Cross-contamination of rapeseed seeds by postharvest insecticide residues during seed storage

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Abstract

A two-year investigation in grain storage companies allowed us to follow the course of rapeseed batches from reception at the storage facility to outloading. Each of these batches was sampled outloading, and was analysed for insecticide residues content. Traceability of oilseeds established by grain-store managers allowed us to identify cross-contamination sources. Substances discovered were mostly pyrimiphos-methyl, malathion, plus chlorpyriphos-methyl and deltamethrin. Pyrimiphos-methyl was the most commonly detected active substance, and caused most cases of non-accordance with regulatory levels in rapeseed. Main cross-contamination hazard resulted from treatment of cereals at their receipt during the same period of rapeseed delivery at the grain store, especially when these cereal treatments were systematically achieved or frequent at that elevator. Another situation led to cross-contamination, but generally at a lower extent: rapeseed stored in bin that contained previously treated cereals, or loaded in empty bins with handling equipment treated before the receipt of rapeseed.

Key words: oilseed storage, cross-contamination, pesticide residues, postharvest insecticides, grain storage companies, food safety, maximum residue levels, seeds

1. Introduction

Post-harvest insecticide residues can sometimes be found on oilseeds, at low levels. But, no insecticide is allowed to be applied directly on oilseeds during storage, even if some secondary pests are found in stored oilseeds (Dauguet et al., 2005). Consequently, maximum residue levels (MRLs) allowed by European regulation are very low (mostly at the lower limit of analytical determination). No MRL existed for malathion during this study, so it should not be found beyond the analytical limit of quantification (10 $\mu g.kg^{-1}$); but since September 2008, the MRL for malathion in oilseeds is 0.02 $mg.kg^{-1}$ (European Communities Commission (ECC) regulation n° 839/2008 of 31 July 2008).

These insecticide treatments are authorised on stored cereals and corn as grain protectants, and on empty storage and handling equipment as control agents for residual insect populations in empty granaries. Pyrimiphos-methyl and still malathion were the most employed substances during this study, except during storage season 2008-2009 for malathion. Dichlorvos and malathion were forbidden since May 2008 in the EU and could be used only until 1st December 2008. As MRL for dichlorvos was newly regulated lower than 0.01 $mg.kg^{-1}$ in cereals in November 2006, this substance, which was largely used until the storage season 2006-2007, could not be used by storage companies anymore. MRL of malathion was not lowered in cereals before withdrawal late in 2008, so it could be still be used until 2007-2008.

Uptake of pyrimiphos-methyl by a single-layer of rapeseed or wheat on galvanized-steel surfaces was demonstrated in a laboratory study (Dauguet et al., 2007). It was shown that, for small bins (less than 50 tons), it could lead to residues contents above regulatory limits.

We hypothesised that a cross-contamination phenomenon can occur, between these various kinds of seeds, cereals and oilseeds, sharing the same grain handling and storage system. This phenomenon has already been demonstrated in Canada on rapeseed (White et al. 1983).

In order to improve our knowledge about this post-harvest insecticide residues cross-contamination, especially in large elevators, an investigation was carried out with the collaboration of several French grain storage companies on sunflower seeds during the storage season 2006-2007 (Dauguet, 2007). An investigation, similar to the previous one, concerned rapeseed from 2007 harvest (Dauguet, 2009). And a third investigation was carried out on rapeseed and sunflower in 2008-2009, to validate the previous findings.

The aim of this work is to check the occurrence of cross-contaminations of oilseeds by post-harvest insecticide residues and to identify the causes.

2. Materials and methods

The process used for these three surveys on oilseeds was:

- Picking up a representative mean sample of outloaded oilseed, in order to get a “final sample”, when traced batch was downloaded for sale (storage time variable from one to eight months after harvesting). The sampling method referred to standard method: Sampling method for moving seeds, for contaminant quantification, with heterogeneous distribution determination, PR EN ISO 24333: 2006. For each grain bin, 25 elementary samples for each 500-Metric-tonnes-batch evenly distributed during the outloading of the grain bin.
- Filling a questionnaire called “traceability from receipt to outloading” in order to record the storage practices and identify cros-contaminations causes.
- Determination of insecticide residues content in final samples by the ITERG’s laboratory (French Institute for Fats and Oils, Pessac, France) in 2007 and 2008, and by the CETIOM’s laboratory (Ardon, France) in 2009. All analyses were performed by GC-NPD (organophosphorus) or GC-ECD (organochlorids and pyrethrinoids).

3. Results

3.1. Residues content in oilseeds and MRL

Twenty-two rapeseed samples were analyzed in 2008 and thirty-two in 2009 : analytical results are summarized in table 1.

Table 1: Insecticide residue contents in µg.kg^{-1} (LQ: limit of quantification, MRL: maximum residue limit, n: samples number) for two storage seasons

Investigation	Substances	LQ	MRL	Mean	Median	Maximum	St. Dev.	% samples ≥ LQ	% samples > MRL
Rapeseed 2007-2008 (n=22)	Pirimiphos-methyl	10	50	130	22	1117	266	55%	32%
	Malathion	10	-	19	0	322	69	18%	18%
	Chlorpyriphos-methyl	10	50	3	0	31	9	9%	0%
	Deltamethrin	10	100	1	0	13	3	5%	0%
	Dichlorvos	10	10	-	-	-	-	0%	0%
Rapeseed 2008-2009 (n=32)	Pirimiphos methyl	10	50	16	0	212	5	37,5%	9.4%
	Malathion	10	20	0	0	13	2	3,1%	0%
	Chlorpyriphos methyl	10	50	1	0	24	40	6,3%	0%
	Deltamethrin	10	100	3	0	64	2	6,3%	0%
	Dichlorvos	10	10	-	-	-	-	0%	0%

The insecticide active substances that are used on cereals or for storage facilities treatment that were detected on rapeseed, were: pyrimiphos-methyl, malathion, chlorpyriphos-methyl and deltamethrin. The most frequently detected substance was pyrimiphos-methyl, that was recovered at a content over the quantification lower limit in 55% of samples in 2008 and 37.5% in 2009 (Table 1). This substance also caused most cases of non-accordance with the MRL, in 32 % of the samples in 2008 and 9.4% in 2009. Mean measured pyrimiphos-methyl contents decreased between 2008 and 2009 from 130 to 16 µg. kg^{-1}.

Dichlorvos was not found anymore since 2007-2008 because of new regulation. Malathion, which caused non-accordance with MRL in 2008 (18% of samples of rapeseed in 2008), was not found anymore in 2008-2009 because of the new regulation.

3.2. Traceability of cross-contamination situations

Five cases leading to cross-contamination were identified:

- K1: treatment of cereals at outloading, just before outloading oilseeds
- K2: outloading of cereals, treated at their receipt, just before outloading of oilseeds
- K3: storage of treated cereals in the same bin just before storage of oilseeds
- K4: treatment of empty bin and handling equipment before receiving oilseeds
- K5: receipt of oilseeds at the same time that cereals treated at receipt (only rapeseed)
- K0 : when none situation leading to cross-contamination is identified

We identified for each sample which cases occurred thanks to traceability questionnaires (Table 2). For one sample, one or several cases could be identified.

Table 2: Distribution of the five cases of cross-contamination

	Storage season 2007-2008			Storage season 2008-2009		
Cases	% in all samples	% in samples with residue content > LQ	% in samples with sum of residue content > 50	% in all samples	% in samples with residue content > LQ	% in samples with sum of residue content > 50
K0	9%	8%	0%	28%	0%	0%
K1	9%	15%	11%	3%	7%	0%
K2	45%	54%	56%	34%	53%	60%
K3	14%	8%	11%	13%	27%	20%
K4	36%	23%	11%	22%	33%	0%
K5	41%	69%	89%	34%	47%	40%

4. Discussion

It appears that the highest cross-contamination level on rapeseed occurred with the situation K5 (1117 $\mu g.kg^{-1}$ of pirimiphos-methyl residues). This case is specific for rapeseed, which is harvested at the same seasonal period than cereals (wheat, barley) during June-July. Most samples with pyrimiphos-methyl above MRL are in the situation K5 in 2007-2008. Looking at each sample, the highest contaminations occurred when treatments on cereals at receipt were systematically carried out. The occurrence of cross-contamination due to a treatment of cereal batches immediately at their delivery increased in 2007-2008 because dichlorvos was already banned. In the 2007-2008 survey, 81% of cereal deliveries were systematically or occasionally treated at their receipt. In the 2008-2009 investigation, 75% of the delivered cereal grain batches were systematically or occasionally treated at their receipt in silo. It was also shown that treatment of cereals at receipt could also lead to cross-contamination situations K2 and K3, which were also frequent on rapeseed in 2008-2009 survey.

The situation K5 can also be linked to a problem with insecticide application equipment:

- leak of insecticide from treatment system,
- cereal treatment not stopped after handling circuit emptying of (leading to direct accumulation of residues on empty handling equipment),
- possible delay for switching off cereal treatment before loading the handling system with a rapeseed batch received just after several cereal batches.

These situations could not be checked by our investigation.

The situation K1 was less frequent than the situation K5, but can also lead to cross-contaminations.

The case K4 caused few problems, except if it was associated to other risky situations.

5. Conclusion

Our study in real situations showed that oilseeds cross-contaminations by post-harvest insecticide residues occurred and can lead to residues contents above the regulatory limits. The highest contamination risk for rapeseed appeared when cereals were systematically treated at receipt, in the same time than rapeseed receipt. Other identified cases can also lead to lower contamination. But, grain store managers have to solve the situations where several risky cases are occurring at the same plant, which can increase the risk level: K2 and K5 for example, often associated.

Nevertheless, other sources of insecticide residues can occur in storage facilities that we could not check in this investigation, as insecticide leak by the application equipment.

Treatment strategy moved from curative to preventive position for the three years of investigation: the withdrawal of dichlorvos led to stop treatments usually performed at cereal grain batch outloading in the past years. This ban of dichlorvos had adverse indirect effect in leading to preventive strategies with more frequent treatments of cereal grain batches at their delivery with persistent organophosphates for preventive protection against storage insect pests.

So, in order to reduce cross-contaminations, we can advise to avoid sharing same receipt circuits when cereals are systematically treated, and to avoid accumulation of risky situations. It is also very important to periodically check the proper functioning of insecticide treatment equipment. This investigation allowed us to make the storage companies aware about this issue, and to help them to understand how cross-contaminations can occur in their silos and how to avoid them, knowing that each silo is different of the others.

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