoring studies do not represent a health risk for humans (ANSES, 2020; ISSEP, 2018; Vinck et al., 2024). The German Federal Office of Consumer Protection and Food Safety (BVL) also performed a pilot study that monitored PPPs in air in 2023–2024 at three sites (two with agricultural influence), in preparation for a future nationwide atmospheric transport monitoring (BVL, 2025).

In addition to direct measurements in air, atmospheric deposition of PPPs has been monitored throughout Europe, with different sampling durations. National monitoring of PPPs in precipitation was conducted from 2002 to 2022 in Sweden at rural background sites located more than 1 km away from agricultural fields, using bulk samplers collecting 12 to 15 samples during the main growing season from April to October (Adielsson & Krueger, 2007; Kreuger & Lindström, 2019; SLU, 2024). Current used pesticides were detected at ng/L levels in rainwater, with occasional findings at μg/L levels (Kreuger & Lindström, 2019). The Danish Centre for Environment and Energy (DCE) monitored 12 pesticides and five degradation products in rainwater during 2-month collection periods at two locations since 2004 (Asman et al., 2005; DCE, 2021). The maximum wet deposition for individual substances observed during the 2-month collection periods ranged from 0.2 μg/m^2 (January–February) to 71 μg/m^2 (September–October). Bulk deposition of PPPs was also measured in pilot studies in Switzerland (Schläpfer et al., 2021) and in Germany (BVL, 2025). The bulk-deposition data from the German study, consisting of samples collected for 4 weeks, indicated no exceedances of regulatory acceptable concentrations for non-target aquatic organisms (BVL, 2025).

Furthermore, atmospheric deposition of PPPs has been investigated using natural vegetation samples, including alpine valley flora (Brühl et al., 2024), playground grass (Linhart et al., 2019), and pine needles (Ratola et al., 2014). In 53 vegetation samples collected along an alpine valley in a large apple-growing area of Italy, 18 current use pesticides (CUP) were detected with maximum substance concentrations ranging from below the limit of quantification (<LOQ) to 1.34 mg/kg (median maximum concentration was 0.012 mg/kg; Brühl et al., 2024). Current use pesticide findings and concentrations in the alpine vegetation samples decreased with increasing distance from orchards and increasing altitudes. In northern Italy, 12 pesticides were detected at 0.035 to 2.024 mg/kg in grass samples collected from public playgrounds located within 50 m from apple orchards and vineyards (Linhart et al., 2019). In pine need samples collected from 12 locations in Portugal, 18 PPP active substances were detected, with total concentrations ranging from 0.019 to 0.056 mg/kg (dry wt basis; Ratola et al., 2014). In a study of similar design to Brühl et al. (2024), levels of 93 CUPs were measured in vegetation samples (grass, herbs and leaves of shrubs, no specific species mentioned) collected in the Upper Rhine Valley of Germany (Mauser et al., 2025). The mean total pesticide concentration measured in vegetation samples was 0.051 mg/kg (dry wt basis), and the single maximum value was 1.97 mg/kg in a sample collected within 10 m from a vineyard. Lower pesticide concentrations were measured in samples collected further away from sites of agricultural activity.

In the EU, pesticide residues on crops for food or feed must be below regulatory maximum residue levels (MRL). If an EU approved pesticide is not authorized to treat a particular crop, a default MRL of 0.01 mg/kg applies (EC, 2005). In Germany, occasional residues of prosulfocarb and pendimethalin on non-target crops have raised concerns in the past. Therefore, since 2016, the BVL requires spray application of PPPs containing these substances with devices achieving ≥90% drift reduction, at tractor driving speeds ≤7.5 km/hr, and at wind speeds ≤3 m/s

(BVL, 2016). In 2019, the Bavarian Environmental Agency (LfU) published a report on biomonitoring of persistent organic and inorganic pollutants, including PPPs, presenting results of measurements from bulk-deposition samplers and bioindicator plants at "agricultural" and "background" sites (LfU, 2019). Nearly 80 PPP substances were detected in bulk precipitation samples, but only pendimethalin and terbuthylazine (and one of its metabolites) were detected in bioindicator plants.

To gain further insight into factors influencing the off-field transport and atmospheric deposition of PPPs, a new study was conducted between 2018 and 2022 in Germany, combining measurements of bulk deposition (collected precipitation) and residues in bioindicator plants at several locations. This study aimed to elucidate spatial and temporal patterns of pesticide residues in environmental samples, and their correlation with surrounding agricultural land use to better understand PPP transport via air. Two bioindicator plant measurement systems were compared, and relationships between residues in precipitation and bioindicator plants were investigated with a view to the potential standardization of pesticide air monitoring protocols.

Materials and methods

Bulk deposition samplers

Bulk deposition samplers comprise a collector bottle maintained at 6 to 10 °C with an open funnel inlet; thus, PPPs can collect in the bottle via wet deposition (i.e., substance dissolved in rain) plus any dry-deposited material that is washed from the surface of the collector funnel by rain. Sampling of bulk deposition was conducted in 4-week (27–29-day) collection periods from mid-March until mid-December in 2018 to 2020 and throughout the whole year in 2021 and 2022 as part of a nationwide monitoring program for inorganic and organic pollutants across Bavaria conducted by the LfU. For this study, bulk deposition sampling was carried out at the two following sites: Neusling (NEU) in Lower Bavaria (48.693904 N, 12.875140 E) and Weibersbrunn (WBB) in northwestern Bavaria (49.904187 N, 9.447742 E). The WBB site is surrounded by forest for a radius of approximately 3 km and is approximately 280 km northwest of NEU. No results are available for September and October 2018 for WBB due to problems with the samples. Please see online supplementary material for a map indicating the locations of the measurement stations and a photograph of the bulk deposition sampler (Figures S3 and S4).

Bioindicator plant samplers

Deposition of pesticide substances was investigated via biomonitoring (i.e., the use of living bioindicator plants that can accumulate gas and particulate-phase material from the air). In the present study, Welsh ryegrass (*Lolium multiflorum* Lam. ssp. *italicum* variety "Gemini"), and Mangold chard (*Beta vulgaris* L. var. *vulgaris* variety "Lucullus") were used in parallel as biomonitoring systems, following guidelines Verein Deutscher Ingenieure (VDI) 3957/2 (VDI, 2020) and VDI 3957/4 (VDI, 2024), respectively. Chard was selected as an additional sampling substrate for comparison to ryegrass, as it is more representative of leafy crops that are cultivated as food. Biomonitoring periods of approximately 4 weeks (26–30 days) were used to sample pesticide deposition from April until October in 2020, 2021, and 2022. Bioindicator-sampling rotation throughout the total measurement period was conducted so that successive sampling periods overlapped by 2 weeks (i.e., 2 weeks into a given 4-week sampling period, the next 4-week sampling period was started). Ryegrass was pre-cultivated under standard greenhouse conditions until ready for exposure sampling, being placed 1.5 m above ground in self-watering containers of 20 cm diameter (see online supplementary material Figure S1). At the end of each 4-week sampling period, the grass was cut. The sampled grass cultures were discarded and a new set exposed. The chard plants were placed in two-level stacked containers (60 × 40 × 22 cm). The upper level was planted with chard (15–20 plants), the lower contained water, and the container stack was supported at a height 1.2 m above ground (see online supplementary material Figure S2). At sampling, the older, outer leaves formed during the 4-week exposure period were harvested, leaving the inner leaves to continue to grow. Collected ryegrass and chard samples were packed in airtight aluminum foil and polyethene bags and transported to the laboratory at <8 °C. Samples were stored frozen at −18 °C and analyzed within 2 weeks after harvest. Bioindicator plants were placed at three sites in agricultural areas in southeast Bavaria (see online supplementary material Figure S4): Neusling (NEU; 48.693904 N, 12.875140 E), Uttenkofen (UTK; 48.814918 N, 12.843189), and Aldersbach (ALR; 48.597127 N, 13.099351 E).

Analysis and selection of substances

Volumes of precipitation collected in the bulk samplers were determined gravimetrically, stored refrigerated (<5 °C), and analyzed within 6 months of collection. For analysis, 10-ml aliquots were transferred into glass high-performance liquid chromatography vials, isotopically labeled internal standards were added to achieve a concentration of 50 ng/L, and the contents were mixed thoroughly. Active substances were analyzed using an Ultimate3000 ultrahigh-performance liquid chromatography system connected to a Q Exactive Orbitrap mass spectrometer (both apparatus from Thermo Scientific) equipped with a heated electrospray ionization source (H-ESI). Samples were enriched online using a C18 solid-phase extraction (SPE) method. Chromatographic separation was achieved using a binary gradient of 0.1% formic acid and acetonitrile on a Zorbax Eclipse plus C18 column (150 × 2.1 mm, 3.5 µm particle size, Agilent). The mobile phase flow rate was 0.3 ml/min, and the column oven temperature was maintained at 35 °C. Samples of 5 ml volume, extracted with the online SPE system, resulted in a calibration range of 1 to 250 ng/L, and an LOQ of 2 ng/L for most analytes. All samples were analyzed in positive and negative ionization mode, in separate runs. Resolution of full scan was set to 140,000, and the range was 100–1,050 *m/z*. Product-ion spectra in data-dependent acquisition mode were collected at a resolution of 17,500, applying stepped collision energies of 20, 40, and 60, and a fixed first mass of 50 Da. In total, 135 active substances and metabolites were determined in precipitation during the time span of 2018–2022 (see online supplementary material Table S1).

Qualification of pesticides was achieved using internal calibration with isotopic labeled standards. Quality (r^2) of weighted linear regression ($1/x$) was >0.95 for all substances. During each analytical run, blank measurements and spike experiments of deposition samples with 10 ng/L and 100 ng/L as quality assurance measures were conducted. For most substances, the recovery rates were in the range of 80% to 120% (90% of substances for spike experiments with 10 ng/L and 93% of substances for 100 ng/L). Final concentrations of pesticides in deposition samples were then calculated with determined recovery rates during each analytical run (for details, see online supplementary material Table S2).

In previous work published by the LfU (2019), it was shown that the pesticide substance concentration in precipitation samples was independent of the magnitude of rainfall. Therefore, the active substance bulk deposition data generated in the present study are presented as time-averaged deposition rates (ng/m^2/day) for each 4-week sampling period.

The analysis of pesticide substances in the biomonitoring samples was performed by an accredited laboratory (DIN EN ISO 17025) using a standard multi-residue method *(Modular QuEChERS method* DIN EN 15662:2018-07). The bioindicator-plant samples were analyzed for multiple substances (∼660 in total, mostly PPP active substances, plus several metabolites), with a standard LOQ of 0.01 mg/kg plant sample (fresh wt). For 17 active substances and the metabolite desethyl-terbuthylazine, selected based on previous data generated by the LfU (2019), additional liquid chromatography tandem mass spectrometry analyses (with spiked sample extracts) were performed to achieve an analysis LOQ of 0.001 mg/kg. These substances comprised the fungicides boscalid, difenoconazole, dimethomorph, epoxiconazole, propamocarb, propiconazole, spiroxamine, tebuconazole, the herbicides ethofumesate, isoproturon, metamitron, S-metolachlor, nicosulfuron, pendimethalin, prosulfocarb (and its sulfoxide metabolite), and terbuthylazine (and its desethyl metabolite), and the biocide carbendazim.

Land use data

For 2020, 2021, and 2022, land use around NEU, UTK, and ALR was characterized at various radial distances (up to 100 m, 100 to 500 m, 500 to 1,000 m, and 1,000 to 1,500 m) regarding the cultivation of different crops (see online supplementary material Table S4 and Figure S5a). Land-use data, based on the central National Integrated Administration and Control System database (Zentral InVeKos Datenbank [ZID], 2024), were provided by the Bavarian State Research Centre for Agriculture (LfL) in an anonymized form. The following land use classifications were used: legumes, fodder grass, cereals, potato, maize, sugar beet, oil seeds, vegetables, permanent grassland, set-aside land, and no agricultural use. For WBB, land use data were provided for a radius of 15 km around the measuring site (see online supplementary material Figure S5b).

Meteorological measurement data

The measurement stations at ALR, NEU, UTK, and WBB are part of the Bavarian agrometeorological measuring network (LfL, 2024). In the present study, daily values for precipitation, mean temperature, relative humidity, and mean and maximum wind speed were obtained for the overlapping 4-week sampling periods.

Active substance approval status, typical use patterns, and intrinsic substance properties

Information regarding registered PPPs in Germany containing active substances detected during the study are provided in Table 1 and in the online supplementary Table S3. The PPP application details (target crops, amounts, and timings) were taken from EFSA publications (Pesticide Peer Review Reports or Reasoned Opinions) or the BVL website (BVL, 2025a). Please see online supplementary material for indicators of potential volatility from plant and soil surfaces (i.e., see columns 2 and 3 of Table S3), based on equilibrium partitioning considerations (as explained in the online supplementary material Box S1). In addition, sales data from 2018 to 2022 for focal active substances are presented in Table 1.

Results and discussion

Bulk-deposition measurements

Annual distributions of the average daily deposition rates (ng/m^2/day) of the 20 substances measured most frequently above the LOQ at the NEU ("agricultural") and/or WBB ("background") sites are represented in Figure 1 (abbreviations are from Table 1). See online supplementary material for time-series plots (Figures S8 and S9). As stated above, the two sites are approximately 280 km away from each other and selected to be contrasting representative locations.

Three main findings can be discerned from the box-and-whisker plots. First, the 10 highest overall substance deposition rates measured at NEU were for prosulfocarb and its sulfoxide metabolite, terbuthylazine and its desethyl metabolite, S-metolachlor, prothioconazole desthio (i.e., metabolite of prothioconazole), flufenacet, epoxiconazole, pendimethalin, and dimethenamid, with mean deposition rates of 1742.5, 587.9, 201.3, 71.09, 196.3, 54, 17.9, 21, 15.0, and 53.6 ng/m^2/day, respectively (considering all periods in which the substances were measured). All these substances were sold in large quantities in Germany (>200 tons/year). The three substances with the highest mean deposition rates—prosulfocarb, terbuthylazine and S-metolachlor—were registered for use on crops grown close (within 1.5 km) to the site during the study period (see online supplementary material Table S3 and Figure S5a), have non-negligible volatilization potential (see online supplementary material Table S1), and were usually applied to fields at rates of 500 g/ha or more (see Table 1). Among the substances with highest deposition rates, only epoxiconazole lost its registration in Germany during the study period, with a PPP stockpile use-up

Table 1 Information on the 20 most frequently observed substances in bulk samplers at the Neusling (NEU) and Weibersbrunn (WBB) sites. The order of the substances is based on average sales data for Germany from 2018 to 2022 (considering only years when there were sales). Further details about plant protection product (PPP) application details are provided in the online supplementary information (Table S1).

Substance[a]	Type	Mean annual PPP sales 2018–2022 (ton)[a]	Vapor pressure (Pa)[b,c]	Water solubility (mg/L)[b]	Half-life in air (days)[c]	Pesticide application details
Metamitron (MMT)[d]	Herbicide	1,258.3	7.4×10^{-7}	1,770	0.55	Spring use on sugar beets up to 3 × 1,392 g/ha.
Prosulfocarb (PSC)[d,e]	Herbicide	1,255.5	7.9×10^{-4}	13.2	0.33	Spring use on potato, legumes, and oilseeds. Autumn uses on cereals. Application up to 4,000 g/ha.
Terbuthylazine (TBA)[d,e]	Herbicide	920.8	1.5×10^{-4}	6.6	1.13	Spring/early summer use on maize up to 750 g/ha.
Flufenacet (FFT)[d,e]	Herbicide	779.6	9.0×10^{-5}	51	0.61	Spring/early summer uses on potato and maize. Autumn use on cereals. Application up to 600 g/ha.
Tebuconazole (TBCZ)[d,e]	Fungicide	750.6	1.3×10^{-6}	36	0.93	Late spring/summer/early autumn uses on beans, cereals, oilseed rape, sunflower and vegetables. Application up to 3 × 300 g/ha.
Pendimethalin (PDM)[d]	Herbicide	677.6	3.3×10^{-3}	0.33	0.35	Spring/early summer uses on bean/pea, vegetables, sunflower, and maize. Late summer/autumn uses on oilseed rape, and cereals. Application up to 2,000 g/ha.
Dimethenamid-P (DMD)[d]	Herbicide	627.3	3.7×10^{-4}	1,200	0.20	Spring uses on legumes, maize, sugar beets, and vegetables. Late summer/autumn use on oilseed rape. Application up to 1,010 g/ha.
Prothioconazole (PTCZ)[f]	Fungicide	625.7	7.4×10^{-10}	22.5	0.095	Summer uses on sugar beet and vegetables. Spring/early summer uses on cereals and oilseed rape. Application up to 3 × 200 g/ha.
S-metolachlor (SMOC)[d,e]	Herbicide	594.9	3.7×10^{-3}	480	0.19	Spring use on maize up to 1,920 g/ha.
MCPA[d]	Herbicide	473.6	4.0×10^{-4}	250,000	0.85	Various crop (cereals, hops) and non-crop (grassland/pasture/meadow, ornamental plants) uses.
Metazachlor (MTZC)[e]	Herbicide	398.2	8.9×10^{-5}	450	0.18	Spring and autumn use on oilseed rape up to 1,000 g/ha.
Azoxystrobin (AZT)[d,e]	Fungicide	288.0	1.1×10^{-10}	6.7	0.23	Late winter/spring uses on oilseed rape and vegetables. Summer/early autumn uses on bean/pea, and vegetables. Application up to 4 × 250 g/ha.
Spiroxamine (SPX)[e]	Fungicide	281.6	5.0×10^{-3}	405	0.083	Late spring/early summer use on cereals up to 2 × 375 g/ha.
Propyzamide (PPYZ)[e]	Herbicide	224.5	5.8×10^{-5}	9.0	0.81	Spring uses on leafy vegetables and autumn uses on oilseed rape. Application up to 1,500 g/ha.
Epoxiconazole (EPZ)[d,e]	Fungicide	208.8	3.5×10^{-7}	7.1	1.22	Late spring and summer uses on cereals and sugar beets up to 2 × 125 g/ha.
Difenoconazole (DFZ)[d]	Fungicide	143.8	3.3×10^{-8}	15.0	0.50	Uses on various crops (potato, sugar beet, oilseed rape, vegetables) up to 150 g/ha.

(continued)

Table 1 (continued)

Substance[a]	Type	Mean annual PPP sales 2018–2022 (ton)[a]	Vapor pressure (Pa)[b,c]	Water solubility (mg/L)[b]	Half-life in air (days)[c]	Pesticide application details
Boscalid (BSC)[d,e]	Fungicide	102.0	7.2×10^{-7}	4.6	1.17	Spring uses on cereals, vegetables and oilseed rape up to 2 × 400 g/ha. Summer/early autumn uses on beans, cereals, sunflower, and vegetables up to 2 × 500 g/ha. Up to 4 × 67 g/ha on potatoes.
Mandipropamid (MDP)[e]	Fungicide	92.7	9.4×10^{-7}	4.2	0.11	Various crop uses (wine grapes, potatoes, tomatoes, leafy vegetables, herbs, hops). Application up to 2 × 150 g/ha.
Fluxapyroxad (FLXP)[e]	Fungicide	86.0	2.7×10^{-9}	3.4	0.69	Various crop uses (pome fruits, stone fruits, grapes, root vegetables, cucurbits, brassica, etc.). Application up to 150 g/ha.
Fluopyram (FPY)[d,e]	Fungicide	83.5	1.2×10^{-6}	16.0	1.73	Various crop (tobacco, berries, herbs, leafy vegetables, cereals, legumes, pome fruit, etc.) and non-crop (ornamental plants, lawns) uses.
Dimethomorph (DMM)[d,e]	Fungicide	67.4	9.7×10^{-7}	29.0	0.10	Summer uses on potato and oilseed rape. Late spring to early autumn uses on vegetables. Application up to 8 × 180 g/ha on potato.
Fluopicolide (FPC)[d]	Fungicide	21.0	3.0×10^{-7}	2.8	2.24	Various crop uses (potato, fruiting vegetables, blackberries, hops, wine grapes).
Prosulfocarb sulfoxide (PSC met)	Metabolite	N/A	3.8×10^{-5} [h]	705 [h]	0.30	N/A. See above for details regarding use of parent active substance.
Terbuthylazine desethyl (TBA met)	Metabolite	N/A	3.5×10^{-4}	327.1	17.3	N/A. See above for details regarding use of parent active substance.
Prothioconazole desthio (PTCZ met)	Metabolite	N/A	5.2×10^{-7} [h]	66.4 [h]	1.18	N/A. See above for details regarding use of parent active substance.
Pendimethalin M455H001 (PMD met)	Metabolite	N/A	1.0×10^{-10}	100	0.35	N/A. See above for details regarding use of parent active substance.
Carbendazim (CBZ)	Biocide	0	9.0×10^{-5}	8.0	0.053	N/A. Agricultural uses in Germany ended in 2015. Biocidal uses are still registered (e.g., to protect painted surfaces).

[a]Abbreviations in parentheses are used elsewhere in the present article. Substance sales data were taken from the BVL database (BVL, 2025b);
[b]Values taken from the University of Hertfordshire Pesticide Properties Database (Lewis et al., 2016);
[c]Half-life with respect to OH-initiated photooxidative degradation in the gas phase, calculated using AOPWIN within the U.S. Environmental Protection Agency Estimation Programs Interface (U.S. Environmental Protection Agency, 2024), assuming 12-hr average [OH] = 1.5×10^6 molecule/cm^3;
[d]Among the 20 most frequently observed substances in bulk samplers at NEU;
[e]Among the 20 most frequently observed substances in bulk samplers at WBB;
[f]Parent substance not observed. Desthio metabolite among 20 most frequently observed substances in bulk samplers at NEU and WBB;
[g]Literature value not available (see online supplementary material Table S1);
[h]Value estimated via the Estimations Program Interface for the web (U.S. EPA, 2024).

date of October 2021. This change in registration status and use is reflected in the bulk deposition data, with very small amounts (<10 ng/m^2/day) of epoxiconazole measured at NEU in 2022 and no detections at WBB in 2021 and 2022.

Second, distributions of bulk deposition rates at the "background" WBB site are generally lower, and with less spread, than at the "agricultural" NEU site. An exception to this trend was carbendazim. This was the only compound among

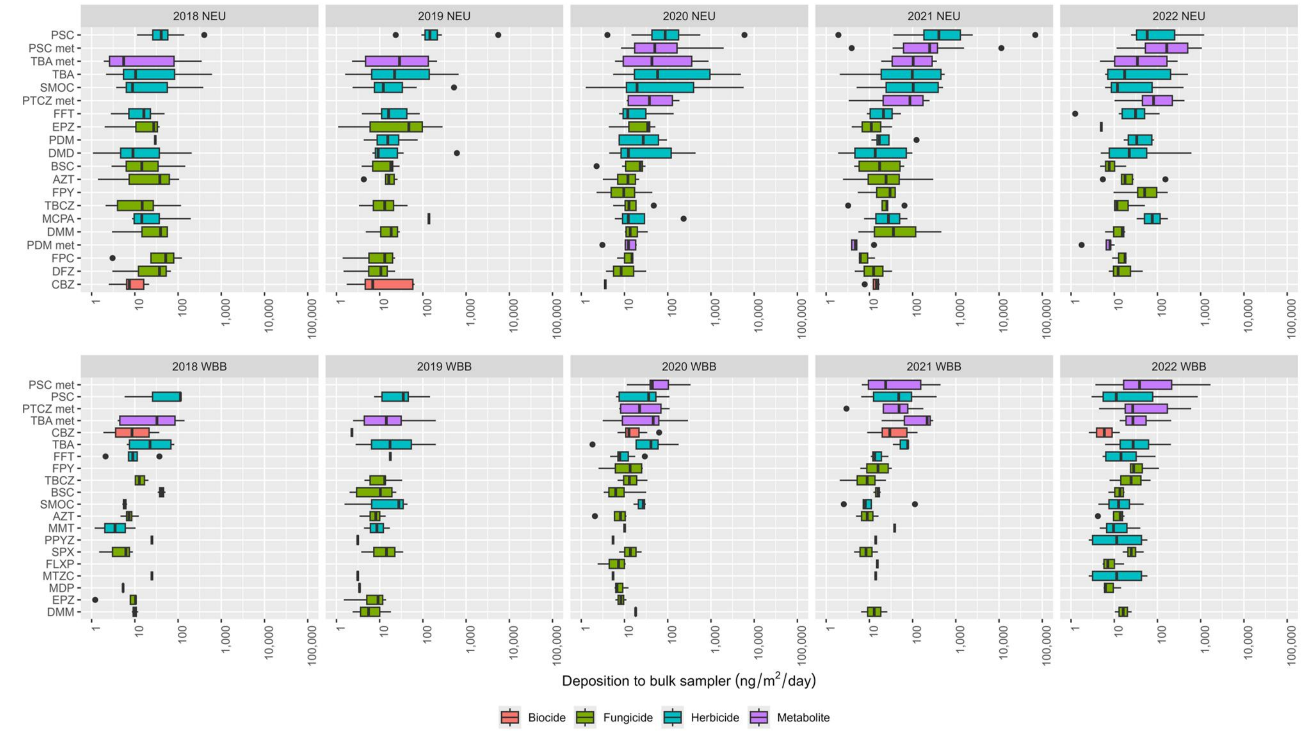

Figure 1 Box-and-whisker plots of bulk-sampler measurements, expressed as deposition rates, for the 20 most frequently detected substances at Neusling (NEU) and Weibersbrunn (WBB), respectively. The boxes represent interquartile ranges (IQR) and the vertical lines within the boxes represent medians. Measurements significantly outside the boxes (defined as above 1.5 × IQR) are indicated as data points. Notes: The horizontal scale is logarithmic. Prosulfocarb sulfoxide and fluopyram were not analyzed in 2018 and 2019. Abbreviated substance names are indicated in Table 1.

the 20 most frequently detected substances at the two sites with significantly overall higher detection frequency (Figure 2) and total deposition at WBB than at NEU (Figure 3). Agricultural pesticide use of carbendazim ceased in 2015. However, this substance is still registered in Germany as a biocide to protect painted surfaces and roofs from algal and fungal growth; hence, it may be washed out of coatings by rain (Umwelt Bundesamt [UBA], 2020) and/or volatilize. Further studies will be necessary to investigate the sources of carbendazim deposition at NEU and WBB. Nonetheless, continuous detections throughout the study years, higher and more frequent findings at WBB, and no detections in bioindicator plant samples (see below) point toward sources outside of agricultural use.

Third, Figure 1 shows exceptionally high deposition values measured at NEU for prosulfocarb and its sulfoxide metabolite in 2021. For the sampling period April 28, 2021 to May 26, 2021, the concentration of prosulfocarb in the rainwater was 14,000 ng/L (=14 µg/L), yielding a deposition rate of 69,000 ng/m^2/day (considering day-average rainfall of 4.93 L/m^2 in this period). Considering the 5-year prosulfocarb data set, this maximum value is >60 times the 95th percentile deposition rate, and approximately 2,000 times the median. Direct spray drift from potato cultivation in a field only 5 m away from the bulk deposition sampler (see online supplementary material Figure S6) is a plausible explanation for the extraordinarily high finding. Prosulfocarb spraying to potato plants in this field in 2021 was confirmed by the farmer (via verbal communication). The peak deposition rate of prosulfocarb corresponds to approximately 0.5% of the maximum registered field application rate of 4 kg/ha for the use in potatoes (considering the entire 28-day sampling period). Similar findings on the low deposited amount in relation to the applied dose in the field (0.0001%–0.1%) were reported in precipitation monitoring in Sweden (Kreuger & Lindström, 2019).

Agricultural activity at the NEU site, and hence emission of pesticide substances to air, is not anticipated to influence observed substances depositions at the WBB site: the "background" site is 280 km northwest of the "agricultural site," and the prevailing wind direction at Würzburg (∼50 km from WBB) is west-southwest (Davis et al., 2023). That said, it is interesting to further compare the distribution of the deposition data between the two sites; the percentage of samples containing levels > LOQ at the WBB site are plotted against the same statistic for the NEU site in Figure 2, considering the entire 2018–2022 data set. Systematic monotonic correlation between the two data sets is apparent, with R^2 = 0.82 for an indicative linear correlation and considering carbendazim an outlier. This suggests that observed deposition to the bulk-deposition samplers at WBB is influenced by local (but >3 km distant) agricultural activity, with a similar combination of use of PPPs as near NEU. In addition, 75th percentile, 95th percentile, and maximum measured deposition rates at NEU were plotted against those at WBB in Figure 3, for the substances most frequently detected at both sites. Within the scatter, a general monotonic relationship is apparent in the data when comparing the different substances—larger deposition amounts at NEU correspond to larger

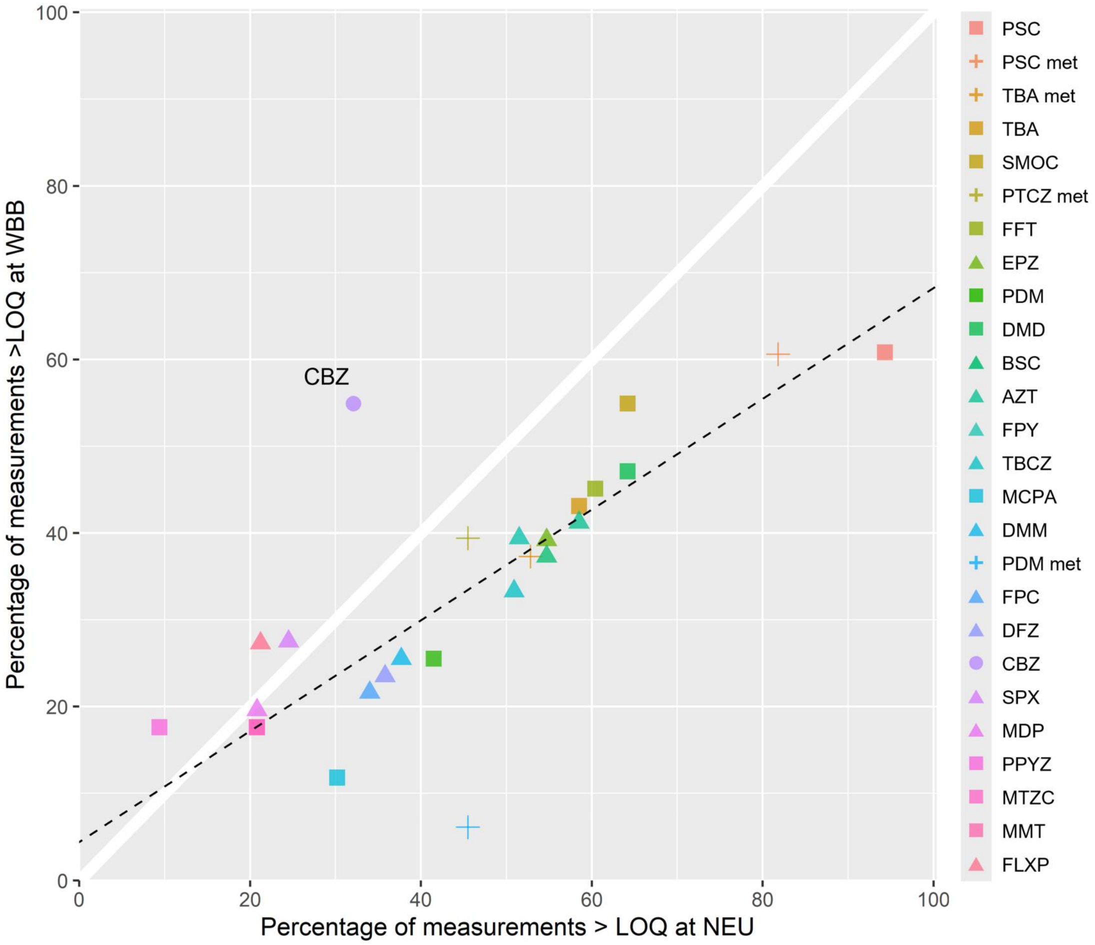

Figure 2 Percentage of measurements > limit of quantification (LOQ) in samples collected at the Weibersbrunn (WBB) site plotted against the respective statistics at the Neusling (NEU) site, for the 20 substances found most frequently at each site (refer to Figure 1). The data marker shapes indicate substance types (square = herbicide, Greek cross = metabolite, triangle = fungicide, circle = biocide). The entire 2018–2022 data set was considered. The white diagonal line represents indicative 1:1 correspondence. The least-squares linear regression for the data, excluding carbendazim (CBZ) is represented by the black dashed line (slope = 0.64, intercept = 4.41, R^2 = 0.82). Abbreviated substance names are indicated in Table 1.

deposition amounts at WBB. However, it is also apparent that the deviation from the indicative 1:1 correspondence line increases as the quantile compared increases (i.e., the individual data points appear further below the 1:1 line), except for carbendazim. This suggests the underlying distributions of deposition measurements at NEU are different (wider and more positively skewed) from those of WBB. One potential reason for this is that certain mechanisms of atmospheric transport, such as spray-droplet drift or soil-dust drift, only operate over short distances due to rapid gravitational settling. The WBB site is further away from agricultural land (see online supplementary material Figure S5b). Therefore, pesticide deposition at this site will more likely result from the atmospheric transport of vapor-phase substance. The transport of small atmospheric particles containing pesticide substances (e.g., formed because of soil erosion at high wind speeds) could in principle also operate over significant distances. However, the measured bulk deposition of metamitron at the WBB site was 1 to 2 orders of magnitude lower than that measured for prosulfocarb, despite the annual average sales of metamitron being slightly greater than those of prosulfocarb (see Table 1). In contrast to prosulfocarb, metamitron is expected to have a very low volatilization potential post spray-application: its vapor pressure is 7.4×10^{-7} Pa, and its water solubility is 1,770 mg/L (giving a calculated Henry's law constant of 8.9×10^{-8} Pa m^3 mol^{-1}). Although the low levels of metamitron deposition measured at WBB may be due to the transport of fine fugitive soil dust particles, the significantly greater deposition of prosulfocarb seems more likely to be associated with its higher volatility and much greater extent of vapor transport. To confirm this assumption, sample collection with physical filters would be necessary, because the bulk samplers used in this study collected both deposited vapor and particles. For terbuthylazine and prosulfocarb, the predominance of vapor transport to WBB is also supported by the parent substance and metabolite distributions at the two sites (see next section *Time-resolved bulk deposition data for active substances and their metabolites*). In some cases, seasonal differences were observed, with higher deposition rates at WBB than at NEU, such as for prosulfocarb in autumn. Nonetheless, overall, the bulk deposition data show that both frequency of findings and

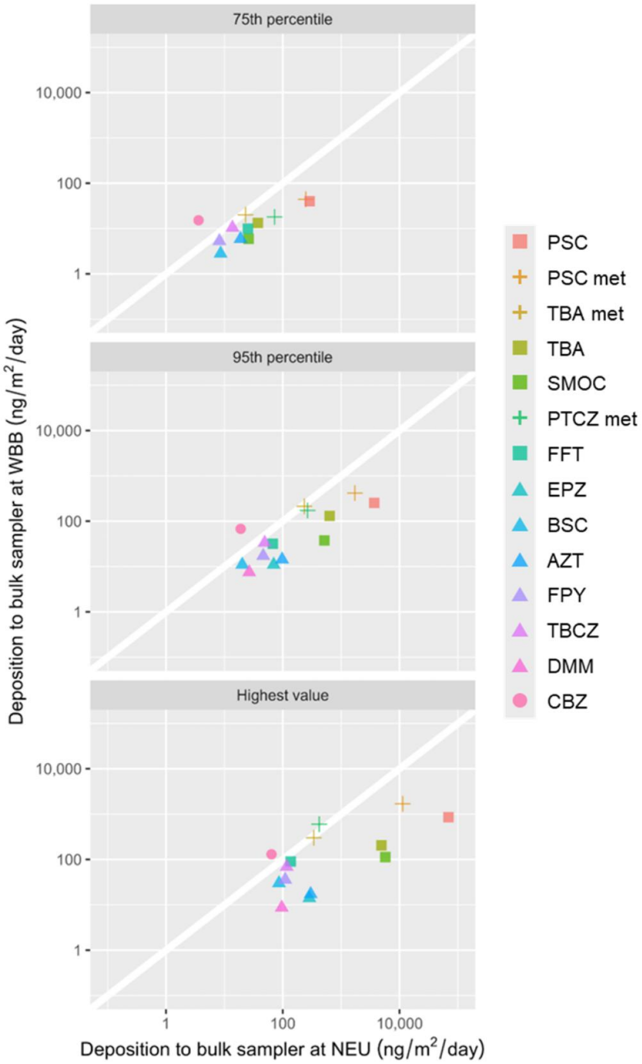

Figure 3 Quantiles of substance deposition rate for WBB plotted against the respective statistics for Neusling (NEU), considering data from 2018 to 2022. The data marker colors indicate different substances. The data marker shapes indicate substance types (square = herbicide, cross = metabolite, triangle = fungicide, circle = biocide). The white diagonal line represents 1:1 correspondence. (Note: the vertical scale is logarithmic.). Abbreviated substance names are indicated in Table 1.

deposition rates of PPP active substances at the WBB site are considerably lower than at NEU. The single highest pesticide active substance deposition rate at WBB was 855 ng/m^2/day, measured for prosulfocarb in 2022. For the entire 28-day sampling period, this deposition corresponds to 0.006% of the maximum registered application rate (4 kg/ha) for the use of prosulfocarb PPPs to treat potatoes and winter cereals.

Time-resolved bulk deposition data for active substances and their metabolites

Some PPP active substances degrade to detectable metabolites in the environment over time. The ratio of metabolite to parent can provide insights into the processes influencing the environmental fate and transport of these compounds. Three metabolites from PPP active substances are among the 20 most frequently measured substances at the NEU and WBB sites: prosulfocarb sulfoxide, prothioconazole desthio, and terbuthylazine desethyl. These are known biotransformation products formed in soil under aerobic conditions (see online supplementary material Box S2). Prothioconazole was not observed in previous LfU bulk-deposition measurement campaigns, most likely due to its fast degradation in the environment (LfU, 2019), and was therefore not measured in the current campaign. Thus, only terbuthylazine, prosulfocarb, and their respective metabolites are considered in the following analysis.

Bulk deposition data for terbuthylazine and its desethyl metabolite are presented in Figure 4. The left panels are for the "agricultural" NEU site, and those on the right for the "background" WBB site. The upper panels present calculated deposition rates for the parent substance and the lower panels present ratios of metabolite to parent active substance concentrations measured in the precipitation collected in the bulk samplers. To aid the following discussion, annotations have been added in red text to the figure. Maximum terbuthylazine deposition occurred in 2020 at both sites (annotation a), within the

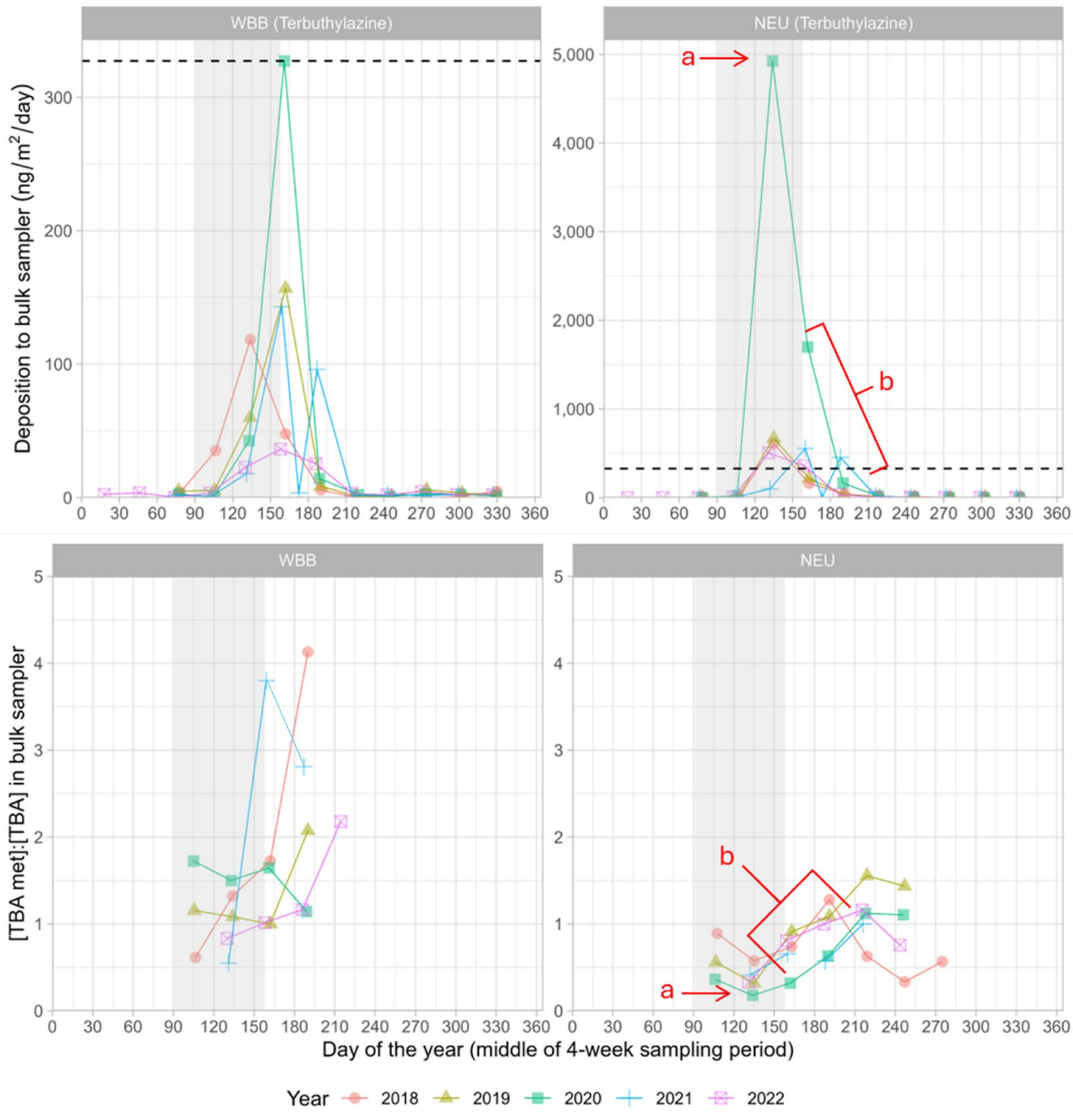

Figure 4 Upper panels: Daily bulk deposition rate for terbuthylazine at Weibersbrunn (WBB) and Neusling (NEU) from 2018 to 2022. Lower panels: Ratio of terbuthylazine desethyl (TBA met) to terbuthylazine (TBA) deposition rate to bulk samples. The shaded rectangle represents the typical time window when terbuthylazine is used in maize. The letter annotations are discussed in the main text. The dashed horizontal lines represent the highest magnitude of TBA deposition observed at WBB.

typical herbicide application time window for maize cultivation. Corresponding to this peak, the metabolite-to-parent ratio is close to zero at NEU, indicating that terbuthylazine captured by the bulk sampler resulted from PPP spraying activity nearby. In 2020, maize was grown in the field directly adjacent to the NEU measurement site, which further suggests that the peak terbuthylazine measurements result directly from spray drift (see online supplementary material Figure S10). For all the measurement years at NEU, after occurrence of the peak terbuthylazine deposition, the magnitude of deposition was observed to decrease while the metabolite-to-parent ratio increased (annotation b). At WBB, peak measured bulk-depositions were observed 1 month (i.e., one 28-day sampling period) later than peak deposition at NEU, with significantly higher measured metabolite-to-parent concentration ratios than for the NEU peak depositions. This suggests that within a period of 28 days, in fields in the general vicinity of (but >3 km from) WBB, a proportion of the sprayed terbuthylazine that initially deposited to soil was metabolized by soil microorganisms and subsequently volatilized, along with some unmetabolized terbuthylazine. The significantly smaller terbuthylazine depositions observed at NEU, after the peaks, also suggest a similar and relatively small influence of post-spray volatilization and subsequent deposition at this site. In samples collected at later times at WBB (after Day 160 and up to Day 210), rates of terbuthylazine deposition (for the 28-day sampling time periods) decreased to effectively negligible levels. At the same time, the ratios of metabolite-to-parent concentration in collected water generally increased. This is consistent with continued substance volatilization from treated fields (located >3 km from the WBB site), transport in air to, and then deposition at, the WBB measurement site, for up to approximately 50 days. Throughout this approximately 50-day period, metabolic processing of terbuthylazine in soil will have continued to occur (increasing the metabolite levels), but additionally/in parallel the terbuthylazine level in the soil will have been decreasing with time (e.g., due to leaching and degradation to various products, including, that is, not exclusively, desethyl terbuthylazine). In absolute quantitative terms, the peak depositions at NEU were much larger than the peak depositions at WBB (as indicated by the dashed lines in the upper panels of Figure 4), indicating that post-spray volatilization and vapor deposition is a relatively much more minor process.

Data for prosulfocarb and its sulfoxide metabolite are presented in Figure 5. Variations in parent bulk-deposition and corresponding metabolite-to-parent ratios through time appear more complex than for terbuthylazine, likely because prosulfocarb PPPs are registered for use on a greater number of crops. As for terbuthylazine, there are indications that the maximum observed bulk deposition rates in each year at NEU were likely due to nearby spray-drift (see annotation a), as they occur during the application period to potatoes, which are grown in the vicinity of the site, and are coupled with low metabolite-to-parent ratios. The exceptionally high measurement of prosulfocarb in 2021 shown in Figure 1 (69,000 $ng/m^2/day$ for the sampling period Day 118 to Day 146) is not included in Figure 5 to maintain readability, but it is further discussed in the context of the bioindicator measurement results below. The metabolite-to-parent ratio associated with this exceptionally high deposition was 0.16, consistent with the hypothesis stated above that the peak deposition of prosulfocarb in 2021 was caused by spray drift. Another, significantly smaller, peak in bulk deposition in 2021 appears at the NEU site outside of the time window for treatment of potatoes or winter-cereals (see annotation b). The broad registration of prosulfocarb for a large variety of crops makes it difficult to further interpret this second peak.

The metabolite-to-parent concentration ratios at NEU in autumn corresponded with much lower magnitudes of parent deposition than in the spring, particularly in 2020 (Figure 5, annotation c). It is unlikely that the observed autumn depositions are due to PPP spray application in the springtime, as prosulfocarb exhibits low persistency in soil. The geometric mean degradation half-life of prosulfocarb in aerobic soil was 11.9 days in laboratory studies and 9.8 days in field studies (EFSA, 2007). Instead, the autumn measurements most likely consisted of vapor phase prosulfocarb originating from autumn PPP application in cereal fields located within a few kilometers from the measurement site. Furthermore, similar deposition rates (within a factor of 2) of prosulfocarb were measured at the "agricultural" and "background" sites during the autumn PPP application window for winter cereals in 2018 to 2022 (see dashed horizontal lines in the upper panels of Figure 5). The corresponding high metabolite-to-parent ratios suggest that the measured autumn deposition at both sites predominantly resulted from vapor transported a few kilometers from recently treated cereal fields. The collected land use data shows that cereal fields cover 24% to 30% and 30% to 35% of the land within a 3 to 15 km radius of the NEU and WBB sites, respectively (see online supplementary material Table S4 and Figure S5b). One additional factor to consider, particularly for herbicides applied at pre- or early emergence, is potential volatilization from soil particles during plowing. This issue was raised in the EXPOPESTEN study in Belgium, as a potential explanation behind the higher prosulfocarb concentrations in autumn, when prosulfocarb is used as pre-emergent treatment of winter cereal fields (ISSEP, 2018).

Similar observations regarding the influence of nearby agricultural land were made in the Danish precipitation monitoring Denmark, where 94% of the measured prosulfocarb deposition in 2019 was from September to December, at the time of PPP treatment of winter cereals fields. For 2007–2019, a correlation was observed between the annual wet deposition amount of prosulfocarb and the surface area of land cultivated with winter cereals (DCE, 2021).

Examination of possible correlations between meteorological parameters and bulk deposition

Comparison of meteorological parameters (temperature, precipitation, relative humidity, and wind speed) with depositions measurements revealed no clear correlations. See online supplementary information (e.g., Figure S11) for bulk-deposition and precipitation data for prosulfocarb and terbuthylazine (and their measured metabolites) at the two measurement sites. Absence of correlation between amounts of deposition and meteorological parameters was also concluded in previous studies by the LfU (2019). In monitoring studies in Denmark conducted from 2007 to 2019 at two measurement sites, annual measured pesticide wet deposition (from "wet-only" precipitation samplers) was generally well correlated with annual levels of rainfall (DCE, 2021).

Deposition to bioindicator plants

An overview of the substances detected in the biomonitoring samples is presented in Table 2 (and in the online supplementary material Figure S12). Of the 18 substances selected for analysis at the lower LOQ of 0.001 mg/kg (see *Materials and Methods*) based on previous experience of the LfU, all substances except carbendazim and isoproturon were observed in bioindicator plant samples at levels above the LOQ (with both of these substances being analyzed for in the years 2020–2022, with a measurement limit of 0.001 mg/kg). It was already mentioned above that carbendazim crop protection uses in

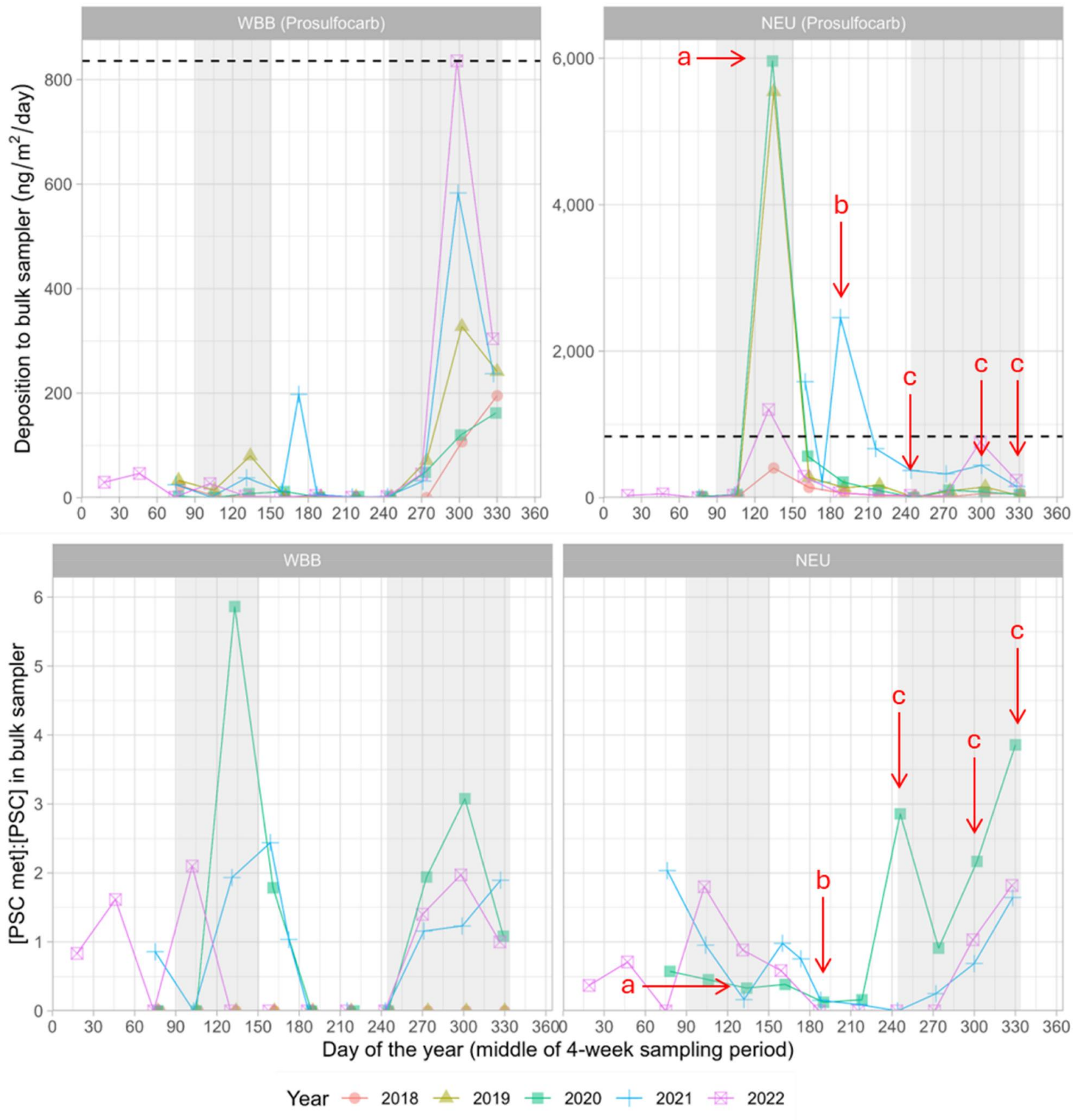

Figure 5 Upper panels: Bulk deposition rates for prosulfocarb at Weibersbrunn (WBB) and Nesling (NEU) for the years 2018 to 2022, omitting the exceptionally high value measured in April/May 2021 at NEU for readability. Lower panels: Ratio of prosulfocarb sulfoxide (PSC met) to prosulfocarb (PSC) concentration in bulk deposition samples. The shaded rectangle represents the typical time windows when prosulfocarb is used to treat potatoes in spring and winter cereals in autumn, respectively. The red annotations of the 2020 and 2021 data are discussed in the main text. The dashed horizontal lines represent the highest magnitude of prosulfocarb deposition observed at WBB.

Germany officially ceased in 2015, although some biocidal uses remain. The agricultural use of isoproturon was discontinued in Germany at the end of 2017 and, to the authors' knowledge, there are no nonagricultural uses of this substance in Germany.

For substances detected at least once ≥0.01 mg/kg in bioindicator plants, the number of detections at the three monitoring sites together during all study years (2020, 2021, and 2022) are shown in Table 2. See online supplementary information for disaggregated data for each site and measurement year (Figure S12). Time-resolved data for these substances are provided in the online supplementary material Figures S13 and S14. Insecticides were not detected in the biomonitoring study. The most frequently measured substances with at least five detections ≥0.01 mg/kg were: fluazinam, fluopyram, metribuzin, pendimethalin, propamocarb, prosulfocarb, and terbuthylazine. It should be noted that due to the sampling interval from April

Table 2 Number of samples containing substance levels ≥0.01 mg/kg and between 0.001 and 0.01 mg/kg, for the Neusling (NEU), Uttenkofen (UTK), and Aldersbach (ALR) sites considered together, for the years 2020–2022. The total number of samples collected in the study was 216.

Substance[a]	No. of detections ≥0.01 mg/kg		No. of detections ≥0.001 and <0.01 mg/kg	
	Ryegrass	Chard	Ryegrass	Chard
Results for substance measurements with LOQ of 0.001 mg/kg				
Dimethomorph	1	0	6	4
Ethofumesate	1	0	13	0
Pendimethalin (PDM)	6	0	22	20
Phenmedipham	4	0	0	1
Propamocarb	17	8	22	23
Prosulfocarb (PSC)	14	6	14	14
S-metolachlor (SMOC)	2	0	9	5
Tebuconazole (TBCZ)	2	0	10	3
Terbuthylazine (TBA)	21	27	24	26
Results for substance measurements with LOQ of 0.01 mg/kg				
Azoxystrobin (AZT)	2	0	N/A [b]	N/A [b]
Aclonifen	4	0	N/A [b]	N/A [b]
Benthiavalicarb	1	0	N/A [b]	N/A [b]
Cyproconazole	1	1	N/A [b]	N/A [b]
Fluazinam	5	0	N/A [b]	N/A [b]
Fluopyram (FPY)	5	0	N/A [b]	N/A [b]
Mandipropamid	1	0	N/A [b]	N/A [b]
MCPA	2	0	N/A [b]	N/A [b]
Metalaxyl-M	3	0	N/A [b]	N/A [b]
Metribuzin	6	10	N/A [b]	N/A [b]
Metobromuron	1	0	N/A [b]	N/A [b]
Prothioconazole	2	0	N/A [b]	N/A [b]

[a]Substances with abbreviations in parentheses were amongst the 20 most frequently observed substances in bulk deposition samplers at the NEU and/or WBB sites (see Table 1);
[b]Not applicable.

until mid-October, not all pesticide applications could be covered with the chosen bioindicator monitoring approach. Spring herbicides in cereals, for example, are typically applied from March to April, whereas autumn herbicide applications can extend into November. Similar to the highest bulk deposition rates observed at NEU, the most frequently observed deposition to bioindicator plants was for substances that: are registered for use on crops known to be grown near the NEU site; have non-negligible volatilization potential; are sold in large quantities in Germany; and have a maximum registered application rate of 500 g/ha or greater. Most substance detections in the biomonitoring dataset occurred within typical PPP application time windows in each of the three measurement years. One exception was terbuthylazine, which was regularly observed in bioindicator plant samples months after the typical PPP application time ended. This could be a substance-specific effect and should be investigated in greater detail in the future.

Differences were observed between the results from the two bioindicator plant measuring systems regarding the frequency and magnitude of substances detected (see Table 2 and online supplementary material Figures S13, S14, and S15). Overall, 99 detections >0.01 mg/kg were made in ryegrass and 53 in chard. Four substances were detected every year at least once in ryegrass ≥0.01 mg/kg: pendimethalin, propamocarb, prosulfocarb, and terbuthylazine (see online supplementary material Figure S12). For most substances, ryegrass was the more sensitive bioindicator plant. This outcome is expected as the ryegrass measuring system was designed with the primary goal to filter small amounts of trace substances from the air, whereas the chard measuring system is designed/intended to investigate possible residues in feed and food crops. Nonetheless, chard appeared to be the more sensitive monitoring system for two substances (terbuthylazine and metribuzin) in this study. This is likely a substance-specific effect that requires further investigation.

Exceptionally high prosulfocarb levels measured in 2021 in bioindicator samples

The highest concentrations observed in bioindicator-plant samples were for prosulfocarb. In 2021, at the UTK and NEU sites, extraordinary high levels of prosulfocarb were measured in chard (1.2 and 0.31 mg/kg, respectively) and ryegrass (3.3 and 1.9 mg/kg, respectively) for the same sampling period (see online supplementary material Figure S14), which was within the typical application time for prosulfocarb in potatoes. The finding at NEU has already been described in the context of the bulk-deposition data, where an exceptionally high prosulfocarb concentration was detected for the same sampling period and is most likely linked to prosulfocarb application in a potato field located just a few meters away from the measuring site. At UTK, during harvesting of the bioindicator plants for this sampling period (April 24, 2021–May 22, 2021), a layer of soil particles was visible on the plants (see online supplementary material Figure S7). This soil particle layer was not observed at any other point throughout the biomonitoring study. In 2021, potatoes were grown on 18% of the area within a 100 m radius (0.6 ha) of the UTK site and on 17% of the area (12.6 ha) located between a 100 to 500 m radius of the UTK site (see online supplementary material Table S3). Thus, it is plausible that soil particle drift from a nearby prosulfocarb-treated field resulted in the extraordinarily high prosulfocarb concentrations in bioindicator samples.

Bioindicator-plant data in the context of agricultural land use

As information on actual PPP applications made around the measurement sites was not available, information on the crops grown in the vicinity of the sites was collected and analyzed to further investigate the sources of pesticide detections. Agricultural land use differed among the measurement sites and changed over the years (see online supplementary material Table S3). For this analysis, bioindicator-plant data were considered for herbicide substances registered for use on maize, potato, and sugar beet. These three crops were grown within a radius of 1.5 km around the ALR, NEU, and UTK sites during 2020 to 2022, but to different extents. Additionally, time-resolved bioindicator-plant and bulk-sampler data sets were compared for NEU.

At the time of the study, terbuthylazine was one of the most widely used maize herbicides in Germany and was only approved for use in this crop. It was also the substance with the most detections in the bioindicator plant samples (Table 2). Findings of terbuthylazine in ryegrass from 2020 to 2022 at the UTK site are shown in Figure 6. The peak concentrations on ryegrass at all sites and study years were found within the typical main PPP spray application period for terbuthylazine (Figure 6 and online supplementary material Figure S14). However, lower concentrations were also measured in the summer of 2021 at the UTK site, outside the typical application period. Most of the other PPP substances were only detected in biomonitoring samples within their typical PPP application periods. Peak concentrations of terbuthylazine in ryegrass at UTK were 0.02 mg/kg, 0.13 mg/kg, and 0.11 mg/kg, in 2020, 2021, and 2022, respectively (Figure 6). These measurements correspond well with differences in land area used for maize cultivation in the vicinity of the UTK site (Figure 6). In 2020, maize was not grown within a 100 m radius of the measuring site, and only 5.7 ha were cultivated with maize at distances between 100 and 500 m away from the site. In 2021, 1.3 ha of maize were grown within a radius of 100 m from the site, and an additional 11.7 ha were grown at 100 to 500 m. In 2022, 0.7 ha and 12.6 ha of maize were grown within a radius of 100 m and at 100 to 500 m, respectively. A similar observation regarding measured levels of terbuthylazine and the area used for maize cultivation was observed at the remaining biomonitoring sites (see online supplementary material Figure S16c).

Although prosulfocarb was registered for use in various crops, the main application of the herbicide during the study period was to potatoes. Maximum bulk deposition and peak concentrations of prosulfocarb in ryegrass (0.29 mg/kg and 0.05 mg/kg at UTK and NEU, respectively) in 2020 were measured within the expected time window of prosulfocarb use in potatoes (see Figure 7). The relative magnitudes of the prosulfocarb biomonitoring results in spring fit well with the relative sizes of the potato cultivation areas around the measuring sites. At UTK, potatoes were grown in 44% (1.4 ha) of the area in a distance up to 100 m around the measuring site and in an additional 22.3 ha in a radial area between 100 and 500 m of site. At NEU, potatoes were not grown within 100 m, but 11.3 ha were cultivated between 100 to 500 m of the site. Similar observations for prosulfocarb were made in 2022 (see online supplementary material Figure S17), and results for 2021 were already discussed above. A very low concentration of prosulfocarb was measured in/on rye at ALR, where potatoes were not grown within 500 m of the measuring site. It cannot be further elucidated if the observed low detection <0.01 mg/kg originated from potato-cultivation activity a greater distance away or potential prosulfocarb treatment of different crops, such as legumes (see online supplementary material Table S3).

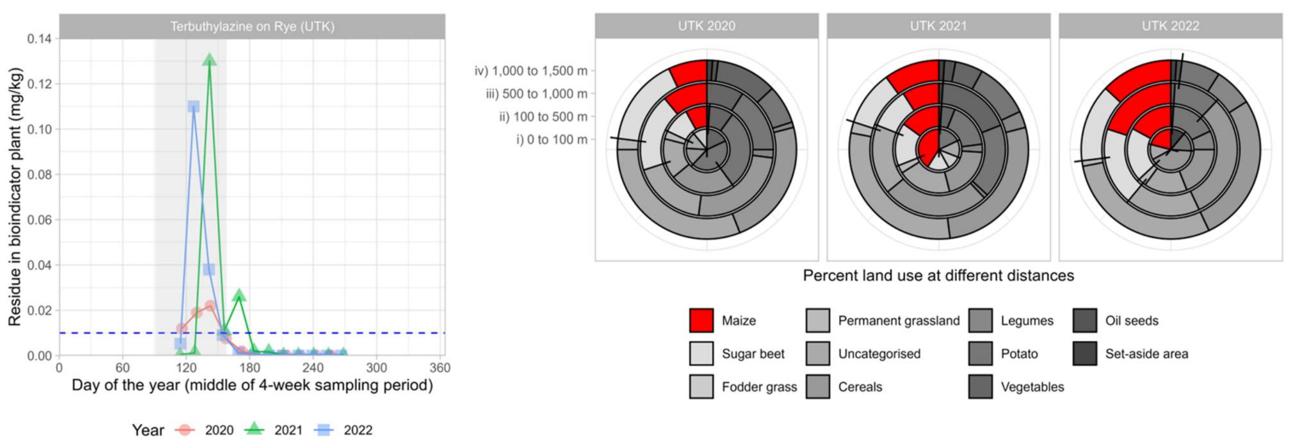

Figure 6 Left panel: Measured residue levels of terbuthylazine in ryegrass bioindicator samples, at the Uttenkofen (UTK) site (2020–2022). The shaded area indicates the typical application time-period of terbuthylazine in maize. The dotted horizontal line corresponds to a residue level of 0.01 mg/kg. Right panel: Proportion of land area surrounding the UTK measurement site used to cultivate different crops in 2020, 2021, and 2022.

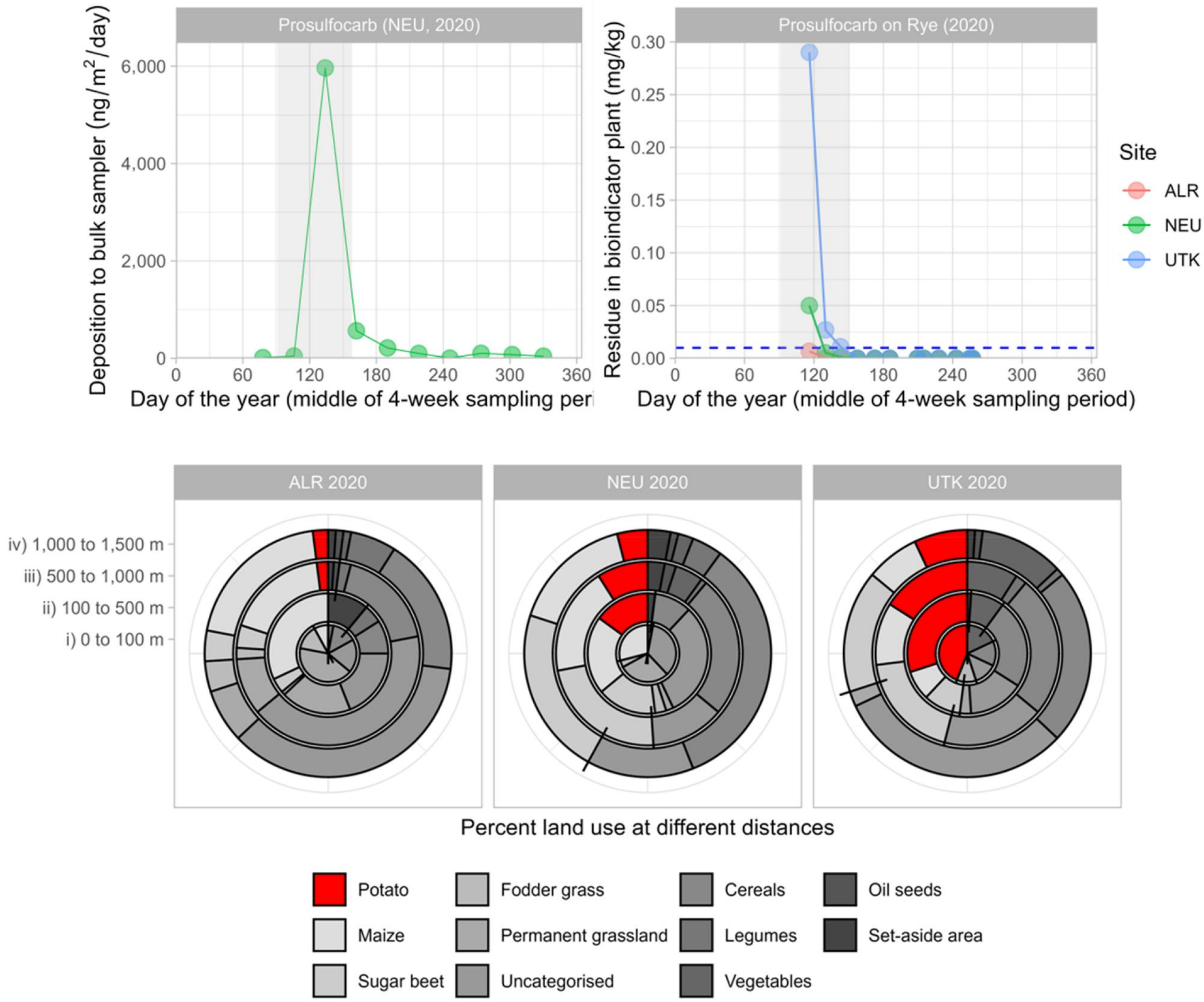

Figure 7 Upper-left panel: Prosulfocarb bulk deposition rates at the Neusling (NEU) site for 2020. Upper-right panel: Residue levels of prosulfocarb in ryegrass bioindicator samples, at the Aldersbach (ALR), NEU, and Uttenkofen (UTK) sites (2020). The shaded area indicates the typical prosulfocarb application period in potatoes. The dashed horizontal line corresponds to a residue level of 0.01 mg/kg. Lower panel: Proportion of land area surrounding the ALR, NEU, and UTK sites used to cultivate different crops in 2020.

Two herbicides registered for use on sugar beet during spring were detected in bioindicator plants, both during the expected application window: ethofumesate and phenmedipham. Measured deposition of phenmedipham to ryegrass was significantly greater than for ethofumesate in 2021 and 2022. Differences in measured ryegrass depositions at the three sites correlate well with the different extents of local sugar beet cultivation. No deposition to ryegrass or chard was observed throughout all study years at the ALR site (see Figure 8 and online supplementary material Figure S18a and b), where sugar beet was only grown in a very small proportion of the area at radial distance 100 to 1,500 m from the site (see Figure 8 and online supplementary material Figure S18c). At the NEU and UTK sites, sugar beet cultivation occurred within a radius of 500 m in 2022, covering 20% and 44% of the area, respectively (Figure 8).

Overall, correlations between the biomonitoring results, agricultural land use, and the usual application windows to target crops suggest that measured deposition of terbuthylazine, prosulfocarb, ethofumesate, and phenmedipham were influenced mainly by agricultural activities near to the measuring stations, likely with minor input from vapor deposition originating from fields further away. Similar correlations between off-field detection of PPP substances and surrounding agricultural land use have been made in other recent publications (e.g., Brühl et al., 2024; Linhart et al., 2019, Zaller et al., 2022, Zivan et al., 2017). Nonetheless, these publications do not discuss the results in the context of good agricultural practice (which determines maximum PPP application rates and timing, and mitigation measures) and of exposure-based risk assessments for humans or non-target organisms. Further studies, such as that planned by the German BVL (Kubiak et al., 2023), would be valuable to elucidate the distances over which field emissions significantly contribute to observed depositions.

The purpose of the present study was not to generate data of direct relevance to the assessment of risk to humans (e.g., via

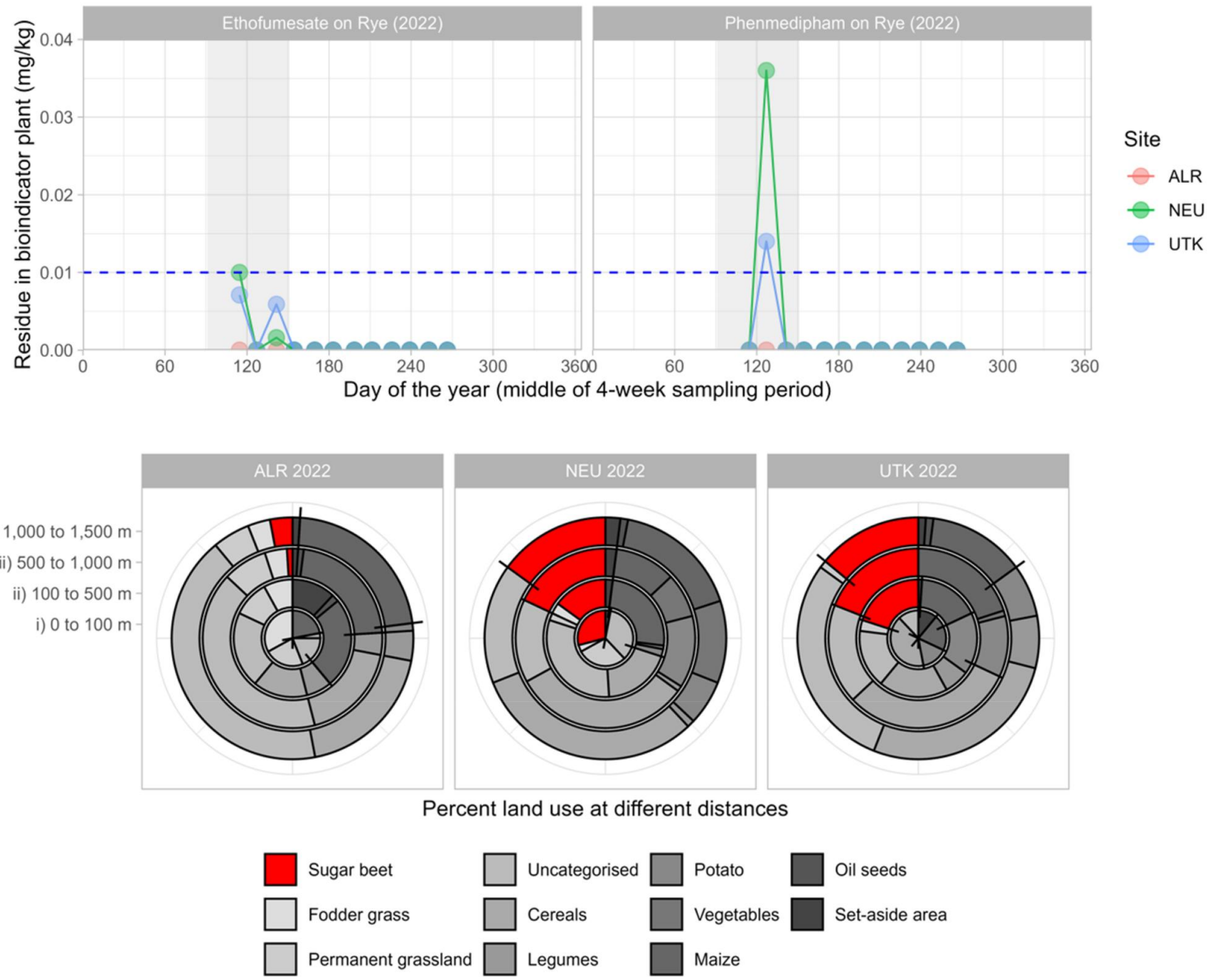

Figure 8 Upper-left panel: Measured residue levels of ethofumesate in ryegrass bioindicator samples, at the Aldersbach (ALR), Neusling (NEU), and Uttenkofen (UTK) sites, in 2022. Upper-right panel: Measured residue levels of phenmedipham in ryegrass bioindicator samples, at the ALR, NEU, and UTK sites (2022). The shaded area indicates the time period when the two herbicides are typically used in sugar beet. The dashed horizontal line corresponds to a residue level of 0.01 mg/kg. Lower panel: Proportion of land area surrounding the ALR, NEU, and UTK sites used to cultivate different crops in 2022.

inhalation exposure to pesticide substances or via consumption of crops with substance residues) or the environment. However, it is important to state that the highest potential exposures, and risks, to both humans and the environment are either within a PPP-treated field (including via treated edible crops) or at its edge. Regulation on the approval of PPP requires that exposure and risk are explicitly assessed for both situations to ensure a safe use. Assuming good agricultural practice, potential exposure further away from the field is lower, as observed in this study, and therefore is covered by the assessed risk “envelope” in/adjacent to the treated field.

It was not possible to identify consistent statistically significant correlations between the bulk-deposition and biomonitoring data sets (see online supplementary material Figures S8 and S13–S15) for the NEU site (where both monitoring systems were set up). This could be consequence of differences in the two measurement setups. For instance, the bioindicator plants were exposed to atmospheric conditions (and thus to variations in temperature and weather), whereas collected rainwater in the bulk samplers was maintained at 6 to 10 °C. Furthermore, the measurement periods do not entirely align. Thus, conclusions about possible MRL exceedances in nearby crops drawn only from data from a single measurement system (passive bulk samplers or bioindicator crops) should be made with caution.

Conclusions

The present study shows the results of 5 years of PPP active substance measurements in bulk precipitation collected at an “agricultural” (NEU) and a “background” (WBB) site and 3 years of biomonitoring at three “agricultural” sites (including NEU) in Bavaria. Substances with the highest deposition to bulk deposition samplers and to bioindicator crops were those with the highest sales, high application rates, and non-negligible volatility potential (based on vapor pressure and equilibrium partitioning between air and moist soil).

A similar suite of substances was observed in bulk samplers at both the NEU and WBB sites from 2018 through 2022. Frequent detections at the "background" WBB site demonstrate off-field atmospheric transport of 3 km or greater. The highest pesticide active substance deposition rate (prosulfocarb) at WBB corresponded to approximately 0.006% of the maximum registered PPP application rate on potatoes and winter cereals. Further research is needed to elucidate the key factors influencing off-field atmospheric transport, including meteorological conditions and substance properties, to derive improved mitigation measures.

While frequencies of detection were similar (within a factor of 3) at both measurement sites, in general, significantly higher active substance deposition rates were measured at NEU compared to WBB. The underlying distributions of deposition rates at NEU compared to WBB suggest that different atmospheric transport mechanisms dominated at each site. This observation was corroborated by analyzing metabolite-to-parent active substance ratios for prosulfocarb and terbuthylazine. For both substance pairs, low metabolite-to-parent ratios were observed at the NEU site and high ratios at the WBB site. This indicates the dominance of a direct exposure pathway at the NEU site—for example, through spray drift and/or dust drift from nearby maize cultivation for terbuthylazine and potato cultivation for prosulfocarb. Conversely, at the WBB site, higher metabolite-to-active substance ratios suggest inputs from volatilized substance that already has undergone degradation on soil or plant surfaces.

Active substance deposition rates to bulk samplers showed clear seasonal patterns, with maximum values occurring during main PPP application periods. This was even more pronounced in biomonitoring samples, with only a few substances being detected outside of typical application times. Biomonitoring of PPP substances was performed from 2020 through 2022 using established ryegrass and chard monitoring protocols. Ryegrass was the more sensitive biomonitoring system, with higher frequencies of detections for most substances. Nonetheless, chard serves as a representative for food and feed crops, making it valuable for studies investigating non-target crop residues. Levels of PPP active substances in bioindicator plant samples generally corresponded with the proportion of land within a 1.5 km radius covered with crops for which the respective PPPs were registered. Crop cultivation within 500 m from the measurement site had the biggest influence on measurable pesticide residues on bioindicator plants. Thus, it is important that all stakeholders continue working toward minimizing such emissions and depositions—for example, through intensified advisory services and appropriate application practices to mitigate spray drift and the potential off-field transport of windborne soil particles.

In summary, the results of the present study demonstrate that different passive monitoring systems such as bulk samplers and bioindicator plants—and even different bioindicator-plant species—yield data that are comparable in some respects but not others. To draw robust conclusions from studies investigating atmospheric transport of pesticides, ideally different deposition measurement systems and types of bioindicator plants should also be combined, if possible, with quantitative active measurements of pesticides in the air compartment itself. Conclusions based on only one monitoring approach should not be transferred directly to another medium. Harmonization in measurement protocols for individual media and, as far as possible, among them (i.e., sampling intervals) is still necessary to enable data comparison within and among studies. Furthermore, to address open questions related to the exposure of non-target crops to PPPs via air, monitoring should allow investigation of the influence of distance from the edge of field in combination with PPP application data. Finally, this study highlighted the importance of clearly characterizing the surrounding environment of a measurement site (particularly land use) and collecting pesticide application data to contextualize and better interpret the results from air monitoring programs.

Supplementary material

Supplementary material is available online at *Environmental Toxicology and Chemistry.*

Data availability

Data are available on request from the corresponding author (Dave.Johnson@Syngenta.com).

Author contributions

Frank Pompe (Data curation, Formal analysis, Investigation, Software, Writing—original draft), Marco U. Reitz (Conceptualization, Funding acquisition, Project administration, Supervision, Writing—review & editing), Björn Brumhard (Conceptualization, Formal analysis, Funding acquisition, Writing—review & editing), Monica Waeber (Investigation, Methodology, Project administration, Writing—review & editing), Annelle A. Mendez Gutierrez (Project administration, Validation, Writing—review & editing), Jutta Köhler (Writing—review & editing), Uwe Kunkel (Investigation, Writing—review & editing), Klaus Gehring (Investigation, Supervision, Writing—review & editing), Markus Scheithauer (Writing—review & editing), and David Johnson (Formal analysis, Visualization, Writing—review & editing)

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Conflicts of interest

The authors declare no conflicts of interest.

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