

## Emission of pesticides during drilling and deposition in adjacent areas

Emission von Pflanzenschutzmitteln während der Aussaat und Deposition in benachbarten Arealen

Udo Heimbach<sup>1\*</sup>, Matthias Stähler<sup>2</sup>, Kristin Schwabe<sup>1</sup>, Detlef Schenke<sup>2</sup>, Jens Pistorius<sup>1</sup>, Pablo-Theodor Georgiadis<sup>1</sup>

<sup>1</sup> JKI, Institute for Plant Protection in Field Crops and Grassland, Braunschweig, Germany

<sup>2</sup> JKI, Institute for Ecological Chemistry, Plant Analysis and Stored Product Protection, Berlin, Germany

\* Corresponding author, [udo.heimbach@jki.bund.de](mailto:udo.heimbach@jki.bund.de)

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### Summary

In seven experiments seeds of maize, oil seed rape and barley, treated with neonicotinoids, were sown using pneumatic drilling equipment with deflectors attached in case of pneumatic suction systems. Directly adjacent to the drilled area of usually about 50 m width were replicated areas with bare soil as well as with crops. During maize (*Zea mays*) drilling flowering oil seed rape (*Brassica napus*) and during drilling of barley (*Hordeum vulgare*) and oil seed rape flowering white mustard (*Sinapis alba*) was adjacent. The amount of residues in the adjacent non crop areas in Petri dishes being distributed on the bare soil declined only slowly from 1 to 20 m distance from the area drilled. Seed batches with more abrasion and higher content of active substances in the dust resulted in higher residues off crop. After drilling of maize in four experiments in Petri dishes in adjacent non crop areas in 1-5 m distance between 0.02 and 0.40 g a.s./ha of neonicotinoids and in the adjacent oil seed rape a total of 0.05–0.80 g a.s./ha were detected. After drilling oil seed rape or barley these values were only 0.02–0.06 g a.s./ha in Petri dishes in non crop areas and 0.03-0.08 g a.s./ha in total in adjacent white mustard. In gauze net samplers installed vertically in 3 m distance in non crop areas up to seven times higher values were detected compared to Petri dishes.

**Key words:** neonicotinoid, seed treatment, dust, drift, adjacent crop

### Zusammenfassung

In sieben Feldversuchen wurde die Abdrift von neonicotinoidhaltigen Stäuben bei der Aussaat von behandeltem Mais, Winterraps bzw. Gerste mit pneumatischen Drillmaschinen bei Nutzung einer Saugtechnik mit Deflektoren zur Abdriftminderung untersucht. Auf den zum etwa 50 m breiten Drillbereich mit echten Wiederholungen benachbarten Flächen blühte Raps bei der Maisaussaat bzw. Senf bei den beiden anderen Kulturen. Staubbürtige Neonicotinoide wurden vergleichend in den blühenden Nachbarkulturen im Pflanzenbewuchs inklusive der Bodendeposition im Bestand und auf offenem Boden in Petrischalen sowie vertikal aufgespannten Gazenetzen analysiert. Die Wirkstoffgehalte in Petrischalen im offenen Bereich und in den Nachbarkulturen fielen bis zu einer Entfernung von 20 m zum Drillbereich nur langsam ab. Saatgutchargen mit höheren Abriebwerten bzw. einem höheren Wirkstoffgehalt im Abriebstaub hatten auch höhere Rückstände im off crop zur Folge. Die nach der Maisaussaat in vier Versuchen ermittelten Wirkstoffgehalte lagen im Abstand von 1 – 5 m in den Petrischalen auf dem Boden im offenen Bereich zwischen 20 - 400 mg/ha, aber als Gesamtsumme deutlich höher in der Nachbarkultur Raps bei 50 – 800 mg/ha. Nach der Aussaat von Gerste (n = 1) bzw. Raps (n = 2) lagen die Werte bei nur 20 – 60 mg/ha in Petrischalen im offenen Bereich und wieder höher bei 30 – 89 mg/ha im benachbarten Senf. In vertikalen Gazenetzen in 3 m Abstand zum Drillen im offenen Bereich ohne Kultur wurden bis zu etwa sieben Mal höhere Gehalte nachgewiesen als im Vergleich zu Petrischalen.

**Stichwörter:** Neonicotinoid, Saatgutbehandlung, Staub, Abdrift, Nachbarkultur

### 1. Introduction

Seeds of many crops are treated with insecticides to protect young seedlings against insect pests with the neonicotinoids imidacloprid, clothianidin and thiamethoxam being frequently used. In 2008 during sowing of maize a bee poisoning of about 12,000 hives was observed in Germany. Relevant amounts of insecticidal substances (neonicotinoids) treated to the seeds drifted into adjacent flowering crops in the form of contaminated dust. Analysis of dust abrasion from different seed batches resulted in up to several g of clothianidin/ha being abraded from seeds of maize and set free in the form of dust (Pistorius *et al.*, 2009; Heimbach *et al.*, 2010). Dust drift containing neonicotinoids such as the three mentioned above which show high effects on bees at very low dose has

been thought to be relevant for bee toxication already in 2003 (GREATTI *et al.*, 2003; 2006) but clear effects associated to drilling of treated maize were identified only later on in Europe (PISTORIUS *et al.*, 2009; GIROLAMI *et al.*, 2012; TAPPARO *et al.*, 2012) and recently in the US (KRUPKE *et al.*, 2012). Dust emission during drilling of treated seeds has opened a new area of research (NUYTTENS *et al.*, 2013) to quantify emission and environmental concentrations in adjacent areas and to predict potential effects on organisms. Therefore in field experiments the emission of pesticides during drilling of maize, barley and oil seed rape was studied and the deposition of the active substance was measured in adjacent areas on bare soil and within adjacent crops to achieve generic drift exposure curves which may be extrapolated to other types of active substances and be used to predict environmental concentration for aquatic and soil organisms and organisms living in adjacent vegetation including crops.

## 2. Material and methods

Field experiments were carried out from 2008 to 2012 using different seeds of different crops as well as different batches of seeds. The abrasion potential of most seed batches was analysed using the Heubach Dustmeter (HEIMBACH, 2008). The content of the active substance in the dust was analysed using the filter within the Heubach-Dustmeter. Pneumatic single corn seeders were used for maize. A deflector was attached to the air outlet, resulting in a 90% drift reduction compared to unchanged systems (ANONYMOUS, 2012). Oil seed rape and barley were sown using pneumatic pressure systems which are frequently used in Germany and only in one case for barley mechanical sowing equipment was used.

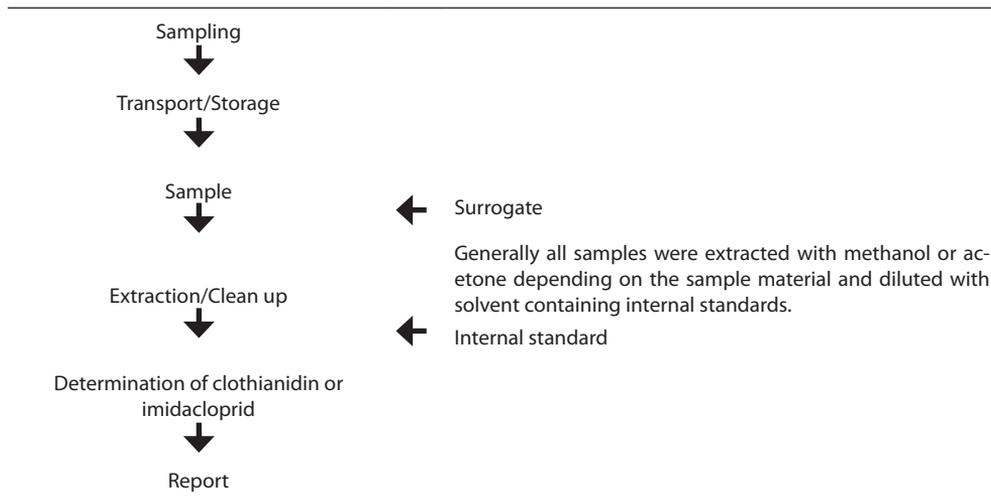
Measurements of drift were carried out on areas with bare ground adjacent in the wind direction of drift during sowing in 2008 and in experiments from 2009 onwards additionally in directly adjacent flowering crops.

Maize of different seed quality (Tab. 1) was sown in every year from 2009 to 2012 with the same pneumatic driller (suction system) with drift deflection (90% reduced drift values). For oil seed rape in 2009 and 2011 and barley (one trial 2008 and 2012) the same pneumatic driller (air pressure system), but for one trial with barley (2008) a mechanical driller was used. Seed treatments used in maize were Poncho/Poncho Pro (0.5/1.25 mg clothianidin/seed), in oilseed rape Elado (10 g clothianidin/kg seeds) and in barley 2008 Manta Plus (35 g imidacloprid/100 kg seeds) and barley 2012 Gaucho 600 FS (35 g imidacloprid/100 kg seeds). The quality of seed treatment was determined using the Heubach method (HEIMBACH, 2008) and the content of the active substance was analysed in dust sampled on the filter of the Heubach filter body. These quality criteria regarding dust abrasion and residue content in dust (Tab. 1) varied depending on the seed batches used. Sowing width was 40–60 m (except 100 m in 2008) and different sowing densities were used (maize usually 100,000 seeds/ha, rape about 700,000 seeds/ha, barley about 140 kg/ha) in most experiments. The soil was quite dry during experiments (between 4 and 23% soil water). Sowing took place at wind speed conditions of 2–5 m/sec. Wind direction varied between experiments and was almost to 90° into the direction of the adjacent areas used for the deposition except the maize drill experiment carried out in 2012 (where it was about 45°).

In 2008, ten Petri dishes (of 143 cm<sup>2</sup>) per distance (with a water/acetone (1:1/v:v) solution) were used as replicates on bare ground placed in several distances from a drilled area of 100 m x 100 m in wind direction. From 2009 onwards the adjacent area of the field sown was divided into 2-3 replicated areas of about 50 m length covered either with winter oil seed rape (maize drill experiments) or mustard (oil seed rape or barley drill experiments) and at least 30 m long areas with bare soil, where the crop had been removed (Fig. 2). Before drilling the adjacent plants were wetted with glycerol/water (1:1/v:v) solution to create a worst case situation for adhesion of dust particles during harvest of plant material for neonicotinoid analysis. In the bare soil area dust drift was measured with 4-5 Petri dishes (143 cm<sup>2</sup> with filter paper wetted with water/acetone 1:1/v:v solution) per replicate, placed at different distances to the area sown from 0.15 m or 1 m up to 30 m. Additionally, in 3 m distance one vertical gauze net was placed (2 m height x 3.50 m length, the net wetted also with glycerol water solution) and three to five samples of 0.25 m<sup>2</sup> size per net at a height of 0.40 m–0.90 m were

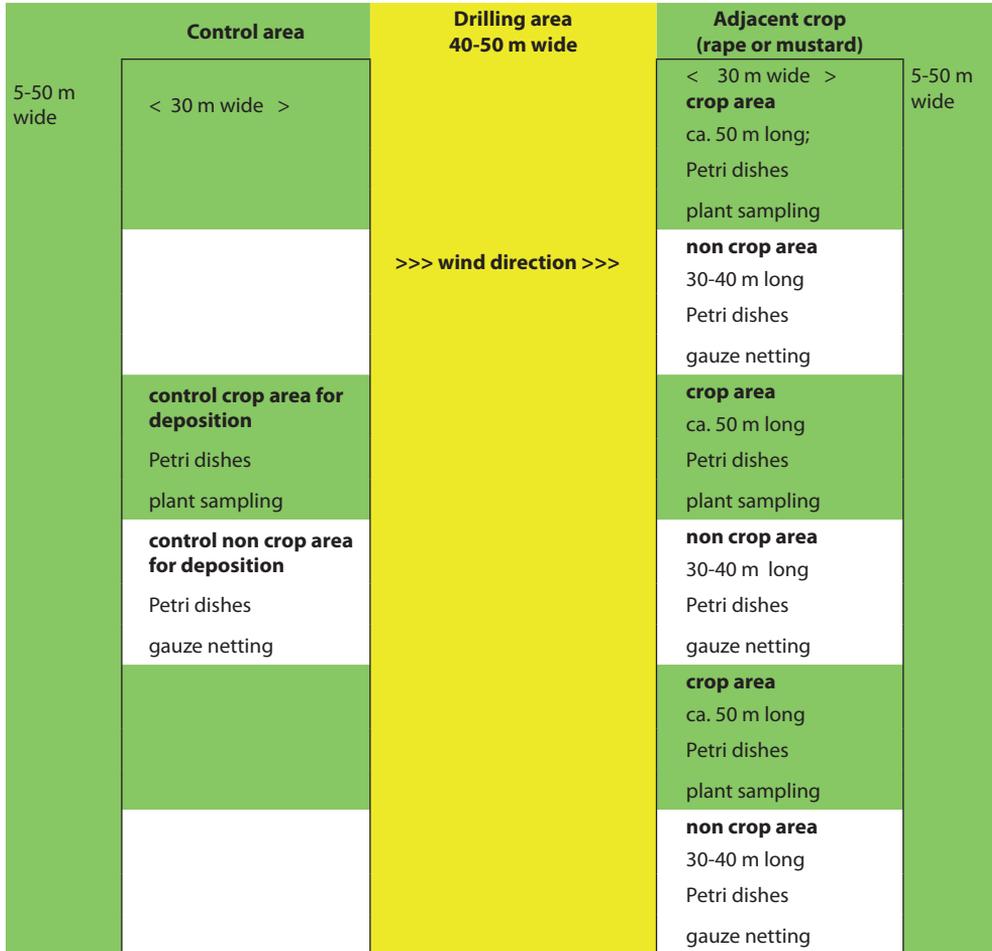
taken above ground level for residue analysis. The filter effect of the directly adjacent vegetation was calculated after measuring the a.s. deposits on all plants harvested in 0.1 m<sup>2</sup> large areas as well as the soil deposition in Petri dishes (143 cm<sup>2</sup>, but only 61 cm<sup>2</sup> in the more dense crop *Sinapis*) within these areas (4 subreplicates for both samples per replicate and distance) and then comparing the values in Petri dishes in non crop areas to total depositions in the adjacent crop areas.

Drilling usually lasted about 1 hour. Plants, gauze nets and Petri dishes were sampled shortly after drilling and 24 hours later. Samples were deep frozen after delivery to the analytical laboratory. For the procedure of determination of insecticide contents see Fig. 1. The identification of different neonicotinoids (e.g. clothianidin and imidacloprid) in Petri dishes/filter collectors, gauze nets, and plants was carried out with a LC/MS/MS- system of Dionex UltiMate 3000 coupled to an AB SCIEX QTRAP 5500. For quantification the use of matrix-matched calibration with internal standards (acetamiprid D3 as surrogate, clothianidin D3 or imidacloprid D4) was necessary. The method was validated by conducting recovery experiments with Petri dishes, and filter inlets, gauze nets, and plants. The limits of quantification were for Petri dishes and filters 0.7 mg/ha (except in 2008 1.5 mg/ha), for gauze 4 mg/ha, and for plants parts with open flowers 1 mg/ha or the rest of the plants 0.1 µg/kg – 0.5 µg/kg per 0.1 m<sup>2</sup> according to the weight of harvested plants/m<sup>2</sup>.



**Fig. 1** Procedure for the determination of neonicotinoid concentration.

**Abb. 1** Ablaufschema für die Bestimmung der Insektizidkonzentration.



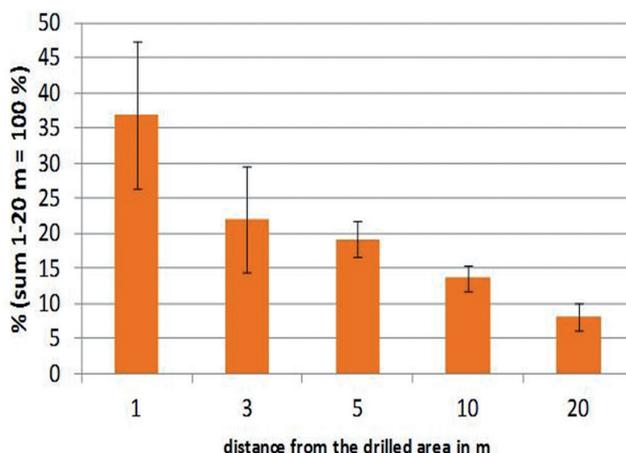
**Fig. 2** Schematic plan for drift experiments with replicated adjacent crop and non crop areas, used 2009-2012.  
**Abb. 2** Schematischer Plan der JKI Abdrift-Versuche mit wiederholten Nachbarkulturen und offenen Bereichen ohne Kultur für 2009 bis 2012.

### 3. Results and discussion

The release of dust into adjacent areas may generally be influenced by the amounts of dust set free during drilling (Heubach value), the percentage of the active substance in dust, dust particle size, amount of seeds drilled per ha, sowing width and type of sowing machinery (ANONYMOUS, 2012; BIOCCA *et al.*, 2011) and also on wind and soil conditions. The exposure of non-target organisms depends on the imission of contaminated dust and the height and density of plants as well as on the morphology of flowers and leaves and especially on the stickiness of adjacent vegetation. Therefore all these aspects need to be considered when conducting drift experiments to derive drift values for risk assessment.

Residues in Petri dishes on bare soil only decreased slowly with increasing distance in average of all experiments as well as in single experiments (Fig. 3). The mean deposition in 1 m, 3 m and 5 m distance to the drilling area was reduced to about 70% in 20 m distance in the nine field experiments with different crops carried out since 2008. Deposition a few cm off the drilling area exceeded the 1 m value clearly with a little less than twice the value of 1 m (average of four experiments). Less

but still detectable drift values were found for mechanical drilling of barley compared to pneumatic drilling in the same experiment (HEIMBACH AND STÄHLER, 2010). In all experiments some samples were taken also 24 hours after drilling but residues were always distinctly lower than shortly after drilling and are not reported here.



**Fig. 3** Mean % and std. of neonicotinoid deposition at different distances (values of 1+3+5+10+20 m = 100%) in Petri dishes placed on bare soil in adjacent non crop areas of nine drilling experiments (4 x maize, 2 x rape, 3 x barley) in 2008–2012.

**Abb. 3** Konzentrationen unterschiedlicher Neonicotinoide in Petrischalen in verschieden Abständen auf offenem Boden auf der unbewachsenen Nachbarfläche in neun Drillversuchen (4 x Mais, 2 x Raps, 3 x Gerste) von 2008 – 2012 (Werte aus 1+3+5+10+20 m = 100%).

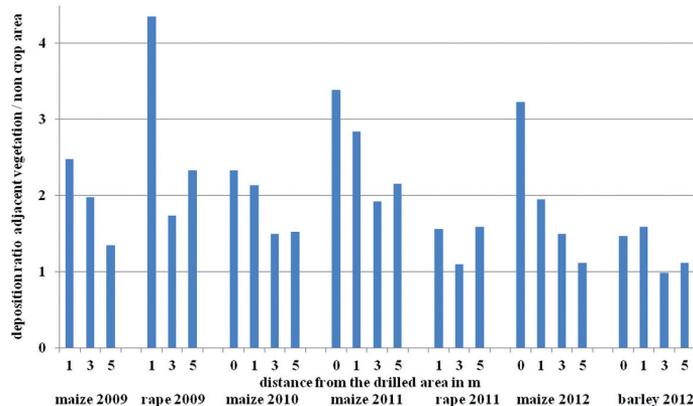
Drift values in Petri dishes on bare soil varied depending mainly on the dust abrasion potential of the seed batch used for drilling. Higher Heubach values or higher amounts of the active substance in the dust resulting in higher drift values (Tab. 1). Contents of the active substance (g/ha) in adjacent flowering plants of several experiments were up to about 4.5 times higher at 1 m distance (average of all experiments in 1 m distance 2.42 times higher) compared to values of Petri dishes on bare soil in non crop areas. The ratio of the active substance values determined on areas with vegetation compared to values on Petri dishes in areas without plants at 1 m distance from the drilling area showed, that flowering oil seed rape filtered dust and resulted in 2.95 times higher amounts in four experiments whereas this value was only 1.70 times if white mustard was adjacent in three experiments (Fig. 4). In both adjacent crops with increasing distance from the drilled area, the difference between content in vegetation and Petri dishes at the same distance decreased. This emphasizes the importance of the filtering capacity of adjacent plants especially directly at the field border when carrying out any risk analysis for organisms being active in adjacent vegetation.

**Tab. 1** Characteristics of seed batches concerning application rate and deposition of neonicotinoids in different samplers in wind direction in areas adjacent to drilling in field experiments 2009-2012. Pneumatic drillers were used for rape and barley (except 1 experiment with mechanical driller for barley) but pneumatic single corn drillers for maize.

**Tab. 1** Charakteristika von Saatgut-Chargen hinsichtlich Applikationsraten und Ablagerungen von Neonicotinoiden auf unterschiedlichen Sammelvorrichtungen angebracht in Windrichtung auf benachbarten Flächen in Aussaatversuchen in den Jahren 2009 bis 2012. Pneumatische Drillgeräte bei Raps und Gerste (außer 1 x Gerste mechanische Aussaat), pneumatische Einzelkornsähergeräte bei Mais.

Crop and year of sowing	g a.s./ha in the experiment	Heubach dust in g sown/ha (% a.s. in dust)	Heubach dust in g a.s. sown/ha	g a.s./ha (Petri dishes, mean of 1-5 m distance)	g a.s./ha (adjacent crop, mean of 1-5 m distance)	g a.s./ha (vertical gauze nets in 3 m distance)
Maize 2009	125	2.1/100,000 seeds (22.1%)	0.469	0.41	0.81	not used
Maize 2010	125	0.86/100,000 seeds (10.6%)	0.091	0.10	0.28	0.685
Maize 2011	50	0.45/100,000 seeds (19.1%)	0.086	0.15	0.27	0.447
Maize 2012	16.7	0.10/13,400 seeds (42%) untreated seeds added	0.041	0.022	0.051	0.026
Rape 2009	46	not determined	not determined	0.058	0.082	not used
Rape 2011	36	0.38/730,000 seeds (6.33%)	0.025	0.021	0.033	0.130
Barley 2008 mechanical driller	51	1.71/147 kg (7.98%)	0.136	0.030*	not used	not used
Barley 2008	47	1.57/135 kg (7.98%)	0.125	0.045**	not used	not used
Barley 2012	46	1.55/132 kg (5.55%)	0.086	0.024	0.030	0.097

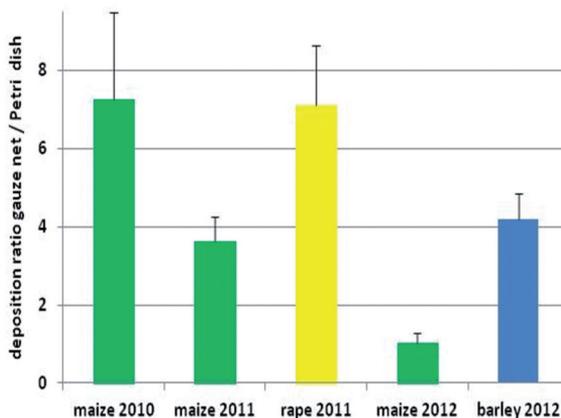
\*Corrected by the author from 0.30 \*\*Corrected by the author from 0.45 (Nov. 2nd, 2015)



**Fig. 4** Ratio of neonicotinoid deposition (at 0.1 m – 5 m distance) in adjacent crops (flowering oil seed rape after drilling of treated maize seeds, flowering white mustard after drilling of treated oil seed rape and barley seeds) and of deposition in Petri dishes placed on bare soil in adjacent non crop areas in drift experiment during sowing of maize, rape, and barley 2009-2012.

**Abb. 4** Verhältnis der Ablagerung von Neonicotinoiden (in 0,1 - 5 m Entfernung) in benachbarten, blühenden Rapsbeständen nach der Aussaat von gebeiztem Maissaatgut, in blühendem Senf nach der Aussaat von gebeiztem Raps- und Gerstesaatgut im Vergleich zur Ablagerung in Petrischalen auf offenem Boden auf unbewachsenen Flächen.

Vertical gauze nets, representing a three-dimensional structure, are easier to handle in drift experiments than adjacent vegetation and may be used to obtain a representative worst case situation of potential drift into adjacent plants. Values of the active substance in these nets ( $\text{g}/\text{m}^2$ ) were about 3 to 10 times higher compared to those in Petri dishes on bare soil, when both samplers were placed in non crop areas in 3 m distance from the drilling area in 5 experiments (Fig. 5).



**Fig. 5** Ratio of neonicotinoid deposition (mean and std. within experiments) in vertical exposed gauze nets (representing a 3-D structure) and of deposition in Petri dishes, both samplers at 3 m distance placed on bare soil in adjacent non crop areas in drift experiments during sowing of maize, oil seed rape or barley 2010-2012.

**Fig. 5** Verhältnis der Ablagerung von Neonicotinoiden (Mittelwert und Standardabweichung der a.s. in 3 m Entfernung) an vertikalen Gazenetzen (die eine 3-D-Struktur darstellen) im Vergleich zur Ablagerung in Petrischalen auf offenen Boden auf unbewachsenen Flächen nach der Aussaat von gebeiztem Mais-, Raps- oder Gerstesaatgut.

#### 4. Conclusions

Risk mitigation for pesticide treated seeds seems possible with an improvement of the seed coating quality regarding content of loose dust, maximum amount of active substances in dust and with improvements of sowing techniques (FORSTER *et al.*, 2012). There are differences between spray and dust drift (e.g. varying form and fraction size of dust particles for different crops and seed batches, evaporation of droplets but not for dust particles during drift), which need to be addressed when carrying out and interpreting dust drift experiments. For risk analysis experimental drift values need to be corrected for the maximum field rate of seeds per ha permitted and for the drilling width in an experiment if it was distinctly less than e.g. 100 m wide, because of the long travelling distance of drift.

The maize drilling in 2010 and 2011 reported here resulted in clear unacceptable effects on honey bees which were exposed in the experiments (GEORGIADIS *et al.*, 2012). A further improvement of the seed quality regarding abrasive or loose dust, a reduction of the content of the active substance in dust and an improvement of the sowing machinery is needed. More data are needed to predict realistic residue exposure values after dust drift in different types of adjacent crops and natural vegetation. In the experiments reported here, adjacent crops (flowering oil seed rape in spring or white mustard in summer) had up to 4.5 times higher residues compared to Petri dishes placed in non crop areas on the soil at the same distance. But data of vertically exposed gauze nets may represent a worst case situation to represent exposure in adjacent vegetation of different types including e.g. hedges.

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