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Determination of the Fipronil content in eggs

Report on the proficiency test organised by the JRC

Corrected version
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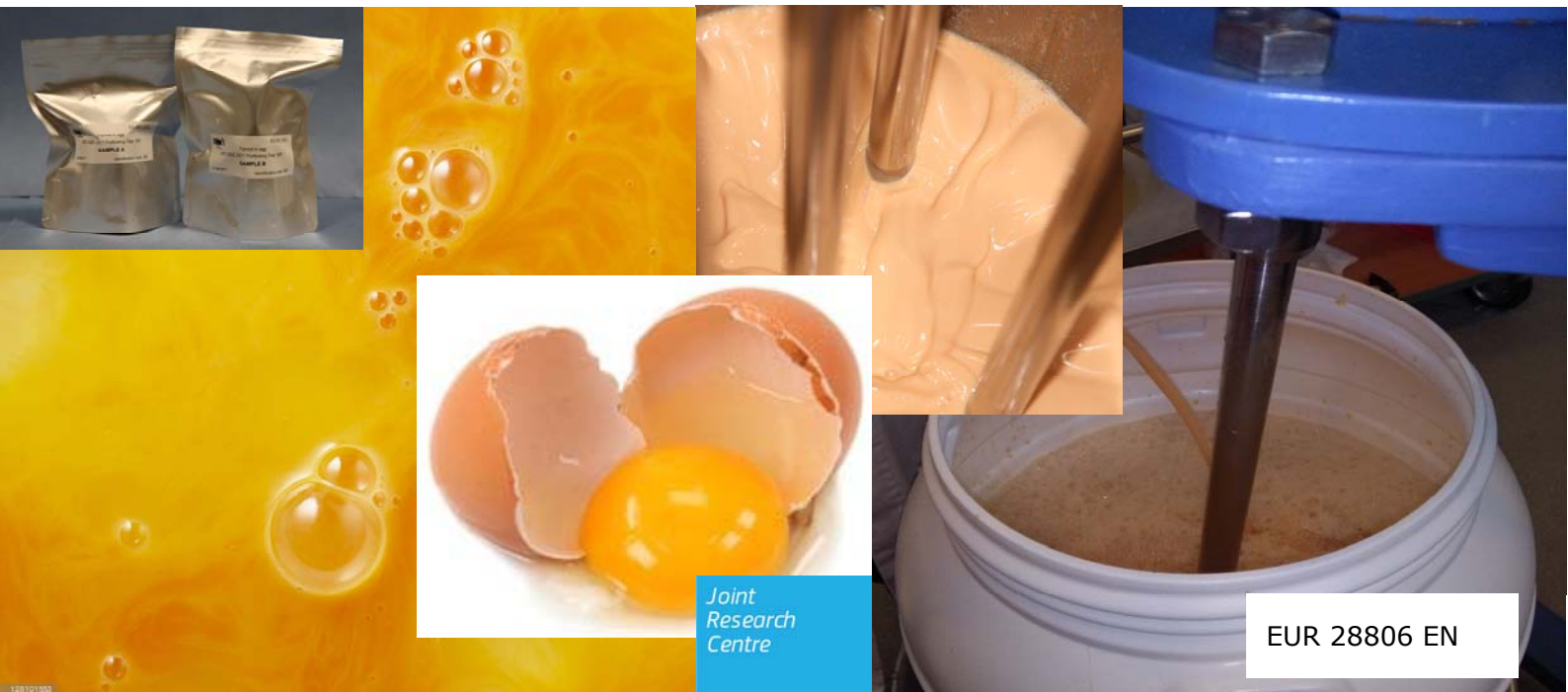

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Erratum

The laboratory codes in Annex 6 are corrected. The codes that were changed have an orange background (see Pages 29, 30, 32-34).

In addition the BELAC logo was added on Page 1

Dr. Stefanka Bratinova (*e-signed*)

PT coordinator

Executive summary

This report presents the results of the proficiency test (PT) organised by the European Commission's Joint Research Centre (JRC) - in agreement with DG SANTE - following a request by the Belgium Government on 16/08/2017. The aim of this PT is to to assess the competence of Official Control Laboratories (OCLs) and National Reference Laboratories (NRLs) in the EU Member States for the determination of the content of the insecticide fipronil in eggs around the regulated Maximum Residue Level (MRL). Two well-characterised, homogeneous and stable sets of samples of frozen liquid eggs were prepared and distributed to participants for analysis, in order to evaluate their capability to identify properly non-compliant food commodities.

The PT was announced on the JRC website on 22/08/2017. Eighty-five NRLs and OCLs from 22 EU Member States, Norway, Serbia and Albania participated to the exercise. Samples were dispatched to participants on 25-27/09/2017, and the deadline for reporting results was set to 12/10/2017.

The liquid egg starting material was purchased from a local supermarket. The material was processed and characterised at the the JRC facilities in Geel. The assigned values for the content of fipronil and fipronil sulfone, respectively (as well as the sum content of these compounds, expressed as fipronil) as well as the corresponding measurement uncertainties have been derived from the gravimetrical formulations, i.e. independently from the participants' results. They were further confirmed by in-house measurements using liquid- or gas chromatography with tandem mass spectrometry.

The participants were free to choose their method of analysis. The reported results were evaluated following the procedures of the JRC Unit F.5 - Food and Feed Compliance, which is accredited for the organisation of PTs according to the international standard ISO/IEC 17043:2010. The performance of the participating laboratories in determining fipronil in the test material was expressed as z scores.

The vast majority of the participants (94 %) obtained satisfactory z scores, thus **confirming the analytical capability of most of the participating NRLs and OCLs to enforce the European Regulations** (EC) 396/2005 and 1127/2014 setting maximum residue levels of pesticides (including fipronil) in or on food and feed of plant and animal origin. The laboratories had also to report their measurement uncertainties in μg/kg. It should be noted that 20 laboratories reported seemingly erroneous uncertainties (expressed in %), and that 14 laboratories did not report uncertainties at all. However, 43 % of the participants set their measurement uncertainty to the maximum tolerable uncertainty (of 50 %) recommended for regulatory compliance assessment.

79 participants (out of 85) correctly classified one of the test materials as non-compliant. Most of them provided a proper justification for their compliance statement, while nine of them did not submit a justification.

268-PT Accredited by the
Belgian Accreditation Body (BELAC)

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1. Introduction

The Belgian Authorities contacted the Joint Research Centre (JRC) in the frame of the Fipronil crisis on the 16/08/2017. They requested to organise a European proficiency test among food testing laboratories to guarantee the quality of the performed measurements. In agreement with DG SANTE the JRC committed to organise a dedicated proficiency test (PT) for Official Control Laboratories (OCLs) in the EU.

The current Fipronil case involves the spread of insecticide contaminated eggs and egg products in EU Member States and outside Europe. According to the RASFF (Rapid Alert System for Food and Feed) alert triggered by the Belgian authorities chicken eggs were found to contain from 0.0031 to 1.2 mg/kg Fipronil in eggs [1]. Several hundred farmers, food producers and supermarkets pulled millions of eggs off the shelves.

Fipronil is an authorised broad spectrum insecticide for the use as a plant protection product. The World Health Organization (WHO) has classified Fipronil as a moderately hazardous class II pesticide. It is toxic by oral, inhalation and dermal acute exposure. It is currently banned by the EU to treat animals destined for human consumption. The European Commission has set a Maximum Residue Level (MRL) for Fipronil in eggs and poultry meat of 0.005 mg/kg (or 5 μg/kg) in Commission Regulations (EC) 396/2005 and (EU) 1127/2014 [2]. This limit includes the sum of Fipronil and its main metabolite, Fripronil Sulfone.

The JRC in Geel organised this PT to assess the competence of OCLs and National Reference Laboratories (NRLs) in the Member States for the determination of Fipronil contents in eggs around the regulated Maximum Residue Level (MRL). Two well-characterised, homogeneous and stable sets of samples of frozen liquid eggs were prepared and distributed to participants for analyses, in order to evaluate their capability to identify properly non-compliant food commodities.

This report summarises the outcome of the PT.

2. Scope

This PT aims to assess the performance of NRLs and OCLs in the determination of the contents of Fipronil (F), Fipronil Sulfone (FS) and the resulting sum (F+FS) in liquid eggs, and to evaluate the measurement uncertainty reported. Finally, the competence of participants in assessing the compliance of the samples against legislative limits was also considered.

The reported results were evaluated following the procedures of the JRC Unit F.5 - Food and Feed Compliance, which is accredited for the organisation of PTs according to ISO/IEC 17043:2010 [3].

This PT is identified by the following code: JRC-PT-2017-01

3. Setup of the exercise

3.1 Participating Laboratories

Only designated OCLs and NRLs of the EU Member States and associated countries were allowed to participate to this PT. Eighty-six laboratories registered to the PT. All except one reported results. The list of participants is provided in the Acknowledgment section.

3.2 Time frame

The PT was announced on the JRC website (see ANNEX 1) and invitation letters were sent to laboratories on the 22/08/2017 (see ANNEX 2). The deadline for registration via the EUSurvey website (see ANNEX 3) was set to 15/09/2017. Test samples were dispatched to participants on 25-27/09/2017, and the deadline for reporting of results was set to 12/10/2017. The documents sent to the participants are presented in ANNEX 4-5.

3.3 Confidentiality

The procedures used for the organisation of this PT are accredited according to ISO/IEC 17043:2010 [3] and guarantee that the identity of the participants and the information provided by them are treated as confidential.

3.4 Design of the proficiency test

Each participant received two plastic screw cap vials containing ca. 40 g of frozen eggs test material (encoded SAMPLE A and SAMPLE B). While SAMPLE A was spiked with Fipronil and Fipronil Sulfone, SAMPLE B was the unaltered starting material with the SUM of Fipronil and Fipronil Sulfone (expressed as Fipronil) far below the MRL.

The participating laboratories were requested to analyse the two test items applying the experimental method of their choice, preferably analytical procedures they would routinely use in the frame of their official control activities.

The target analytes were (1) **Fipronil**, (2) **Fipronil Sulfone** and (3) the **SUM** (Fipronil and Fipronil Sulfone) expressed as Fipronil. Participants were requested to report their results and the corresponding expanded uncertainty (specifying the coverage factor used) in **µg/kg**. Results were to be corrected for recovery.

z scores were assigned to results reported for SAMPLE A only.

Participants were requested to assess the compliance of the test items they have analysed, according to the relevant legislation. They were also asked to report - in a dedicated questionnaire (see ANNEX 6) - details of the analytical method used and the respective method performance characteristics.

4. Test materials

4.1 Preparation

Fifty 1-litre Tetrabriks of pasteurised liquid whole eggs originating from free-ranging chicken were purchased from a local supermarket. Three different production lots were included: #L5864-0; #L6279-0; and #L6235-0 (corresponding to 1; 33; and 16 packs, respectively).

An IKA-mixer (Turbotron, RS G-01-P750, Janke Kunkel, Stafufen, DE) was used on a tripod (SFH-type, Janke-Kunkel) to mix the eggs in a 60-litre polypropylene drum for 30 minutes at moderate speed to avoid foaming. The 60-litre drum was placed in a larger drum with ice to create a double jacket, thus keeping the egg slurry cold during mixing. Manual filling of about 40-g portions of liquid egg was performed using plastic syringes from BD Plastipak (Reading, UK). The mass filled was controlled on a balance to ensure that all units contained more than 40 g. A total of 250 units of the 50 mL polypropylene plastic containers were filled. All these samples were stored at -20 °C. This constituted the blank batch of SAMPLE B. Several of these samples were later analysed by (i) Gas Chromatography with Electron Capture Detection (GC-ECD) and (ii) Liquid Chromatography tandem Mass Spectrometry (LC-MS/MS) to check the absence of Fipronil and Fipronil Sulfone in SAMPLE B.

The remaining egg liquid (38.700 kg) was divided over five 25-litre plastic drums and placed at -20 °C. The eggs were thawed over-night before spiking. Similarly, units (40 g portions) of blank eggs were also thawed to prepare the highly concentrated spike in liquid egg, thus ensuring better blending efficiency of the spiked compounds into the egg bulk. The spiked egg (638.85 g) was mixed to the bulk egg (38.06 kg) for two hours in a 100-litre stainless steel drum with a Silverson GX20 emulsifier - operated at the lowest speed on the frequency converter to avoid foaming (Chesam, UK). The stainless steel drum was then placed in a larger drum with ice to create a double jacket.

The spiked egg concentrate was prepared gravimetrically in a 1-litre stainless steel vessel. At first, 50 mL of water was put in the vessel. Then the individual standard stock solutions of Fipronil (0.973 mg/g) and Fipronil Sulfone (0.993 mg/g) in acetonitrile were added in a very

small volume (~ 600 µL) to minimise the egg protein denaturation. A portion of 597.6 g of the blank egg material was then added in the vessel and stirred properly at low rotation speed to prevent the formation of foam. The target mass fractions of Fipronil and Fipronil Sulfone were set to 10 and 5 µg/kg, respectively. During mixing, lights of the laboratory room were switched off and the openings of the drum were covered with aluminised plastic sheets to avoid degradation of Fipronil by light. 40 g portions of egg slurry were then filled in plastic containers as described above. A total of 248 units (SAMPLE A) were prepared and stored at -20 °C. The sample units were manually labelled and numbered in fill-order. Sample units of blank (SAMPLE B) as well as spiked liquid egg (SAMPLE A) were placed and sealed in pre-labelled aluminised pouches to prevent light exposure. The final material was stored at -20 °C to prevent any degradation. Shipment was foreseen on dry-ice so that the participants would be delivered with well-preserved frozen samples.

4.2 Homogeneity and stability

The egg test materials were investigated for "significant inhomogeneity" and for "sufficient homogeneity" according to ISO 13528:2015 [4]. LC-MS/MS was used for analysis. The method precision complied with the requirements laid down in this documentary standard.

The homogeneity experiment consists of duplicate analysis on 10 samples randomly selected along the filling sequence. The analyses were performed in random order. The test material was rated sufficiently homogenous at a sample intake of 5 g and no trend was observed. Details of the homogeneity test results are given in ANNEX 7.

The stability of the test material was evaluated following the requirements in ISO 13528:2015. Nine randomly selected samples were stored at different conditions for 3 weeks, covering the whole period of the PT exercise, from the dispatch of the test items to the end of the submission of the results. The first set of 3 samples was stored in a refrigerator (+4 °C) for 1 week (mimicking the possible temperature increase during transport), followed by 2 weeks in a freezer (-18 °C). The second set of 3 samples was stored for three weeks in a freezer at (-18 °C). The third set was stored in a deep freezer for three weeks at the reference temperature (-80 °C). After the deadline for reporting of results had expired, all 9 samples were analysed in duplicate under repeatability conditions using LC-MS/MS. No significant differences of the analyte contents of the test samples were found. Hence stability of the test samples over the whole period of the study can be assumed, provided that the recommended storage conditions were applied.

4.3 Assigned values, corresponding uncertainties, and standard deviations for proficiency assessment

Assigned values were determined for the two test items (SAMPLE A and SAMPLE B).

Test samples (5 g) of homogenised eggs (SAMPLE A or SAMPLE B) were subjected to acetonitrile extraction – after the addition of an isotopically labelled internal standard ($^{13}C_4$-Fipronil). The extraction was carried out using a wrist-arm shaker and samples were centrifuged. For the determination of Fipronil and Fipronil Sulfone by LC-MS/MS, the supernatant was filtered through a paper disk before analysis. When GC-MS/MS was to be used, the supernatant was filtered through paper filter with sodium sulfate and the filtrate was evaporated to dryness. The extract was then cleaned-up by gel permeation chromatography before the GC-MS/MS determination. The experimental results obtained by the two analytical methods (Table 1) indicate that:

- No Fipronil could be detected in SAMPLE B;
- The amount of Fipronil spiked in SAMPLE A was experimentally confirmed by LC-MS/MS and GC-MS/MS. Hence, the gravimetric value and the corresponding measurement uncertainty were set as the assigned value $x_{pt}(F)_A$ and uncertainty contribution due to the characterisation $u(x_{char}(F)_A)$;

- A non-negligible amount of endogenous Fipronil Sulfone ($x(FS)_B$) was detected in SAMPLE B by LC-MS/MS and GC-MS/MS. This amount was then added to the spiked amount in SAMPLE A ($x_{spike,A}$) to derive the assigned value: $x_{pt}(FS)_A = x(FS)_B + x_{spike,A}$. The corresponding uncertainty contribution was estimated by propagating the respective uncertainties;
- The total amount of Fipronil Sulfone in SAMPLE A determined by LC-MS/MS and GC-MS/MS confirmed the expected content as calculated above ($x_{pt}(FS)_A$).

The associated standard uncertainties of the assigned values ($u(x_{pt})$) are calculated following the law of uncertainty propagation, combining the standard measurement uncertainty of the characterization (u_{char}) with the standard uncertainty contributions from homogeneity (u_{hom}) and stability (u_{st}) studies, in compliance with ISO 13528:2015 [4]:

$$u(x_{pt}) = \sqrt{u_{char}^2 + u_{\text{hom}}^2 + u_{st}^2} \qquad \text{Eq. 1}$$

The uncertainty contribution deriving from the homogeneity study (u_{hom}) was calculated using SoftCRM [5] (Table 1).

The study confirmed that the material was stable and the corresponding uncertainty contribution was set to zero (u_{st} = 0) for all analytes.

The uncertainty of the assigned value for the SUM, $u(x_{pt})_{SUM}$, has been calculated applying the law of error propagation of the uncertainties of the individual parameters [6] (Table 1):

$$u(x_{pt})_{SUM} = \sqrt{u(x_{pt})_F^{\,2} + u(x_{pt})_{FS}^{\,2}} \qquad \text{Eq. 2}$$

The standard deviation for proficiency assessment, σ_{pt}, was set by using a maximum tolerated standard uncertainty of 25% following the SANTE/11945/2015 Guidance document on analytical quality control and method validation procedures for pesticide residues analysis in food and feed [7].

5. Evaluation of laboratory performance

5.1 General

The individual laboratory performance was expressed in terms of z scores according to ISO 13528:2015 [4]. They were calculated for the results reported for Fipronil (F), Fipronil Sulfone (FS) and the SUM (F+FS, expressed as Fipronil) in SAMPLE A. No scores were computed for results related to SAMPLE B.

The **"reference" assigned values were independent from the results reported by participants**. They derive from the gravimetric preparation.

The results as reported by participants are listed in ANNEX 8. The statistical evaluation was performed using the PROLab® software [8].

Table 1: Assigned values (x_{pt}), relevant uncertainties, and standard deviation for proficiency assessment (σ_{pt}). The experimental results obtained by LC-MS/MS and GC-MS/MS are also presented.

		Fipronil (F)	Fipronil Sulfone (FS)	SUM (F+FS) *expressed as Fipronil*
SAMPLE A	Gravimetric value x_{spike} (µg/kg)	10.39	5.62	
	$u(x_{spike})$, k=1 (µg/kg)	0.03_3	0.05_4	
	Assigned value, x_{pt} (µg/kg)	10.39	6.73 [(a)]	16.88
	u_{char}, k=1 (µg/kg)	0.03	0.13 [(b)]	
	u_{hom}, k=1 (µg/kg)	0.14 (1.3 %)	0.11 (1.6 %)	
	$u(x_{pt})$, k=1 (µg/kg)	0.14	0.17	0.22
	Expanded uncertainty $U(x_{pt})$, k=2 (µg/kg)	0.28	0.34	0.44
	σ_{pt} (µg/kg)	2.60	1.68	4.22
	σ_{pt} (in %)	25 %	25 %	25 %
	$u(x_{pt})/\sigma_{pt}$	0.05	0.10	0.05
SAMPLE B	Assigned value, x_{pt} (µg/kg)		1.11	
	u_{char}, k=1 (µg/kg)		0.12	
	LC-MS/MS (µg/kg), k=2		1.10 ± 0.25	
	GC-MS/MS (µg/kg), k=2		1.12 ± 0.22	

(a) $x_{pt}(FS)_A = x_{pt}(FS)_B + x_{spike}(FS)$ = 1.11 + 5.62 (µg/kg)

(b) $u(x_{pt}(FS)_A) = \sqrt{0.12^2 + 0.054^2}$ (µg/kg)

5.2 Evaluation parameter

z scores are calculated as follows:

$$z = \frac{(x_i - x_{pt})}{\sigma_{pt}} \qquad \text{Eq. 3}$$

where x_i is the measurement result reported by the laboratory; x_{pt} is the assigned value; and σ_{pt} the standard deviation for proficiency assessment (set to 25 % of the assigned value).

The interpretation of the z score is done according to ISO 13528:2015 [4]:

$|z| \leq 2.0$ = satisfactory performance
$2.0 < |z| < 3.0$ = questionable performance
$|z| \geq 3.0$ = unsatisfactory performance

The z scores compare the participant's deviation from the assigned value with the deviation for proficiency assessment (σ_{pt}) used as common quality criterion.

5.3 Evaluation of results

Each participant had to report a total of 6 results, obtained by applying their analytical method of choice. Two participants reported two sets of results obtained with two different analytical methods, while one participant did not report any results. Consequently, a total of 87 datasets were reported.

Lab 78 reported result only for Fipronil, thus resulting in an underestimated value for the SUM parameter, due to the neglected contribution of Fipronil Sulfone.

ANNEX 8 consists of tables including the reported results and measurement uncertainties together with the corresponding z scores. Each laboratory is denoted by his unique "lab code" (from 1 to 102). The graphs for each of the investigated measurands are provided in ANNEX 9, while the corresponding Kernel density plots are included in ANNEX 10.

Furthermore, Algorithm A+S described in ISO 13528:2015 [4] was applied to compute the robust means and robust standard deviations, as additional information. The confidence intervals of the assigned values are overlapping with the confidence intervals of the robust means calculated from the results reported by the participants (see Kernel distributions in ANNEX 10). However, the robust standard deviation of the results (16-18 %) was lower than the target standard deviation for proficiency assessment of 25 % set in compliance with SANTE/11945/2015 [7]). Hence, the variation among the measurement results reported by participants is smaller than the maximum one tolerated by the legislation.

Figure 1 shows that **94.2 % of the participants obtained satisfactory z scores**. Only 5 % of the results (13 individual results reported by 6 laboratories) fall into the unsatisfactory performance range ($|z| \geq 3$).

Figure 2 confirms that 80 datasets (out of 87) are rated with satisfactory z scores (see blue bars), while the results of 4 participants for the SUM parameter (F+FS, expressed as Fipronil) were classified as non-satisfactory (red bars).

Figure 1: Histogram of z scores

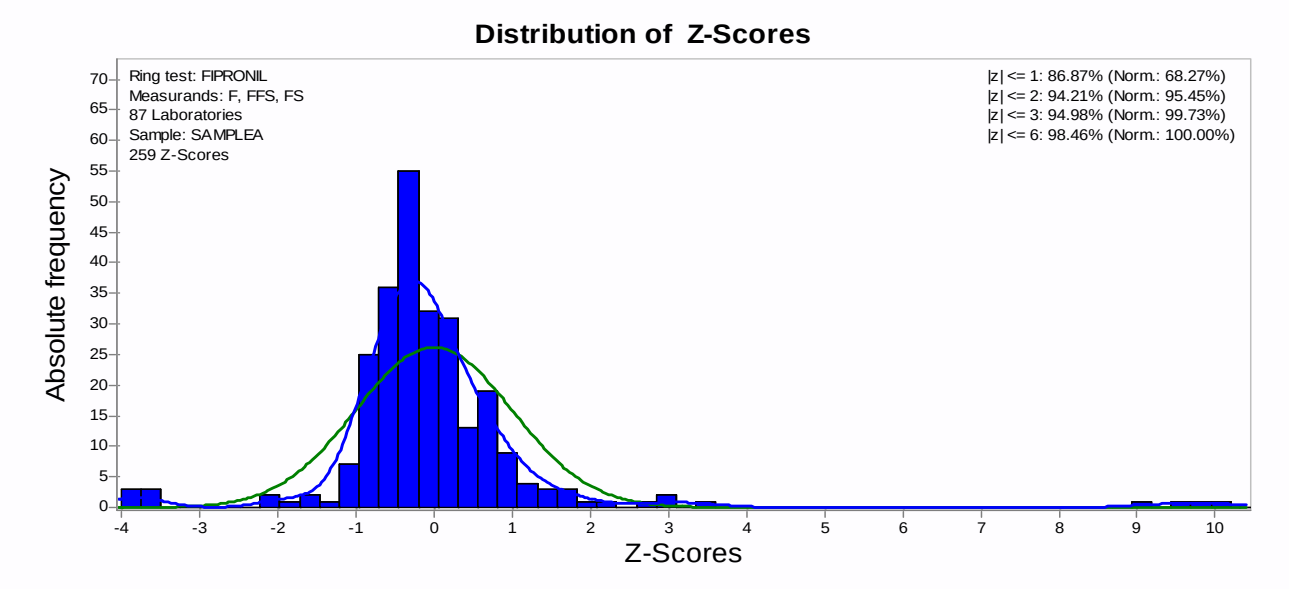


Figure 2: Graphical overview of z scores, related to results reported for SAMPLE A

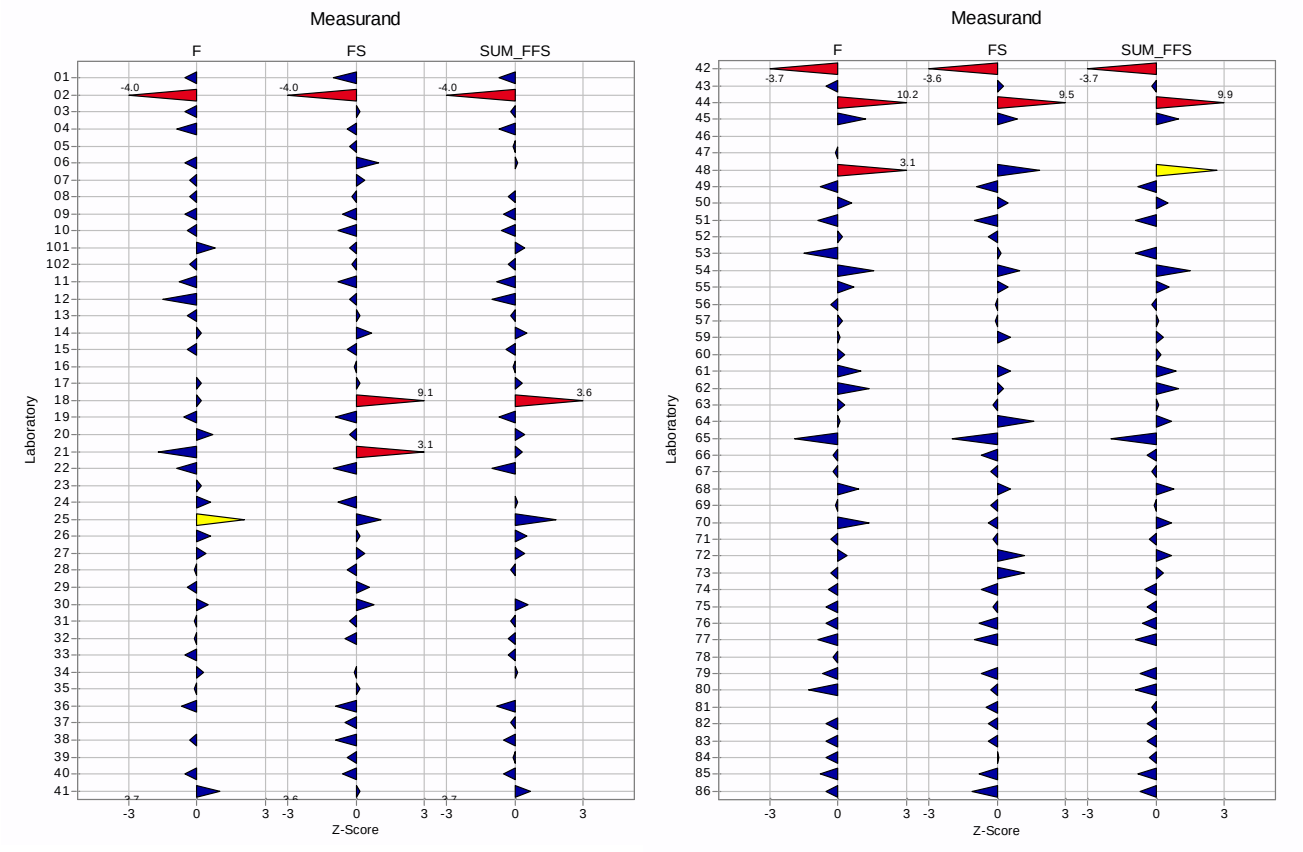


Figure 3: Youden plot: Fipronil vs Fipronil Sulfone (in SAMPLE A)

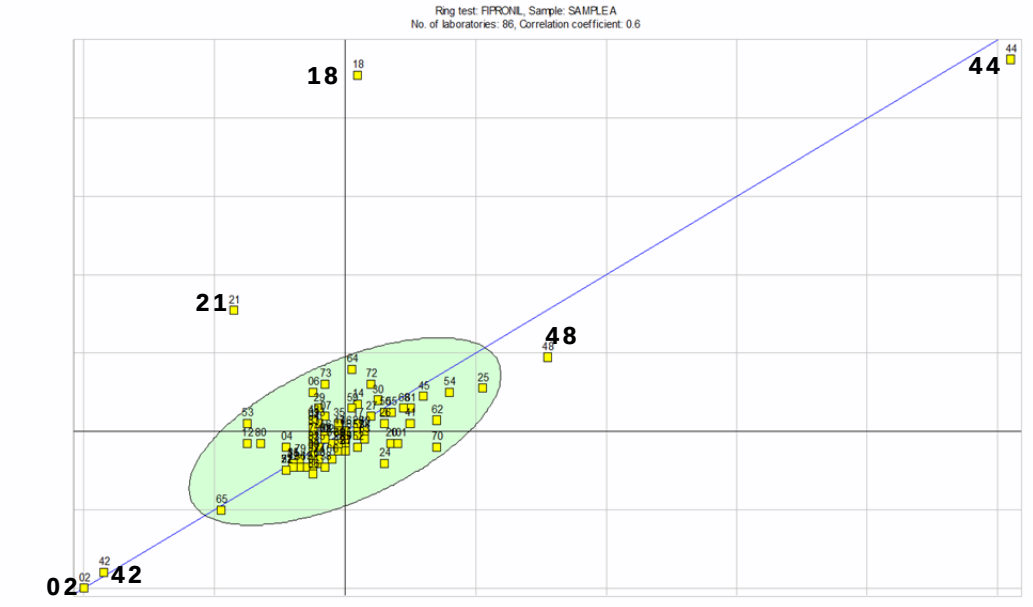


Table 2: Remarks related to poor performing laboratories

Lab	F	FS	SUM	The laboratory may have ….
02	U	U	U	… reported in other units (mg/kg instead of µg/kg); Note 1: Their compliance assessment confirms this assumption Note 2: mg/kg is the unit commonly used by the pesticide community
18		U	U	… erroneously inverted the results for Fipronil and Fipronil Sulfone
21		U		… erroneously inverted the results for Fipronil and Fipronil Sulfone
42	U	U	U	… made a calculation mistake (by one order of magnitude)
44	U	U	U	
48	U		Q	

The general remarks presented in Table 2 could be drawn from the results reported for SAMPLE A and evaluated as "Questionable" or "Unsatisfactory" (see ANNEX 8) which are also clearly identified in the Youden plot (Figure 3, outside the green ellipse).

Nevertheless laboratories 02, 18, 21, 42, 44 and 48 are advised to conduct a thorough root-cause analysis to identify the reasons of such underperformance, in order to avoid inaccurate results in the future. This is of special relevance to laboratories performing routinely such analyses (e.g. Lab 18 declared performing 100-500 sample tests in the last couple of months).

The results for the SAMPLE A are close to normal distribution (see Kernel distributions, ANNEX 10). The major mode is close to the assigned value and to the robust mean calculated from the reported results. This confirms that the measurement of Fipronil in eggs is under statistical control. No influence from the analytical techniques used (GC-MS/MS or LC-MS/MS) could be identified.

No laboratories reported quantitative results for Fipronil in SAMPLE B. Few laboratories reported results for Fipronil Sulfone in SAMPLE B (from 0.9 to 2 µg/kg) that are in agreement with the one determined by the PT provider (Table 1). However, due to the low amount present, many laboratories reported truncated values (less than) (ANNEX 8). Lab 21 reported a 10-times higher content, most probably due to a calculation mistake. Another four laboratories reported elevated values ranging from 3 to 5 µg/kg. However, having in mind that these levels are around the LOQ of the method, it is not necessary to follow this up.

5.4 Reported measurement uncertainties

Participants were requested to report their expanded measurement **uncertainties** in µg/kg (specifying the coverage factor used) (ANNEX 8).

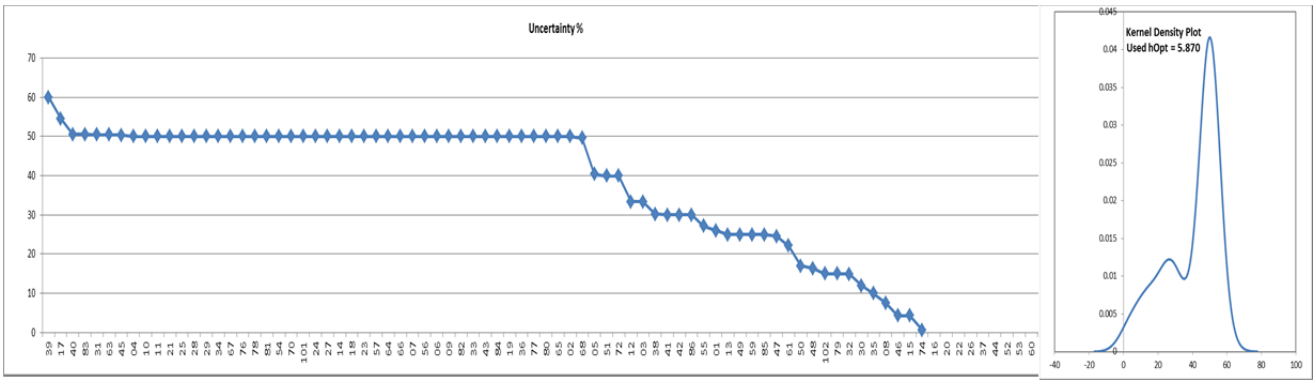
14 laboratories did not report uncertainties. 20 laboratories reported seemingly erroneous uncertainties (with uncertainty values larger than the result itself), simply because they reported in %, instead of the requested unit (see Lab codes marked with # in ANNEX 8).

ANNEX 9 displays two sets of graphs for each of the measurands: (i) with uncertainties "as reported" (i.e. Lab 4: $x(F)_A = 8 \pm \underline{50}$ (k=2) in µg/kg)) in the top graph; and (ii) with "converted uncertainties" (i.e. Lab 4: $x(F)_A = 8 \pm \underline{4}$ (k=2) in µg/kg)) in the lower graph.

Figure 4 compares the reported **relative expanded uncertainties** (after conversion to the same reporting unit). Half of the population (43) had set their uncertainty to the maximum tolerable uncertainty (of 50 %) recommended for regulatory compliance assessment without evaluating their own laboratory uncertainty. 28 laboratories made the effort to evaluate and report their measurement uncertainties.

The Kernel density plot displays a bimodal distribution with one mode at 50 %, and the second one around 25 %. Looking into the details from the questionnaire these groups could not be correlated to the use of multi-residue or target methods.

Figure 4. Graphical presentation of the relative expanded uncertainties for Fipronil, after the conversion of the reported uncertainties to the same unit.



5.5 Compliance assessment

The proper interpretation of results is as important as the accurate result itself. This proficiency test was specially designed to evaluating the capability of the participating laboratories to properly identify non-compliant food commodities in view of the recent Fipronil crisis [1].

SAMPLE A was designed to be non-compliant even at the expanded uncertainty of 50 % recommended to be used by regulatory authorities in cases of enforcement decisions (MRL exceedances) according to SANTE/11945/2015 [7]. The Fipronil content in SAMPLE A (0.017 mg/kg), resulting from the sum of Fipronil and Fipronil Sulfone (expressed as Fipronil), was higher than the maximum residue level (0.005 mg/kg) [2], even at 50 % uncertainty level.

Seventy nine participants (93 %) classified SAMPLE A as non-compliant (ANNEX 6); most of them provided the correct justification(#) for their compliance assessment, while nine of them (Labs 05, 33, 35, 57, 64, 72, 83, 84 and 86) provided no justification for their assessment.

Five participants wrongly declared SAMPLE A to be compliant: Labs 42 and 65 – based on the low measurement results reported; while Labs 11, 20, and 77 provided no justification for their assessment.

Lab 78 determined only Fipronil in SAMPLE A, and did not make any compliance assessment.

Lab 21 assessed SAMPLE B as non-compliant based on their outlying result reported.

5.6 Truncated values

Participants were not explicitly requested to report their limits of detection or limits of quantification (LOD or LOQ) together with the test results. However the low content of Fipronil Sulfone in SAMPLE B (of 1.1 μg/kg) triggered the reporting of truncated values ("less than") by many participants.

Two groups of participants can be identified (see Fipronil Sulfone in SAMPLE B in ANNEX 9): (i) 14 laboratories reported "< MRL" (of 5 μg/kg for the SUM); and 10 laboratories reported "< MRL/2"; and (ii) those that targeted their method to lower LOQ levels to be in compliance with the analytical requirements.

Most probably Labs 02 and 08 reported their "less than" values in mg/kg (0.005 and 0.0025).

(#) $x(SUM)_A - (50\ \%) * x(SUM)_A >$ MRL

5.7 Additional information extracted from the questionnaire

Additional information was gathered from the questionnaire filled in by the participants (ANNEX 6). Data are presented as reported by the participants.

Table 3 presents the overview on years of experience and number of samples analysed in the past 6 months by the laboratories. Nearly half of the participants have more than 5, or even more than 10 years of experience with the method. One third of the participants were heavily involved during the past couple of months in the analysis of Fipronil in eggs (from 100 to 1000 samples). Another third of the participants was not involved intensely in control activities in the last couple of months although they have long experience with the method. Seven of the participants declared to have no experience at all, while 21 obtained some experience after July 2017.

Table 3: Number of laboratories vs. (a) the years of experience and (b) the number of samples analysed in the last 6 months

Years of experience	Number of labs
> 10 y	12
5 – 10 y	25
2 – 5 y	15
1 – 2 y	6
0.5 y	21
none	7

Number of samples in the last 6 months	Number of labs
> 1000	4
501 – 1000	5
101 – 500	18
11 – 100	30
0 – 10	29

It should be noted that one participant (Lab 18) with a very unsatisfactory result for Fipronil Sulfone analysed 100-500 egg samples since July 2017, which might have resulted in false positive results.

51 participants were accredited for the determination of Fipronil in eggs, while 68 were accredited for the analysis of Fipronil in food in general. In 15 laboratories the applied method for the determination of Fipronil in eggs was neither validated nor accredited.

54 of the participants applied a multi-residue method for analysis, while 28 used a method targeted only to Fipronil and metabolites. No significant difference in the performance is observed for both classes of methods, neither for both major instrumental measurement techniques (LC-MS/MS with 48 datasets; GC-MS/MS with 14 datasets; and applying both methods with 13 datasets; Figure 7).

Most of the participants (54) used internal standards, but only seven of them used Fipronil ^{13}C. The majority of the internal standards consist of other pesticides or even PCBs. 27 laboratories did not use an internal standard.

Almost half of the participants prepared their calibration solutions starting from neat substances. The other half used commercial standard mixtures in a solvent.

No significant difference was noticed between the results of the different approaches.

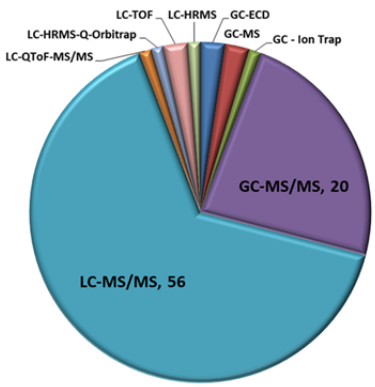


Figure 7: Method of analysis used and the corresponding number of laboratories

6. Conclusions

The JRC organised this proficiency test to assess the performance of National Reference Laboratories (NRLs) and Official Control Laboratories (OCLs) of Member States and associated countries in the determination of the content of Fipronil, Fipronil Sulfone and the resulting SUM (expressed as Fipronil) in liquid eggs. The competence of participants in assessing the compliance of the sample against legislative limits was also evaluated.

Eighty-five NRLs and OCLs (from 22 EU Member States, Norway, Serbia and Albania) reported results.

The performance has been assessed against **independent reference values**.

The main outcome of this proficiency test is the high quality of reported results. The vast majority of the participants (94 %) obtained satisfactory z scores, thus **confirming the analytical capability of most of the participating NRLs and OCLs to enforce the European Regulations** (EC) 396/2005 and 1127/2014 *setting maximum residue levels of pesticides (including Fipronil) in or on food and feed of plant and animal origin*.

Seventy-nine participants (out of 85) correctly classified SAMPLE A as non-compliant: most of them provided proper justification for their compliance statement, while nine of them provided no justification.

While requested to report measurement uncertainties in μg/kg, 20 participants reported seemingly erroneous uncertainties (expressed in %), and 14 reported no uncertainties. However, half of the participating laboratories set their expanded uncertainty to the maximum tolerable uncertainty (of 50 %) recommended for regulatory compliance assessment. This maximum uncertainty was found to be larger (less stringent) than the observed variation among the measurement results reported by participants.

Acknowledgements

The authors wish to thank colleagues from the JRC Geel for their valuable contribution to this report. The experimental contribution of Ms. Eva Jerabkova to the characterisation of the Fipronil content in the starting material is highly appreciated. Furthermore, the 86 laboratories listed hereafter are kindly acknowledged for their participation in the proficiency test.

Institute	*Country*
Food Safety and Veterinary Institute	Albania
AGES	Austria
ANALYTEC® Labor für Lebensmitteluntersuchung und Umweltanalytik	Austria
Institut Dr. Wagner Lebensmittel Analytik GmbH	Austria
AFSCA	Belgium
Bodemkundige Dienst van België	Belgium
CER Groupe	Belgium
Laboratorium ECCA NV	Belgium
LOVAP NV	Belgium
Primoris Belgium	Belgium
WIV-ISP	Belgium
SGS Bulgaria	Bulgaria
Croatian Veterinary Institute	Croatia
Euroinspekt-Croatiakontrola d.o.o.	Croatia
Sample Control d.o.o.	Croatia
State General Laboratory, Ministry of Health	Cyprus
State Veterinary Institute Prague	Czech Republic
University of Chemistry and Technology Prague	Czech Republic
Danish Veterinary and Food Administration	Denmark
National Food Institute, Technical University of Denmark	Denmark
Finnish Customs Laboratory	Finland
Finnish Food Safety Authority	Finland
ANSES - LSAl	France
INOVALYS	France
Laboratoire du SCL de Montpellier	France
Laboratoires des Pyrénées et des Landes	France
Service Commune des Laboratoires	France
Bayerisches Landesamt für Gesundheit und Lebensmittelsicherheit	Germany
Chemical and Veterinary Analytical Institute Rhine-Ruhr-Wupper	Germany
CVUA Freiburg	Germany
Federal Office of Consumer Protection and Food Safety	Germany
GALAB Laboratories GmbH	Germany
Hessisches Landeslabor (Hessian State Laboratory)	Germany
Institut für Hygiene und Umwelt, Hamburg	Germany
Labor Friedle GmbH	Germany
Landesamt für Umwelt- und Arbeitsschutz, Saarbrücken	Germany
Landeslabor Berlin-Brandenburg	Germany
Landesuntersuchungsamt, Institut für Lebensmittelchemie, Speyer	Germany
LAV Sachsen-Anhalt	Germany
LAVES -LVI Braunschweig/Hannover, Standort Hannover	Germany

LAVES, LVI Oldenburg	Germany
Lower Saxony State Office for Consumer Protection and Food Safety, Braunschweig	Germany
Lower Saxony State Office for Consumer Protection and Food Safety, Oldenburg	Germany
LUFA-ITL GmbH	Germany
State Laboratory Schleswig-Holstein	Germany
State Office for Agriculture, Food Safety and Fisheries, Rostock	Germany
Thüringer Landesamt für Verbraucherschutz	Germany
Benaki Phytopathological Institute	Greece
General Chemical State Laboratory	Greece
Food Chain Safety Centre Non-profit Ltd.	Hungary
National Food Chain Safety Office	Hungary
Department of Agriculture, Food & the Marine	Ireland
ARPA Bolzano	Italy
ARPA Lazio	Italy
Istituto Superiore di Sanità	Italy
Istituto Zooprofilattico Sperimentale Brescia	Italy
Istituto Zooprofilattico Sperimentale Cuneo	Italy
Istituto Zooprofilattico Sperimentale dell'Abruzzo e del Molise ''G. Caporale''	Italy
Istituto Zooprofilattico Sperimentale delle Venezie	Italy
Istituto Zooprofilattico Sperimentale dell'Umbria e delle Marche	Italy
Istituto Zooprofilattico Sperimentale Genova	Italy
Istituto Zooprofilattico Sperimentale della Puglia e Basilicata	Italy
Istituto Zooprofilattico Sperimentale Lazio e Toscana	Italy
National Food and Veterinary Risk Assessment Institute	Lithuania
Laboratoire National de Santé	Luxembourg
Norwegian Institute of Bioeconomy Research	Norway
National Veterinary Research Institute	Poland
Voievodship Sanitary-Epidemiological Station in Rzeszow	Poland
Voivodship Sanitary-Epidemiological Station in Warsaw	Poland
Laboratório Regional de Veterinária e Segurança Alimentar - Madeira	Portugal
Institute for Hygiene and Veterinary Public Health	Romania
Institute of Public health of Belgrade	Serbia
SP Laboratorija A.D.	Serbia
Public Health Authority of the Slovak Republic	Slovak Republic
Centro Nacional de Tecnología y Seguridad Alimentaria (CNTA)	Spain
EURL for pesticide residues in Fruits and Vegetables-University of Almería	Spain
Laboratori Agència de Salut Pública de Barcelona	Spain
Eurofins Food & Feed Testing Sweden AB	Sweden
National Food Agency Sweden (Livsmedelsverket)	Sweden
Eurofins Lab. Zeeuws-Vlaanderen B.V.	The Netherland
DUCARES	The Netherlands
Groen Agro Control	The Netherlands
Nofalab B.V.	The Netherlands
TLR	The Netherlands
Fera Science Limited	United Kingdom
Kent Scientific Services	United Kingdom

References

[1] RASFF- the Rapid Alert System for Food and Feed. https://ec.europa.eu/food/safety/rasff_en

[2] REGULATION (EC) NO 396/2005 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 23 February 2005 on maximum residue levels of pesticides in or on food and feed of plant and animal origin and amending Council Directive 91/414/EEC

COMMISSION REGULATION (EU) No 1127/2014 of 20 October 2014 amending Annexes II and III to Regulation (EC) No 396/2005 of the European Parliament and of the Council as regards maximum residue levels for amitrole, dinocap, Fipronil, flufenacet, pendimethalin, propyzamide, and pyridate in or on certain products

[3] ISO/IEC 17043:2010 "Conformity assessment - General requirements for proficiency testing providers". International Organization for Standardization, Geneva, Switzerland

[4] ISO 13528:2015 "Statistical Methods for Use in Proficiency Testing by Interlaboratory Comparisons". International Organization for Standardization, Geneva, Switzerland

[5] SoftCRM, (n.d.). http://www.eie.gr/iopc/softcrm/index.html

[6] ISO/IEC Guide 98-3:2008 "Evaluation of measurement data – Guide to the expression of uncertainty in measurement" International Organization for Standardization, Geneva, Switzerland

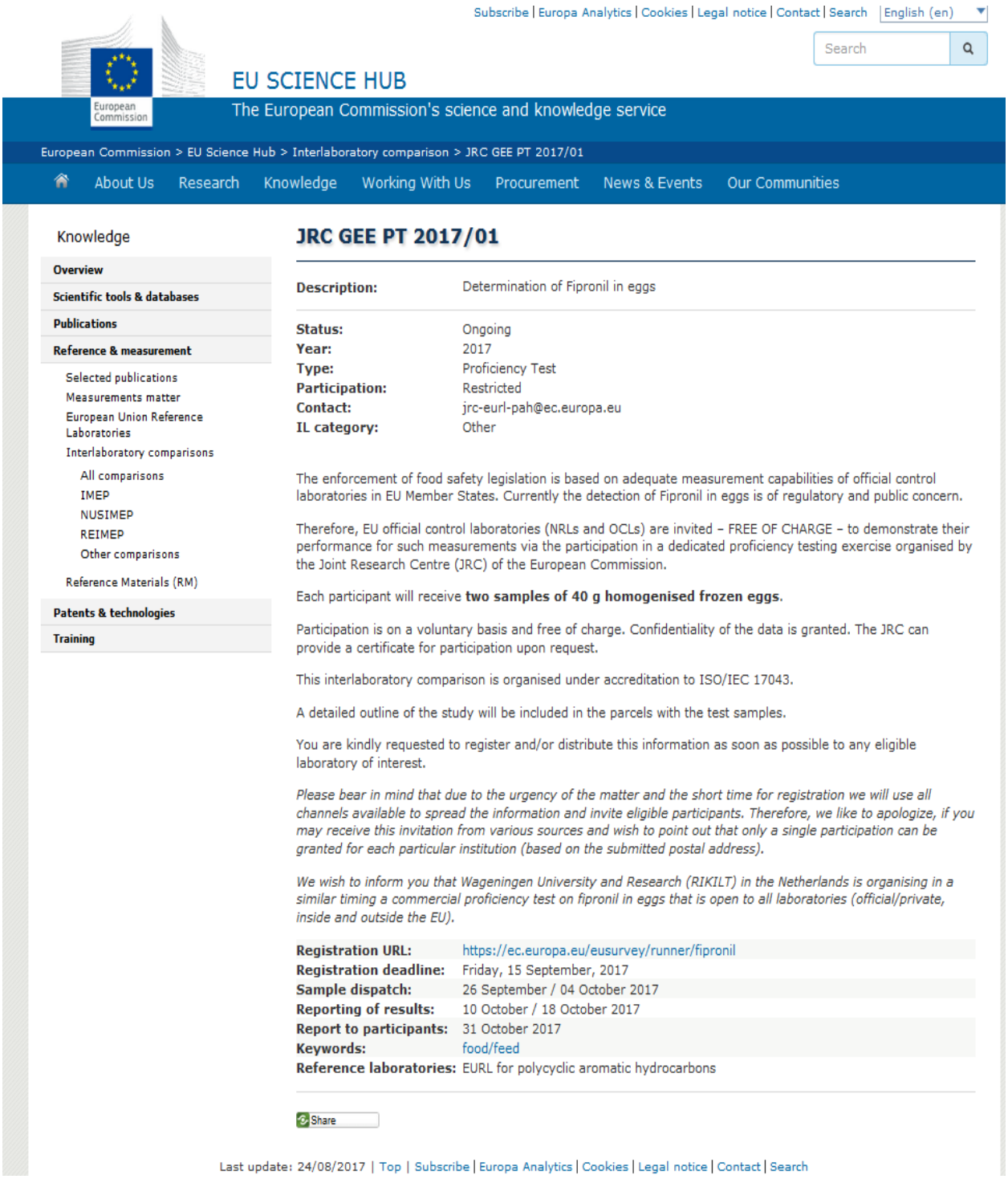
[7] Guidance document on analytical quality control and method validation procedures for pesticides residues analysis in food and feed SANTE/11945/2015

[8] PROLab, Software for PT programs and collaborative studies; http://quodata.de/en /software/for-interlaboratory-tests.html

List of abbreviations and definitions

EC	European Commission
EU	European Union
F	Fipronil
FS	Fipronil Sulfone
GC-MS/MS	Gas chromatography tandem mass spectrometry
ILC	Interlaboratory comparison
ISO	International Organization for Standardization
JRC	Joint Research Centre
LC-MS/MS	Liquid chromatography tandem mass spectrometry
LOD	Limit of Detection
LOQ	Limit of Quantification
MRL	Maximum residue level
NRL	National Reference Laboratory
OCL	Official food control laboratory
PT	Proficiency test
SUM	Sum of Fipronil and Fipronil Sulfone contents, expressed as Fipronil (also denoted FFS)

ANNEX 1: Announcement of the PT on the JRC website



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    - NUSIMEP
    - REIMEP
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- Training

JRC GEE PT 2017/01

Description: Determination of Fipronil in eggs

Status: Ongoing
Year: 2017
Type: Proficiency Test
Participation: Restricted
Contact: jrc-eurl-pah@ec.europa.eu
IL category: Other

The enforcement of food safety legislation is based on adequate measurement capabilities of official control laboratories in EU Member States. Currently the detection of Fipronil in eggs is of regulatory and public concern.

Therefore, EU official control laboratories (NRLs and OCLs) are invited – FREE OF CHARGE – to demonstrate their performance for such measurements via the participation in a dedicated proficiency testing exercise organised by the Joint Research Centre (JRC) of the European Commission.

Each participant will receive **two samples of 40 g homogenised frozen eggs**.

Participation is on a voluntary basis and free of charge. Confidentiality of the data is granted. The JRC can provide a certificate for participation upon request.

This interlaboratory comparison is organised under accreditation to ISO/IEC 17043.

A detailed outline of the study will be included in the parcels with the test samples.

You are kindly requested to register and/or distribute this information as soon as possible to any eligible laboratory of interest.

Please bear in mind that due to the urgency of the matter and the short time for registration we will use all channels available to spread the information and invite eligible participants. Therefore, we like to apologize, if you may receive this invitation from various sources and wish to point out that only a single participation can be granted for each particular institution (based on the submitted postal address).

We wish to inform you that Wageningen University and Research (RIKILT) in the Netherlands is organising in a similar timing a commercial proficiency test on fipronil in eggs that is open to all laboratories (official/private, inside and outside the EU).

Registration URL: https://ec.europa.eu/eusurvey/runner/fipronil
Registration deadline: Friday, 15 September, 2017
Sample dispatch: 26 September / 04 October 2017
Reporting of results: 10 October / 18 October 2017
Report to participants: 31 October 2017
Keywords: food/feed
Reference laboratories: EURL for polycyclic aromatic hydrocarbons

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ANNEX 2: Announcement of the PT via e-mail

Ref. Ares(2017)4155033 - 24/08/2017

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate F - Health, Consumers and Reference Materials

Geel, 24 August 2017
JRC.F.5/SB/mt/ARES(2017) 17-059

Subject: Proficiency test on the determination of Fipronil in eggs

Dear Madam, dear Sir,

The enforcement of food safety legislation is based on adequate measurement capabilities of official control laboratories in EU Member States. Currently the detection of Fipronil in eggs is of regulatory and public concern.

Therefore, EU official control laboratories (NRLs and OCLs) are invited – FREE OF CHARGE – to demonstrate their performance for such measurements via the participation in a dedicated proficiency testing exercise organised by the Joint Research Centre (JRC) of the European Commission. The following timing is planned:

- Deadline for registration: 15 September 2017
- Dispatch of samples: 26 September / 04 October 2017
- Deadline for reporting of results: 10 October / 18 October 2017
- Submission of Report: **31 October 2017**

Registration for participation is **open until 15 September 2017** via the EU Survey webpage.
https://ec.europa.eu/eusurvey/runner/fipronil

Each participant will receive **two samples of 40 g homogenised frozen eggs**.

Participation is on a voluntary basis and free of charge. Confidentiality of the data is granted. The JRC can provide a certificate for participation upon request.

This interlaboratory comparison is organised under accreditation to ISO/IEC 17043.

Information will soon be available on the website:

https://ec.europa.eu/jrc/en/eurl/pahs/interlaboratory-comparisons

A detailed outline of the study will be included in the parcels with the test samples.

You are kindly requested to register and/or distribute this information as soon as possible to any eligible laboratory of interest.

Retieseweg 111, B-2440 Geel - Belgium. Telephone: (32-14) 571 800. https://ec.europa.eu/jrc/
E-mail: jrc-eurl-pah@ec.europa.eu

Please bear in mind that due to the urgency of the matter and the short time for registration we will use all channels available to spread the information and invite eligible participants. Therefore, we like to apologize, if you may receive this invitation from various sources and wish to point out that only a single participation can be granted for each particular institution (based on the submitted postal address).

We wish to inform you that Wageningen University and Research (RIKILT) in the Netherlands is organising in a similar timing a commercial proficiency test on fipronil in eggs that is open to all laboratories (official/private, inside and outside the EU).

PT coordinators

Stefanka Bratinova & Joerg Stroka
Tel: + 32 (0)14 571800
E-mail: jrc-eurl-pah@ec.europa.eu

Should you require further clarification, please do not hesitate to contact us via:

JRC-EURL-PAH@ec.europa.eu

With kind regards,

Stefanka Bratinova
Project Officer EURL PAHs

ANNEX 3: Registration form

JRC-GEE 2017 PT on the determination of Fipronil in eggs

Fields marked with * are mandatory.

European Union Reference Laboratory
Polycyclic Aromatic Hydrocarbons

JRC-GEE PT 2017/01 - Fipronil in eggs - Registration

The enforcement of food safety legislation is based on adequate measurement capabilities of official control laboratories in EU Member States. Currently the detection of Fipronil in eggs is of regulatory and public concern.

Therefore, EU official control laboratories (NRLs and OCLs) are invited – FREE OF CHARGE – to demonstrate their performance for such measurements via the participation in a dedicated proficiency testing exercise organised by the Joint Research Centre (JRC) of the European Commission.

You are kindly requested to register and/or distribute this information as soon as possible to any eligible laboratory of interest.

We wish to point out that only a single participation can be granted for each particular institution.

IMPORTANT !

Athough the deadline for registration is 15th September, we would like to inform youi that we are reaching our limits very fast and that we would be forced to limit future registrations to not more than 10 participants

* Organisation

Department

* Address (for DHL shipment)

* City

* Postal code

* Country

* Name of the contact person

* Email

* Telephone (DHL requirement)

* Are you going to participate in the PT on determination of fipronil in eggs, organised by JRC-GEE? The dispatch is foreseen in the period 26th September - 04th October.

- Yes
- No

Which instrumental method you are going to apply

- GC/MS
- LC/MS-MS
- both

* Do you have a status of official food/feed control laboratory?

- Yes, NRL
- Yes, OCL
- other, please describe below

ANNEX 4: Instructions to participants

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate F - Health, Consumers and Reference Materials
Food & Feed Compliance

Geel, 15 September 2017
JRC.F.5/XX/xx/ARES(2017)

«Name_of_the_contact_person»
«Organisation»
«Postal_code», «City»
«Country»

Subject: JRC-GEE EURL PT Fipronil in eggs

Dear «Name_of_the_contact_person»,

Thank you for the registration in the proficiency test organised by the JRC-GEEL on the determination of the content of the fipronil in liquid eggs. It will starts with the dispatch of the samples, foreseen for the last week of September (26th-27th). Please note that some delay may occur due to unforeseen circumstances.

The target analytes are **Fipronil** and **Fipronil sulfone** expressed as **µg Fipronil//kg egg**. Each participant will be provided with two plastic screw caps vials containing a portion of frozen liquid eggs.

Outline of the study.

The participating laboratories shall apply for the analyses a method of their choice.

The laboratories shall report the results two weeks after the dispatch date at the latest, following the instructions provided later on.

The participants will be requested to report the results in **µg** Fipronil (total) /kg egg as received, corrected for recovery and accompanied by their associated uncertainties.

Additionally participants will be asked to perform compliance assessment according to the CURRENT legislative limits.

Participants will be also invited to report, together with the results, details of the applied analysis method and some method performance characteristics.

Retieseweg 111, B-2440 Geel - Belgium. Telephone: (32-14) 571 800. https://ec.europa.eu/jrc/
E-mail: jrc-eurl-pah@ec.europa.eu

Test material and analytes

Two 50 ml plastic screw caps vials containing approximately 40 g portion of frozen liquid eggs, packed in aluminium bags for light protection, labelled as" Fipronil in eggs, JRC-GEE 2017 PT 1901, Sample A - No XXX and Sample B - No XXX"

If not analysed immediately after receiving, please store the eggs test samples protected from light in freezer (-18°C).

Reporting the results

Reporting of the results will be open upon sample dispatch. The laboratories shall report the results two weeks after the sample dispatch at the latest via the ILC web interface

https://web.jrc.ec.europa.eu/ilcReportingWeb

using the participation (password) key, mailed to you after the sample dispatch.

Scoring system

The assigned values will be obtained independently from the participants results

The proficiency of the participating laboratories for the determination of the Fipronil content in eggs will be evaluated by assigned **z-scores** for the reported results. The target standard deviation of the assigned value will be set using a Fit-For-Purpose Relative Standard Deviation (FFP-RSD) approach to **25%.**

In case of questions please do not hesitate to contact:

With kind regards,

Stefanka Bratinova

PT coordinator

2

ANNEX 5. "Sample Receipt" Form

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate F - Health, Consumers and Reference Materials
Food & Feed Compliance

PROFICIENCY TESTING MATERIAL RECEIPT FORM

2017 PT- Fipronil in eggs

Contact person	
LabCode	

Content of the parcels:

1. Two plastic screw cap vials with about 40 g of frozen eggs;
2. Documents with instructions;
3. One sample receipt form (= this form), which is e-mailed as well to be filed and send electronically

TEST SAMPLE SHOULD BE STORED
PROTECTED FROM LIGHT IN A FREESER AT -18ºC

Please ensure that the items listed below have been received undamaged, and then describe the relevant statement:

Date of the receipt of the test materials	
All items have been received undamaged	YES ☐ / NO ☐
If NO, please list damaged items	

Please return the completed form to

Stefanka Bratinova

Retiesweg 111, B-2440 Geel - Belgium. Telephone: (32-14) 571 800. http://jrc.ec.europa.eu
Telephone: direct line (32-14) 571 229. Fax: (32-14) 571 3.
E- mail: jrc-EURL-PAH@ec.europa.eu or Stefanka-Petkova.BRATINOVA@ec.europa.eu

ANNEX 6. Questionnaire & Answers from participants

Lab Code	1. Is SAMPLE A compliant with the current MRL?	2. Please justify your statement.	3. Is SAMPLE B compliant with the current MRL?	4. Please justify your statement.
	1	2	3	4
01	No	Fipronil sum in SAMPLE A is greater than the current MRL	Yes	Fipronil sum in SAMPLE B is smaller than the current MRL
02	No	0.0176-(0.0176/2) >0.005	Yes	<RL=0.0025
03	No	the value obtained (0.016) is above 0.005 which is the MRL	Yes	Fipronil is below LOD and Fipronil sulfon is lower than LOQ 0.005. The sum is lower than 0.005 MRL
04	No	exceedance of MRL of concidering an expanded Measurement uncertainty of 50%	Yes	no exceedance of MRL, detected concentrations below LOD (Fipronil) and LOQ (Fipronil-Sulfone) respectively
05	No		Yes	
06	No	(RESULT - UNCERTAINTY) ABOVE MRL 5 PPB	Yes	BELOW MRL 5 PPB
07	No	Fipronil + -sulfon > 0.005 mg/kg even by consideration of the uncert. value	Yes	Fipronil + -sulfon < 0.005 mg/kg
08	No	The MRL for Fipronil and Fipronil sulfon expressed as Fipronil in egg is 0.005 mg/kg. The determined concentration in sample A-013 is above this value.	Yes	The MRL for Fipronil and Fipronil sulfon expressed as Fipronil in egg is 0.005 mg/kg. The determined concentration in sample B-030 is below this value.
09	No	Even considering the measurment uncertainty, the maximum residue limit according to the EU regulation 396/2005 is exceeded.	Yes	The maximum residue limit according to the EU regulation 396/2005 is not exceeded.
10	No	the final concentration after taking into account the MU is above MRL	Yes	the final concentration after taking into account the MU is below MRL
11	Yes	Sample A is 015	Yes	Sample B is 035
12	No	The LMR for Fipronil in eggs is 5 µg/kg. Taken into account the uncertainty, the value would be between 6.3 and 18.9 µg/kg, so above the LMR.	Yes	Fipronil was not detected for this sample. As our limit of detection is equal to LMR, sample B is compliant with the current LMR.
13	No	Sum expressed as Fipronil - Uncertainty>MRL	Yes	Sum expressed as Fipronil - Uncertainty<MRL
14	No	the measured value exceeds the MRL by more than the expanded uncertainty	Yes	the measured value not exceeds the MRL by more than the expanded uncertainty
15	No	above MRL (5 µg/kg)	Yes	below MRL (5 µg/kg)
16	No	Above MRL 5 ug/kg	Yes	Below MRL
17	No	The value found in the sample for Fipronil + Fipronil Sulfone is 0.018 mg/kg, being its MRL in egg 0.005 mg/kg for the residue definition. It is not compliant, it is almost 3 times above the MRL.	Yes	We introduce a value of 0.001 mg/kg because it is not possible to say < LOQ. Our LOQ is 0.002 mg/kg for each compound, by that we have not calculated its Uncert. Value
18	No	Fipronil sum minus 50%uncertainty is more than 5 µg/kg (MRL)	Yes	Fipronil sum is less than 5 µg/kg (MRL)
19	No	0,0139 ± 0,0070 (mg/kg) > MRL (0,005 mg/kg)	Yes	"< 0,005 mg/kg" is < MRL
20	Yes		Yes	
21	No	MRL for Fipronil+Fipronil Sulfone is 0.005 and the results exceeds that even taking off uncertainity measurement of 50%.	No	MRL for Fipronil+Fipronil Sulfone is 0.005 and the results exceeds that even taking off uncertainity measurement of 50%.
22	No	Value about MRL	Yes	
23	No	the value found (17 µg/kg) - uncertainity (8.5 µg/kg) exceeds the MRL (5 µg/kg)	Yes	the value found is below the MRL of 5 µg/kg
24	No	Result (sum of Fipronil and Fipronil Sulfone expressed as Fipronil) above the MRL taking in account MU	Yes	Result (sum of Fipronil and Fipronil Sulfone expressed as Fipronil) below the MRL
25	No	result > MRL (5 µg/kg)	Yes	result < MRL (5 µg/kg)
26	No	Current MRL = 5 µg/kg. Result found is above this limit	Yes	No Fipronil detected
27	No	Sum is >MRL, if the egg is produced after 1.1.2017, if before the sample is complaint taking into account a measurement uncertainty of 50% according to SANTE document (AQC guidelines)	Yes	<0.005 mg/kg
28	No	Considering a measurement uncertainty of 50% the determined concentrations exceeds MRL of 0,005 mg/kg.	Yes	No Fipronil or Fipronil Sulfone above LOQ were detected.
29	No	results > 0.005 mg/kg	Yes	results < 0.005 mg/kg
30	No	MRL=0.005 mg/kg, Sum of Fipronil and firpronil Sulfone (0.019 mg/kg) taking into account measurement uncertainty is > MRL.	Yes	MRL=0.005 mg/kg, Sum of Fipronil and firpronil Sulfone (<0.002) mg/kg) is <MRL.
31	No	Mrl= 5ppb. Result is greater.	Yes	Mrl=5ppb, 1.2 ppb is less than.
32	No	results were higher then the MRL	Yes	results were lower then the MRL
33	No		Yes	
34	No	17.34 - 8.67 (50% uncertainty) > MRL	Yes	result < LOQ, which is equal to the MRL
35	No		Yes	
36	No	Regarding the expanded MU of 50%, the sample is not conform the EU regulation 396/2005 and updates.	Yes	The sample is conform the EU regulation 396/2005 and updates.
37	No	16.19 µg/kg - 8.095 µg/kg (MU) > 5 µg/kg (current MRL)	Yes	Fipronil was not detected and Fipronil-Sulfone was < LOQ. LOQ < MRL of 5 µg/kg
38	No	Reg. 396/2005/EC	Yes	Reg. 396/2005/EC
39	No	>5 µg/kg	Yes	<5 µg/kg
40	No	BECAUSE (RESULT OF FIPRONIL+FIPRONIL SULFONE-U)>MRL (=0.005 mg/kg)	Yes	Both Fipronil and Fipronil Sulfone <LOQ (=0.0025 mg/kg)
41	No	The sample A is higher than the current MRL.	Yes	The sample A is lower than the current MRL.
42	Yes	MRL = 5 µg/kg	Yes	MRL = 5 µg/kg

43	No	Results exceeded MRL	Yes	
44	No	The results is far much than MRL (10 times more)	Yes	the Fipronil Sulfone is lower then <5 , and there is no Fipronil
45	No	The value obtained is >0.005 mg/Kg	Yes	
46	No	considering uncertainty the MRL is exceeded	Yes	proven content is lower than quantitation limit = MRL
47	No	The residue found is greater than the MRL set for birds eggs which is is 0.005* for Fipronil	Yes	
48	No	SUM of Fipronil+Fipronil Sulfone = 28.287 µg/kg > MRL	Yes	SUM of Fipronil+Fipronil Sulfone = 2.069 µg/kg < MRL
49	No	Our result is above National MRL (5ug/kg)	Yes	
50	No	The MRL of 0,005 mg/kg for Fipronil in chicken eggs according Reg (EC) No 1127/2014 amending Annexes II and II to reg. (EC) No. 396/2005 is exceeded even in consideration of the expanded measurement uncertainty of 50%.	Yes	The MRL of 0,005 mg/kg for Fipronil in chicken eggs according Reg (EC) No 1127/2014 amending Annexes II and II to reg. (EC) No. 396/2005 is kept.
51	No	VO(EG) 396/2005 0.005 mg/kg	Yes	VO(EG) 396/2005 0.005 mg/kg
52	No	Given the default uncertainty of 50% the sample is exceeding the MRL value of 0.005 mg/kg	Yes	The concentration in the sample is lower than the MRL-value.
53	No	The result including the U is above the MRL (0.005 mg/kg)	Yes	It is present an amount of Fipronil Sulfone but lower than the LOQ of the method (0.002 mg/kg)
54	No	Sum Fipronil and FipronilSulfone, expressed as Fipronil on the product as is (wet weight) > 5 ug/kg	Yes	Sum Fipronil and FipronilSulfone, expressed as Fipronil on the product as is (wet weight)< 5 ug/kg
55	No	The detected levels for Fipronil and Fipronil-Sulfone were >2.5 µg/kg and therefore the sum of Fipronil and Fipronil-Sulfone is above the current MRL (5 µg/kg for the sum of Fipronil and Fipronil-Sulfone)	Yes	No levels of Fipronil or Fipronil-Sulfone were detected >2.5 µg/kg so the sum of Fipronil and Fipronil-Sulfone is below the MRL (5 µg/kg)
56	No	The Result - Expanded Uncertainty > MRL	Yes	No residues found
57	No		Yes	
59	No	Result of Sum Fipronil after subtraction exp. MU exceeds MRL for egg of 0.005 mg/kg	Yes	Result of Sum Fipronil far below MRL for egg of 0.005 mg/kg
60	No	EU VO Nr. 396/2005 MRL 5 µg/kg; Sante/11945/2015 uncert. value: +/- 50% (=8,912 µg/kg> MRL)	Yes	EU VO Nr. 396/2005 MRL 5 µg/kg;
61	No	measured value minus uncertaitny above MRL according regulation nr. 1127/2014/EC (396/2005/EC)	Yes	measured value minus uncertaitny lower than MRL according regulation nr. 1127/2014/EC (396/2005/EC)
62	No	Sum of Fipronil and Fipronil Sulfone is above the MRL of 5 µg/kg. The ratio of Fipronil/Fipronil Sulfone is unusual. It is assumed that the amounts of Fipronil and Fipronil Sulfone are spiked.	Yes	Sum of Fipronil und Fipronil Sulfone is below the MRL of 5 µg/kg.
63	No	The Sample A is not compliant with the current MRL, that is 0.005 mg/kg.	Yes	The Sample B is compliant with the current MRL, that is 0.005 mg/kg.
64	No		Yes	
65	Yes	Expanded measurement uncertainty according to SANTE 11945/2015 is 50%, if we have to make decision about MRL exceedances.	Yes	Concentration of Fipronil in egg is below MRL value and below of LOQ of method
66	No	Fipronil content in sample A (0,0076mg/kg) > MRL (0,005mg/kg)	Yes	Fipronil content in sample B < MRL (0,005mg/kg)
67	No	MRL in egg 5 µg/kg (whole product basis)	Yes	level below MRL(even below our reporting limit of 2 µg/kg)
68	No	Because MRL for the sum is 0.005 mg/Kg	Yes	Because MRL for the sum is 0.005 mg/Kg
69	No	concentration is above the MRL of 5 µg/kg	Yes	concentration is lower than the MRL of 5 µg/kg
70	No	exceedance of MRL for Fipronil_total (MRL=0.005 mg/kg)	Yes	no exceedance of MRL for Fipronil_total (MRL=0.005 mg/kg)
71	Yes	because MRL is 5 µg/kg	Yes	because MRL is 5 µg/kg
72	No		Yes	
73	No	The Fipronil concentration of sample A exceeded the MRL of 0,05 mg/kg.	Yes	The Fipronil concentration of sample B was below the MRL of 0,05 mg/kg.
74	No	higher then MRL (5 µg/kg)	Yes	lower then MRL (5 µg/kg)
75	No	MRL above 0,005 mg/kg	Yes	MRL under 0,005 mg/kg
76	No	concentration of Fipronil (sum Fipronil + Sulfone metabolite (MB46136) expressed as Fipronil) in Sample A is above the MRL for bird eggs (chicken) of 0,005 mg/kg	Yes	there is no Fipronil or Fipronil-Sulfone in Sample B
77	No		Yes	
79	No	In accordance with residue definition, the sum of value of parent compound and its sulfon corrected by conversion factor is higher than the 2xMRL (due to 50% rel. uncert.)	Yes	In accordance with residue definition, the sum of value of parent compound and its sulfon corrected by conversion factor is lower than the MRL.
80	No	Sum of Fipronil and Sulfone - U above MRL	Yes	
81	No	MRL=0,005 ug/kg Commission Regulation (EU) No 1127/2014 and No 396/2005	Yes	MRL=0,005 ug/kg Commission Regulation (EU) No 1127/2014 and 396/2005
82	No	MRL 0.005 mg/kg	Yes	MRL 0.005 mg/kg
83	No		Yes	
84	No		Yes	
85	No	In accordance with residue definition, the value of Fipronil (sum of Fipronil + Sulfone metabolite (MB46136) expressed as Fipronil) is higher than the 2xMRL (due to 50% exp. uncert.)	Yes	In accordance with residue definition, the value of Fipronil (sum of Fipronil + Sulfone metabolite (MB46136) expressed as Fipronil) is lower than the MRL
86	No		Yes	A trace level of Fipronil Sulfone was detected in sample B
101	No	The value obtained is >0.005 mg/Kg	Yes	
102	No	In accordance with residue definition, the sum of value of parent compound and its sulfon corrected by conversion factor is higher than the 2xMRL (due to 50% uncert.)	Yes	In accordance with residue definition, the sum of value of parent compound and its sulfon corrected by conversion factor is lower than the MRL.

Lab Code	5. Do you have previous experience with determination of Fipronil as pesticide residue in food and how long?	6. How many egg samples have you analysed in the last couple of months?	7. Are you accredited for the quantitative analysis of Fipronil content in eggs?	8. Are you accredited for the quantitative analysis of Fipronil content in food in general?	9. Is your method multiresidue method?	10. Is the applied method for determination of the content of Fipronil (Fipronil+Fipronil Sulfone) validated?	11. What is your sample intake in grams?
	5	6	7	8	9	10	11
01	5-10 years	10-100	No	Yes	Yes	No	5 grams
02	5-10 years	10-100	No	Yes	Yes	Yes	5
03	since July 2017	10-100	Yes	Yes	Yes	Yes	5
04	5-10 years	500-1000	Yes	Yes	Yes	Yes	5
05	since July 2017	10-100	Yes	Yes	Yes	Yes	5
06	2-5 years	0-10	Yes	Yes	Yes	Yes	5 grams
07	since July 2017	100-500	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	10
08	2-5 years	0-10	Yes	Yes	Yes	No	2.5 g
09	> 10 years	500-1000	Yes	No	Yes	Yes	10
10	5-10 years	10-100	No	Yes	Yes	Yes	10 grams
11	since July 2017	10-100	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	10
12	5-10 years	0-10	Yes	Yes	Yes	Yes	5
13	since July 2017	100-500	Yes	Yes	Other	Yes	5
14	2-5 years	10-100	No	Yes	Yes	Yes	
15	> 10 years	10-100	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	
16	no	0-10	No	Yes	No, targeted to only Fipronil and methabolites	No	5
17	5-10 years	10-100	No	Yes	Yes	Yes	5
18	1-2 years	100-500	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	5
19	2-5 years	0-10	No	No	Yes	Yes	10
20	5-10 years	10-100	No	Yes	Yes	Yes	10
21	> 10 years	0-10	No	Yes	Yes	No	5g for GC-MS and 7.5g for LC-MS
22	since July 2017	0-10	No	Yes	Yes	Yes	10
23	5-10 years	0-10	No	No	Yes	No	5
24	1-2 years	0-10	No	No	Yes	Yes	10
25	1-2 years	100-500	No	No	No, targeted to only Fipronil and methabolites	No	10
26	> 10 years	100-500	Yes	Yes	Yes	Yes	10 g
27	5-10 years	100-500	Yes	Yes	Yes	Yes	5
28	2-5 years	10-100	Yes	Yes	Yes	Yes	5
29	2-5 years	10-100	Yes	Yes	Yes	Yes	5
30	since July 2017	10-100	No	No	No, targeted to only Fipronil and methabolites	Yes	7
31	2-5 years	10-100	No	Yes	Yes	No	15
32	since July 2017	0-10	No	No	No, targeted to only Fipronil and methabolites	Yes	5 g
33	since July 2017	10-100	No	Yes	Yes	No	5
34	5-10 years	0-10	No	Yes	Yes	No	5
35	5-10 years	10-100	No	No	No, targeted to only Fipronil and methabolites	Yes	5
36	2-5 years	500-1000	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	1 g (egg fat)
37	no	0-10	No	No	No, targeted to only Fipronil and methabolites	No	5
38	5-10 years	0-10	Yes	Yes	Yes	Yes	5
39	5-10 years	0-10	No	Yes	Yes	No	5
40	no	10-100	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	5
41	5-10 years	10-100	Yes	Yes	Yes	Yes	5 g
42	no	0-10	No	No	No, targeted to only Fipronil and methabolites	No	5
43	5-10 years	10-100	Yes	Yes	Yes	Yes	5
44	no	0-10	No	No	Yes	No	5
45	since July 2017	0-10	Yes	Yes	Yes	Yes	5
46	5-10 years	10-100	Yes	Yes	Yes	Yes	5
47	2-5 years	100-500	Yes	Yes	Yes	Yes	5
48	since July 2017	10-100	No	No	No, targeted to only Fipronil and methabolites	Yes	5.0 g
49	no	0-10	No	Yes	No, targeted to only Fipronil and methabolites	No	5 g

	5	6	7	8	9	10	11
50	5-10 years	100-500	Yes	Yes	No, targeted to only	Yes	5
51	2-5 years	> 1000	Yes	Yes	Yes	Yes	2
52	5-10 years	0-10	No	Yes	No, targeted to only Fipronil and methabolites	No	5
53	> 10 years	0-10	Yes	Yes	Yes	Yes	10 g
54	2-5 years	100-500	Yes	Yes	Yes	Yes	10 g
55	> 10 years	10-100	Yes	Yes	Yes	Yes	5
56	5-10 years	0-10	No	Yes	Yes	Yes	5 g
57	since July 2017	100-500	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	1 g
59	5-10 years	10-100	No	Yes	Yes	Yes	10
60	5-10 years	10-100	No	Yes	Yes	Yes	5
61	since July 2017	100-500	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	1 gram
62	since July 2017	10-100	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	
63	> 10 years	> 1000	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	5.00± 0.050
64	2-5 years	10-100	Yes	Yes	Yes	Yes	5
65	since July 2017	0-10	No	No	Yes	Yes	5
66	2-5 years	10-100	Yes	Yes	Yes	Yes	10
67	> 10 years	100-500	Yes	Yes	Yes	Yes	5,0
68	since July 2017	10-100	Yes	Yes	Yes	Yes	5.0
69	since July 2017	0-10	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	5
70	> 10 years	500-1000	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	5.0
71	5-10 years	0-10	Yes	Yes	Yes	Yes	5
72	5-10 years	> 1000	Yes	Yes	Yes	Yes	2,5
73	1-2 years	500-1000	Yes	Yes	Yes	Yes	1
74	since July 2017	100-500	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	5 gram
75	no	0-10	No	No	No, targeted to only Fipronil and methabolites	Yes	5
76	since July 2017	10-100	Yes	Yes	Yes	Yes	5 g
77	5-10 years	> 1000	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	2.5
79	> 10 years	0-10	Yes	Yes	Yes	Yes	5
80	1-2 years	10-100	Yes	No	No, targeted to only Fipronil and methabolites	Yes	5
81	2-5 years	100-500	Yes	Yes	No, targeted to only Fipronil and methabolites	Yes	6g
82	2-5 years	100-500	Yes	Yes	Other	Yes	5
83	1-2 years	100-500	No	Yes	Yes	Yes	5
84	since July 2017	100-500	Yes	No	Yes	Yes	10
85	> 10 years	100-500	Yes	Yes	Yes	Yes	5
86	5-10 years	0-10	No	Yes	Yes	Yes	5
101	since July 2017	0-10	Yes	Yes	Yes	Yes	5
102	> 10 years	0-10	Yes	Yes	Yes	Yes	5

Lab Code	12. What are the extraction solvent and conditions (solvent type, volume, T, time?	13. What type of clean up you performed if any?	14. Which other metabolite and degradation products of Fipronil you analyse?	15. What type of measurement techniques you applied?
	12	13	14	15
01	Acetonitrile, 10 mL; 25ºC; 20 min	Magnesium sulfate and PSA	Fipronil Sulfone and Fipronil desulfinyl	GC-MS/MS
02	acetonitrile, 10 mL, ambient temp, 1 min	900 mg MgSO4, 150 mg PSA	Fipronil desulfinyl	LC-MS/MS
03	QuEChERS method		None	LC-MS/MS
04	extraction solvent 10ml acetonitril, water addition (10ml), extraction time 20min, RT	no	Fipronil-desulfinyl, Fipronil-sulfid	LC-MS/MS
05	CH3CN, 10 ML, 10 min QuEhERS, citrate-buffered	PSA after freezing	Fipronil-Sulfid, Fipronil-Desylfinyl	LC-MS/MS
06	acetonitrile 10mL, ambient, 30 minutes	PSA, MgSO4, C18	none	LC-MS/MS
07	Acetonitril, 10 ml, ambient, 10 min	EMR (Agilent)	no	LC-MS/MS
08	ethyl acetate, 10 ml, ambient temperature, 1 hour	clean up as dispersive SPE with PSA and C18	Fipronil-sulfide, Fipronil-desulfinyl	GC-MS/MS, LC-MS/MS
09	ACN, 10ml, Quechers procedure	PSA, MgSO4	Fipronil-sulfid, Fipronil-desulfonyl and Fipronil-carboxamid	GC-MS/MS
10	extraction solvent: ethyl acetate	GPC clean up/filtration	Fipronil Sulfone	LC-MS/MS
11	ACN, 10 ml, room temperature, 2 min	1. MgSO4+NaCl 2. MgSO4+PSA	none	LC-MS/MS
12	Our extraction solvent is acetonitrile (10 mL) at ambient T. Shaking time is 1 minute.	d-SPE PSA	none else	other
13	5 ML ACETONITRILE	Quechers (PSA; C18, MgSO4)	Fipronil Sulfone	LC-MS/MS
14	acetonitrlile, 10 ml,	QuEChERS	Fipronil- Sulfone,Fipronil- desulfinyl	GC-MS/MS, other
15				LC-MS/MS
16	ACN 10mL short	QUECHERS	Fipronil-de sulfinyl and Fipronil-sulfid	LC-MS/MS
17	Acetonitrile, 10 mL, 8 minutes	Freezen out + EMR sorbent	Fipronil sulfide and Fipronil desulfynil	other
18	Add 10 mL water, then 10 mL Acetonitrile and quechers extraction , ambient temperature, ten minutes	dSPE with PSA and MgSO4	Fipronil Sulfone	LC-MS/MS
19	acetonitrile (acidified 0,1% with acetic acid); 10 mL; 20±4ºC; 10 min.	None		other
20	ACN,10ml,vortex 1 min, centrifugalize		Fipronil-sulfid, Fipronil-carboxamid, Fipronil-desulfinyl	LC-MS/MS
21	GC-MS:Acidified Acetonitrile, 10mL, 30 secs shake, 3 min spin, 5min evaporate at 70. LC-MS: Acidified acetonitrile, 1min shake, 3min spin, 10min evaporation at ambient temp.	Solid Phase	None	GC-MS, LC-MS/MS
22	Methanol, Room temperature,	modifiied L 00.00 113	Fipronil-desulfinyl, Fipronil-sulfon (MB46136)	LC-MS/MS
23	ACN, 10 mL, RT, 10 min	DSPE with PSA	none	GC-MS/MS, LC-MS/MS
24	10 ml acetonitrile, nanual shaking 1 min	QuEChERS, EN method	None	LC-MS/MS
25	Acetonitrile (20 ml), water (10 ml)	Quechers	Fipronil-Sulfone	LC-MS/MS
26	10 ml Acetonitrile. Extraction time = 10 min	DSPE C18	Fipronil Sulfone + sulfide + desulfinyl + carboxamide	other
27	acetonitrile, QuEChERS	PSA	Fipronil-desulfinyl, Fipronil-sulfid	GC-MS/MS, LC-MS/MS
28	acetonitril, 10mL	dispersive SPE	Fipronil sulfide, Fipronil desulfinyl	GC-MS/MS, LC-MS/MS
29	10 mL acetonitrile, r.t., 1h	dispersive SPE (Quechers)	fipronil-Sulfone, Fipronil-desulfinyl	LC-MS/MS
30	Hexane:Acetone, 15+10 ml, Room Temp, 15 min+15 min	dispersive SPE	-	LC-MS/MS
31	Acetonitrile 1% acetic acid	PSA	Desulfynil, Sulfide.	LC-MS/MS
32	acetonitrile, 10 mL, ambiant, 10 min shake	PSA dspe	f desulfinyl and f sulfide	LC-MS/MS
33	ACN, 10 ml, room temperature, 1 min.	Freeze out	Fipronil Sulfone	GC-MS
34	Acetonitrile, 10ml, room temperature, 15 min	quechers extraction (NaCl/MgSO4) follow by dispersive SPE (150 MgSO4 and 25 mg PSA)	Fipronil Sulfone and disulfinyl	LC-MS/MS
35	10 ml water and 10 ml acetonitril, shake 5 min	PSA C18	Sulfone and sulfide	LC-MS/MS
36	Acetonitrile, 10 mL, ambient, 2 minutes	dSPE EMR-lipid and C18/PSA/MgS04		LC-MS/MS
37	10 ml acetonitrile, 10 minutes (shaker)	dispersive SPE with 25 mg PSA/ml, freezing fat out	Fipronil-sulfide	LC-MS/MS
38	Acetonitrile (10 ml), ambient temperature, 3 min	dSPE with PSA	Fipronil Sulfone, Fipronil desulfinil	LC-MS/MS
39	Ethylacetate 10 ml	PSA and C18	Fipronil Sulfone, Fipronil-Desulfinyl	GC-MS/MS
40	acetonitrile, 10ml, 10 min	QuEChERS and d-SPE	only Fipronil Sulfone	LC-MS/MS
41	10 mL ACN and extraction during 20 min	PSA, C18, Magnesium sulfate at -15°C	Fipronil desulfinil, Fipronil sulfide	LC-MS/MS
42	10ml water/ 10 mlacetonitrile	PSA and magnesium sulfate	Fipronil Sulfone	LC-MS/MS

	12	13	14	15
43	10ml ACN; room T; 10min	PSA	None	GC-MS/MS
44	Ethyl Acetate	PSA/C18		LC-MS/MS
45	Quechers extraction: 10 mL water + 10 mL Acetonitrile	Quechers -dSPE KIT 900mg MgSO4, 150mg PSA, 150mgC18E	Fipronil desulfinyl	LC-MS/MS
46	ACN, 10 ml, ambient, 30 min	PSA	Fipronil-sulfon	LC-MS/MS
47	acetoniltrile	none	Fipronil de-sulfinyl	LC-MS/MS
48	10.0 ml ACN, Room temperature, 10.0 min.	UNI:EN:15662:2009	Fipronil Sulfone	GC-MS/MS
49	10 ml Water, 10 min shaker, 10 ml Acetonitrile, 10 min shaker, room temp.	150 mg PSA and 900 mg magnesium sulphate for an aliquot of 6 ml	Fipronil Sulfone and Fipronil-desulfinyl	GC-MS/MS, LC-MS/MS
50	ACN, 10ml, 30min, 40°C	dSPE	Fipronil-desulfinyl, Fipronil-sulfide	GC-MS/MS, LC-MS/MS
51	Acetonitrile, 10 mL, 15 min	PSA	Fipronil-desulfinyl, Fipronil-sulfid	LC-MS/MS
52	EURL method: "Analysis of Fipronil and metabolites with modified QuEChERS method in Egg" dated 08.08.2017	Dispersive SPE with PSA	Fipronil-Sulfone	LC-MS/MS
53	Acetonitrile 10 mL	dSPE by PSA	Fipronil Sulfone	GC-MS/MS
54	5 ml H20 + 10 ml AcN, room temperature, 15 min	dSPE	None	GC-MS/MS, LC-MS/MS
55	We use the SweEt method. In short 5 g sample shaken with 5g NaSO, 200 mg C18 and 10 mL Ethyl Acetate for 3 minutes.	dSPE is used as a small amount of C18 is used in the extraction.	None in eggs.	GC-MS/MS
56	10 ml Acetonitrile	PSA		LC-MS/MS
57	water, acetonitril	quetchers, DSPE	no	GC-MS/MS
59	acetonitrile, 10ml, RT, 15min	QuChERS with deep freezing (-70°C, 15min) and C18 material	none	LC-MS/MS
60	ACN, extraction salts			LC-MS/MS
61	5 millilitres acetonitril	liquid-liquid-extraction with hexan	-sulfon, -sulfid	LC-MS/MS
62				other
63	Acetonitrile, 10mL, Room Temperature, 15 minutes	Dispersive SPE 15mL Fatty Samples, EN	Fipronil Sulfide, Fipronil Desulfinyl	other
64	ACN, 10ml, QuEChERS	PSA+C18+MgSO4	Sulfone	GC-MS/MS
65	Acetontrile, 10 mL, 1 minute	No clean up, just extraction	Just Fipronil Sulfone	GC-MS/MS
66	n-hexan/acetone = 2/1, cold column extraction, appr. 400ml, room temperature, appr. 3 - 4 hours	GPC	-	GC-MS/MS
67	20 ml Acetonitril / Water (1:1); 1 min + 30 min	5 ml n-Hexane and then dSPE with Zsep+	Fipronil, Fipronil-Sulfone, Fipronil-desulfinyl	LC-MS/MS
68			Only Fipronil Sulfone	GC-MS/MS
69	Quecher methode - 10 mL Acetonitrile	clean up with quecher-kit	Fipronil and Fipronil-Sulfone	GC-MS/MS
70	acetonitrille	dessication with MGSO4	Fipronil-sulfon, Fipronil-desulfynil (for babyfood)	LC-MS/MS
71	Quechers extraction (Acetonitril, 10ml, room temperature)	SPE (Quechers)		LC-MS/MS
72	10 ml acetonitril, ambiant, 5 min	quechers	Fipronil sulfon	LC-MS/MS
73	Extraction of the samples takes 2 hours; 10 mL acetonitril + 10 mL water per sample at both room- and freezing-temperature	quechers-clean up	Fipronil-sulfide and Fipronil-desulfinyl	GC-MS/MS, LC-MS/MS
74	acetonitril	none	Fipronil Sulfone	LC-MS/MS
75	acetonitrile and water 10 ml each	quechers	Fipronil desuflinil	LC-MS/MS
76	Acetonitrile, 10 mL, 20 min	no clean up	Fipronil-desulfinyl	LC-MS/MS
77	5 ml acetone, 5 ml petroleum ether, 2.5 ml dichloromethane, extraction salt added, mechanically shaken	Freezing step and florisil clean up	None in this method	GC-MS/MS
79	10 ml water and 10 ml acetonitrile and shake for 10 min	freesing at night and then dSPE with PSA+MgSO4 clean up mixture	Fipronil desulfonyl but only for baby foods and infant products	GC-MS/MS, LC-MS/MS
80	Water+ACN 10 mL + 10 mL T= amb 20'	dSPE PSA+C18	none	GC-MS/MS
81	Dichlormethane/acetone (2/1)	GPC	Fipronil- desulfinyl	other
82	Acetonitrile, 10 mL	QuEChERS		LC-MS/MS
83	acetonitrile	Quechers (citrate)	Fipronil-Sulfone	GC-MS/MS, LC-MS/MS
84	Acetonitrile, 10 ml, ambient temperature, 2 min	dSPE (Magnesium Sulfate, C18, PSA)		LC-MS/MS
85	Acetonitrile (10 ml), 8 min., water addition (10 ml), t=24C	freezing-out (1 night), centrifugation, DSPE (PSA-MgSO4)	Fipronil-Desulfinyl	GC-MS, LC-MS/MS
86	10 ml of acetonitrile, room temperature, time 2 min.	SPE C-18 sorbent and frezzing (-20C) 30 min.	only Fipronil Sulfone	LC-MS/MS
101	Quechers extraction: 10 mL water + 10 mL Acetonitrile	Quechers -dSPE KIT 900mg MgSO4, 150mg PSA, 150mgC18E	Fipronil desulfinyl	GC-MS/MS
102	10 ml water and 10 ml acetonitrile and shake for 10 min	freesing at night and then dSPE with PSA+MgSO4 clean up mixture	fipronin desulfonyl but only for baby foods	GC-MS/MS, LC-MS/MS

Lab Code	16. Did you use IS? Please specify.	17. What were your calibrants - neat material or pre-prepared commercial standard solutions? Please specify the supplier.	18. Did you perform matrix match calibration?	19. Are your results corrected for recovery?
	16	17	18	19
01	Triphenylphosphate	Neat material	Yes	No
02	NO	NEAT FROM BASF AND Dr ERHENSTORFER	YES	No
03	TFP	neat material -solutions prepared in laboratory	Yes	No
04	TPP (ESI+), Nicarbazin (ESI-)	neat material	Sigma Aldrich	No
05	no	neat material	yes	No
06	no	neat material	yes	No
07	no	dissolved solids; LGC	no	No
08	no	neat material, Sigma-Aldrich Chemie GmbH	no	Yes
09	Triphenylphosphate	Neat, Ehrenstorfer	Yes	No
10		Neat material: Dr.Ehrenstorfer	Yes	Yes
11	no	Fipronil (Sigma 46451), Fipronilsulfon (Sigma 32333)	no	No
12	Isoproturon D6	Restek	Yes	No
13	Fipronil 13C2 15N2	Standards (powder) from Sigma	yes	No
14		Reference material, solution of 57 components, CPA chem, Stara Zagora, Bulgaria	Yes	Yes
15				No
16	Yes but not used in the calculatioon	neat	yes	No
17	Only procedural internal standard, not for correction.	Neat material, Dr Ehrenstorfer	Yes, additionally we used standard addition.	No
18	no	Fipronil and Fipronil Sulfone from Dr Ehrenstorfer	yes	No
19	No	neat material - Dr. Ehrenstorfer	Yes	No
20	Fipronil 13C4	Neochema	no	No
21	Yes- BNB	Neat Material	Yes	No
22	no	commercial standard	yes	No
23		pure substance; solution in ACN prepared in the lab; supplier: Dr. Ehrensdorfer - Germany	no, but we did standard addition to extract portions	No
24	No	Neat material, Fipronil - Dr Ehrenstorfer; Fipronil Sulfone - ChemService	Yes	No
25	Yes, Fipronil-Sulfone isotope	Sigma-Aldrich: Fipronil (46451-100mg) ; Fipronil-Sulfone (32333-50mg)	Yes	No
26	No		Yes	No
27	Dichlorprop-D6, Fipronil-sulfon labelled	neat material	yes	No
28	Yes, Nicarbazin	neat material from LGC	Yes	No
29	external, Nicarbazin; 2,4-D3	0.0002; 0.0005; 0.001; 0.005; 0.01; 0.02; 0.05 ng/uL	yes	No
30	no	Ehrenstorfer standards	procedural calibration using blank egg matrix	Yes
31	No	Neat stds. Dr Ehrenstofer	No	No
32	yes, Fipronil 13 C2 15N2 Fipronil	Dr. Ehrenstorfer	NO	No
33	TPP	Dr.Ehrenstorfer	Yes	No
34	no	neat material	yes	Yes
35	no	commercial standard solutions	yes	No
36	For LC/MSMS: no IS but recovery standards: TDCPP and d5-terbutylazine	Pre-prepared commercial standards (Dr. Ehrenstorfer)	Yes	No
37	Dichlorprop D6	neat Material by Dr. Ehrentorfer	yes	No
38	Nicarbazine for volume correction	neat material, Fipronil - Sigma Aldrich, Fipronil sulfon - Dr. Ehrenstorfer	yes	No
39	Parathion-d10	Ehrenstorfer	Yes	No
40	no	neat material from Sigma-Aldrich	yes	No
41	Bentazone D6	SIGMA ALDRICH	Yes	No
42	Fipronil C13	powder by Sigma-aldrich	No	No
43	PCB 31	Sigma 46451	Yes	No
44	No	Commercial	no	No
45	TPP	Neat material	YES	No
46	Nicarbazin, but without recovery correction	neat material, Dr. Ehrenstorfer	yes	No
47	na	reference materials matrix matched egg	yes	No

	16	17	18	19
48	Yes, PCB 209 at 100.0 µg/l	LabStandard-Lab Instruments and Dr. Ehrenstorfer	Yes, matrix match calibration and procedural calibration	Yes
49	TPP for GC-MS/MS	Dr. Ehrenstorfer GmbH	Yes	No
50	TPP	neat material from h-pc	Yes	Yes
51	no	own prepared standard solution	yes	No
52	No	Neat material, Dr. Ehrenstorfer - Fipronil, Sigma Aldrich - Fipronil-Sulfone	Yes	No
53	TriPhenylPhosphate	neat material by ChemService	yes	No
54	No	Fipronil - CPA standard solution; Fipronil Sulfone - DrEhr neat standard	Yes for LC-MS-MS	No
55	No	Commercial standard solution.	Yes, procedural standards were used.	Yes
56	No	Neat material, Ehrenstorfer	No	No
57	Fipronil C13, Fipronil sulfon C13	standard solution	no	No
59	yes; Desmetryn mainly for control method application	neat material; Dr. Ehrenstorfer/LGC	yes for the first test; standard addition for confirmation	No
60	Nicarbazin		Yes	No
61	no	substance, supplier sigma-aldrich	no: we use matrix calibration	No
62				No
63	Yes. 2,4,6-(trimethyl phenoxy)-acetic acid (TMA)	Neat material. Dr.Ehrenstorfer and Sigma Aldrich	Yes.	No
64	TPP	Dr. Erhenstorfer	Yes	No
65	PCB 138	Pre-prepared commercial standard solution from Restek	Yes	Yes
66	no	neat material, Fipronil: Dr. Ehrenstorfer, Fipronil sulfon: Sigma-Aldrich	yes	No
67	No	Fipronil from Ehrenstorfer, all others from HPC	yes, however using procedural calibration	Yes
68	Yes		Yes	No
69	Fipronil C13	Calibration standards of Dr. Ehrenstorfer - controle with standards of Sigma-Aldrich	Matrixaddiition at one sample of a batch	No
70	yes, before extraction	derived from commercial standards	no	No
71	no	Standard solutions, Dr. Ehrenstorfer	no	No
72	Yes,		Yes	No
73	Yes, Atrazine-D5 and Terbutryn-D5	De calibrants from Sigma-Aldrich are further diluted at our laboratorium.	yes	No
74	yes	neat material from Sigma	yes	Yes
75	yes	sigma	yes	Yes
76	yes: 2,4-D 13C6	commercial standard from HPC (Fipronil), LGC (Fipronil-sulfon) and Sigma-Aldrich (Fipronil-desulfinyl)	yes	No
77	PCB-153	Calibration line made from Dr Ehrenstorfer pure compounds	No, apple matrix in calibration standards	Yes
79	triphenyl phosphate	neat material diluted by techniciants - Dr. Ehrenstorfer	yes	No
80	Triphenylphosphate	Ultra scientific	Yes	No
81	trans-Nonachlor	neat material	yes	No
82	Yes, Acetamiprid-D3		Procedural calibration	Yes
83	TCPP	Pre-prepared commercial standard solution supplied by A2S Analytical Standards	Yes, we did	No
84	Nicarbazin	neat materials - dr. Ehrenstorfer	yes	No
85	No	neat material; Dr. Ehrenstorfer	Yes	No
86	yes, TPP	neat material, supplier Dr. Ehrenstorfer	Yes	No
101	TPP	Neat material	yes	No
102	triphenyl phosphate	neat material diluted by technicians - Dr. Ehrenstorfer	yes	No

Lab Code	20. Please explain how and give any recovery correction factor if applied.	21. Did you experience difficulties during the analyses or reporting?	22. What is your current tool for ensuring the traceability of your results (e.g. reference material, in-house calibrator, reference method)?	23. Do you use CRM for quality control (if available)?
	20	21	22	23
01	No recovery correction is applied	No	In-house calibrator	No
02	NO FACTOR WAS APPLIED	NO		NO
03	75%	No	in house calibrators	No
04	no recovery correction if recavery 70-120 % in general	no	determination of recovery with every sequence	yes
05		no	spiking of analytes (QC); (each working day)	In the moment it is not available.
06	not applied	no	reference method	not available
07	50%	no	Spiking of Blancs; standard-addition	no
08	standard addition	no	reference material, in-house calibrator	yes
09	recovery tested, around 100%, therefore no correction for recovery	No	In-house reference sample	In-house reference sample
10	Recovery figures obtained using 3fold recoveries measured in the same batch. Spiking levels were close to the submited result concentrations. Fipronil correction factor = 1.416. Fipronil Sulfone correction factor = 1.511	No		No
11	the recoveries are on the average of 100%	no	in-house PT	no
12	No recovery correction factor is applied.	None	Our quality control is a "blank" with an addition of an amount of Fipronil and Fipronil Sulfone (corresponding to the LOQ).	No
13		no	none	no
14			comparison with technique GC ECD	No
15	standardaddition			
16	no recovery correction		ref. material EURL AO12 egg	no
17	No corrected, recoveries between 90-100%	No	in-house calibrator, in house validated method	No
18	we don't apply the recovery correction	no	in house spiked samples	no
19	No recovery correction since recoveries acceptable: Fipronil (105.3%); Fipronil Sulfone (99,9%)	No	European Union Proficiency Tests	No
20	corrected with IS	no	in-house reference material	no
21		Had to half the weight of sample to obtain sufficient supernatant during extraction.	Recoveries, accredited method	No
22	Recovery for Fipronil: 83,4 % , Fipronil-sulfon: 84,9 %			no
23	No recovery correction was done	No		No
24	Not corrected		In-house calibrator	Yes
25		No		No
26	No correction			
27	No	no	reference material	no
28	.	MRL is indicated in mg/kg, the reporting is in µg/kg	spiked sample	No
29		no	spiked blank material (quality control sample)	no
30	Procedural calibration was used (spiking to blank sample portions before extraction)	yes, extraction efficiency varieties from validation, was not time to repeat analyses	spiking to blank sample portions	no
31	NA	Yes, difficulties separating the layers in initial extraction. less supernatent achieved than normal.	Certified standards.	No
32	we used isotopic dilution so no correction applied	yes for reporting about coverage factor (no definition) so we applied 2, for final (Fipronil + Fipronil Sulfone) results we had applied residus definition = Fipronil *1 + Fipronil Sulfone*0.96	reference material given by EURL AO	no
33	Recovery 80-120%	No	Reference material, spiked samples	No
34	procedural calibration	no	in-house calibrator, EURL method	no
35		no	in-house	no
36	Recoveries are within 70%-120% (SANTE), no correction applied	No	ISO 17025	No
37	we did not correct for recovery because our recovery was 108 %			
38		No	reference material (neat certified standard)	no matrix CRM
39	Not corrected for recovery	No	Reference material	yes
40	we do not correct for recovery	no	in-house calibrator	no

	20	21	22	23
41	No recovery correction factor for the results	No	With MRI	
42	recovery correction is applied when the recovery on a spiked blank matrix was under 70% or above 120%	No	No	No
43	Not corrected	No	Reference standard solution	No
44	i have not apply correction factor	no	reference method	no
45	not applied	no	Reference material, validation, reference method	yes
46	3 LL (2,5 ng/ml): 104 %, 102 %, 106 %; 3 HL (10 ng/ml): 105 %; 107 %; 102 %	no	recovery samples	no
47	NA	no		not available
48	The results were corrected using the correction factor derived from the average recovery obtained in method validation studies	No.	Reference material and Reference method	Yes
49	We didn't apply correction factor	No	Reference method	No
50	matrix matched calibration			
51		no	standard addition	no
52	Recoveries in the range of 100-110%, no correction applied.	No	Proficiency tests.	No
53	no used any correction for recovey	no	spiked samples in house for recoveries checks	no
54	No	No	reference material	
55	The standards were procedural standards and thus the results are recovery corrected	No	QCs and PTs	No
56	No correction factor applied	The calculation of the uncertainties of the results could have been better clarified with examples.	Control samples for estimating recovery	No
57	-	no	in house	no
59	no recovery correction factor applied	no (no labels on the sample pots, only on the bags)	procedure control standard (recovery test)	no
60	no correction			no
61	no recoverycorrection in case of matrix calibration	no	inhouse-validation and sample measurement by differnt laborities (intern PT)	seldom1-
62				
63	No recovery correction applied.	No.		No.
64		No	in-house calibrator	No
65	92	No	Reference material	Yes
66	results are NOT corrected for recovery	no	in-house calibrator	no
67	procedural calibration	No	quality control samples, test material from EUPT-AO12 (egg)	no
68		No	in-house calibrator	No
69		no	in-house calibrator	no
70		no	3rd line controls	no
71	standard addition (recovery between 90-100%)	no		
72		No		No
73	Not applicable, no correction factor was used.	no	several tools, such as storage of digital results, storage of sample, storage of raw data	no
74	use of internal standard implies correction for eventual losses	no	in-house QC-samples and reference material obtained from NRL	no
75	sample is spiked prior to extraction	no		no
76	118 % Fipronil and 119 % Fipronil-sulfon	no	-	CRM not yet available but planed (material from BVL in Berlin)
77	Corrected for standard additions to the sample	No	Standard additions and control samples	No
79		no		no
80	104% Fipronil; 108% Fipronil Sulfone	no	spiked sample	no
81		no	in-house calibrator	yes
82	automatically via procedural calibration			
83	The results are not corrected for recovery	No, we didn't	In house calibrator and reference material	No, we don't
84	Results are NOT corrected for recovery. (Recovery: Fipronil - 102%, Fipronil sulfon-104%)	no	spiked samples, different determination techniques (GC-MS/MS, GC-ECD)	yes
85	not applied	no	reference material	no
86	1	No	in-house calibrator	No
101	not applied	NO	Reference material, validation, reference method	yes
102		no	reference material	no

Lab Code	24. Are you interested in availability of matrix CRM for QC in determination of Fipronil residue in food?	25. Which matrices, please indicate such as eggs (liquid or powder) or food products containing egg (please specify).	26. What concentration levels of Fipronil and Fipronil Sulfone are you interested in?	27. How frequently would you use such a CRM in your laboratory?
	24	25	26	27
01	No	None	0.004 mg/kg	Never
02	NO			
03	Yes	Liquid eggs	1-2 x MRL	Monthly
04	yes (if material contains other pesticides additionally)	pasta, bakery products	between 5µg/kg and 50µg/kg	quaterly
05	yes	egg, meat	2 and 5 µg/kg	each working day
06	no	no	-	-
07	yes	egg powder; Egg liqueur	3 µg/kg	one per batch
08	yes	egg powder	0.005 µg/kg	always when egg samples are analysed
09	No			
10				
11	yes	eggs, noodles, bisquite, advocaat	0,5 µg/kg - 25 µg/kg	at every Fipronil-project, maybe 4times a year
12	yes	eggs and transform product containing eggs such as cakes	5 µg/kg and 10 µg/kg	Never
13	yes	meat	around MRL (5 µg/kg)	for pesticide : never
14	Yes	eggs liquid, powder,baby food containing egg, egg white,egg yolk	0,002 mg/kg, 0,005 mg/kg	as needed
15				
16	yes	liquid egg	5-20 ug/kg	1-3 times p. year
17	Yes	Fruits and vegetables	0.002 mg/kg	None
18	yes	liquid eggs, pasta, chicken meat	near the MRL	every analytical batch
19	Yes	liquid	0.005 to 0.015	Never
20	no			
21	Yes	Both	Low or near the MRL	For method validation, training and possible QC Check. Monthly.
22	yes			
23	No			
24	No need for egg; not in routine method scope			
25	Yes	Liquid eggs; chicken muscle	5 µg/kg	12
26				
27	yes	egg, muscle, fat	low level, below 0.010 mg/kg	once a year
28	Yes	egg powder	0.010 mg/kg	once a quarter
29	no	-	-	-
30	possibly	eggs	low, near MRL level	
31	yes	egg, poulrty meat.	Our target reporting limit is 5 ppb for Fipronil +metabolites.	As required.
32	y	eggs powder, processing food containing egg	MRL levels	for each serie of analysis if available
33	Yes	Eggs (liquid or powder)	10 ppb	2 times per year
34	no			once. to confirm method efficiency at the end of the method validation
35	no			
36	Yes	Eggs: liquid or powder	0.005 mg/kg to 0.250 mg/kg	
37				
38	yes	eggs (powder or liquid)	close to MRM	regularly in each series of sample
39	No			
40	yes	liquid egg/liquid albumen/liquid yolk	near MRL	every batch (depending on the cost)
41				
42	Yes	liquid egg	1/2 MRL	for each series
43	Yes	Powder	0,008mg/kg	Internal control
44	yes	liquid egg	MRL	Confirmatory
45	yes	Food products containig eggs	5-10 ug/kg	once or Twice a years
46	no			
47	yes	eggs	0.005 - 0.02 mg/kg	once a month
48	No in this moment	Eggs, Pasta, Cookies, Mayonnaise, Feed, Meat, Fat Tissues.	1.0-2.5-5.0-10.0-25.0 µg/kg in matrix and 1.0-2.5-5.0-10.0-25.0 µg/l in solvent	All time
49	Yes	liquid	5 ug/kg	once per month
50				
51	yes	mayonnaise	0.005 mg/kg	once a week
52	No			
53	no			
54	yes	powdered egg products	5 ug/kg	For each non-compliant sample
55	No			

	24	25	26	27
56	No			
57	yes	eggs, food containing eggs, water, manure	around MRL	monthly
59	yes	eggs (liquid)	0.003 mg/kg	
60	no			
61	yes	egg-powder, muscle (lyophil.)	0,003 until 0,020 mg/kg	1-2 per year
62				
63	Yes.	Eggs (liquid).	About MRL.	Once a week.
64	Yes	Fat	> 0.005- < 0.020 mg/kg	Twice a month
65	Yes	Eggs in powder form	Around 5 ppb	For every positive detection
66	yes	eggs	concentration levels corresponding to sample A or higher	for each series of analysis
67	yes	whole eggs, liquid, powder, all kinds of eggs	0,005 mg/kg	if availible quite often
68	Yes	Eggs liquid and powder, pasteurized eggs	from 0.002 to 1.0	One/month
69	yes			
70	possibly			
71				
72	No			
73	no	not applicable	not applicable	not applicable
74	yes, always	whole egg	round MRL (5 µg/kg)	Quatraly by frequent analysis
75	yes			
76	yes	eggs	0,005-0,02 mg/kg	3-6 times a year
77	Yes	All	5-100 ug/kg	Monthly
79	no			usually not
80	Yes	eggs	0.005 mg/Kg	each batch
81	yes	eggs (liquid and powder),baby spikes, mayonnaise	10-20 ug/kg	monthly
82	Yes	egg		
83	Yes, we are	Eggs, fresh pasta	0.005 mg/kg	It dipends on the number of analytical sessions in the future
84	yes	eggs - liquid and powder	about or above 0,005 mg/kg	in confirmatory analysis (in cases of MRL exceedances)
85	yes	liquid egg	2.5 ug/kg	once a year
86	Yes	liquid eggs and powder	from 0.005 to 0.5	not very often
101	yes	Food products containig eggs	5-10 ug/kg	once OR Twice a years
102	no			usually not

Erratum

The laboratory codes in Annex 6 are corrected. The Lab Codes that were changed have an orange background (see Pages 29, 30, 32-34)

ANNEX 7: Homogeneity of the liquid egg material

SAMPLE A - Fipronil

Standard Statistics :Fipronil - Fipronil

sample	Replicate 1		Replicate 2		Replicate 3		Sample Mean	Sample StDev
	anal.#	result	anal.#	result	anal.#	result		
8	1	9.25	20	9.27	21	9.73	9.417	0.272
30	2	9.48	19	9.01	23	9.32	9.270	0.239
48	3	9.44	18	9.17	25	8.99	9.200	0.226
69	4	8.83	17	9.01	27	9.21	9.017	0.190
84	5	9.39	16	9.47	29	9.06	9.307	0.217
98	6	9.16	15	9.09	30	9.07	9.107	0.047
117	7	9.48	14	9.34	28	9.62	9.480	0.140
129	8	9.49	13	9.42	26	9.32	9.410	0.085
159	9	9.29	12	9.57	24	9.15	9.337	0.214
189	10	9.74	11	9.43	22	9.41	9.527	0.185

Source of Variation	SS	d.f.	MS	StDev	F	F-crit 95%	F-crit 99%
Between Units	0.717	9	0.080	0.119	2.126	2.393	3.457
Within Units	0.749	20	0.037	0.194			
Total	1.466	29					
Snedecor F-Test RESULTS	Differences between units statistically significant? (a=95%) :No						
	Differences between units statistically significant? (a=99%) :No						
Homogeneity Results	Weight.Avg.	Sbb	Sbb (%)	Swb	Swb (%)	Ubb*	Ubb* (%)
	9.307	0.119	1.3%	0.194	2.1%	0.063	0.7%

SAMPLE A - Fipronil Sulfone

Standard Statistics :Fipronil - Fipronilsulfone without IS

sample	Replicate 1		Replicate 2		Replicate 3		Sample Mean	Sample StDev
	anal.#	result	anal.#	result	anal.#	result		
8	1	6.054064759	20	6.040045208	21	5.713552282	5.936	0.193
30	2	6.222591086	19	5.728601674	23	5.888782049	5.947	0.252
48	3	6.40113798	18	5.890271635	25	6.559923527	6.284	0.350
69	4	5.633893882	17	5.732491679	27	6.183444137	5.850	0.293
84	5	6.238095884	16	5.584571146	29	6.486749829	6.103	0.466
98	6	6.138846258	15	5.953447267	30	5.679766145	5.924	0.231
117	7	5.948348727	14	5.193931486	28	5.879393315	5.674	0.417
129	8	5.658910469	13	5.824597444	26	5.756803765	5.747	0.083
159	9	5.870860623	12	5.898902539	24	5.476075701	5.749	0.236
189	10	5.500072084	11	5.924638122	22	5.958214698	5.794	0.255

Source of Variation	SS	d.f.	MS	StDev	F	F-crit 95%	F-crit 99%
Between Units	0.911	9	0.101	0.066	1.149	2.393	3.457
Within Units	1.763	20	0.088	0.297			
Total	2.674	29					
Snedecor F-Test RESULTS	Differences between units statistically significant? (a=95%) :No						
	Differences between units statistically significant? (a=99%) :No						
Homogeneity Results	Weight.Avg.	Sbb	Sbb (%)	Swb	Swb (%)	Ubb*	Ubb* (%)
	5.901	0.066	1.1%	0.297	5.0%	0.096	1.6%

SAMPLE B - Fipronil Sulfone

Standard Statistics :Fipronil - Sample B fipronil sulfone without IS

sample	Replicate 1		Replicate 2		Replicate 3		Sample Mean	Sample StDev
	anal.#	result	anal.#	result	anal.#	result		
6	1	1.139623844	20	1.124057436	21	1.094267841	1.119	0.023
48	2	1.120411719	19	1.092901793	23	1.055270522	1.090	0.033
55	3	1.060758983	18	1.107172727	25	1.021899178	1.063	0.043
78	4	1.131919526	17	1.061539043	27	1.053440984	1.082	0.043
108	5	1.068548805	16	1.055758424	29	1.091960583	1.072	0.018
123	6	1.052996125	15	1.016030915	30	1.051787749	1.040	0.021
144	7	1.102306921	14	1.056034895	28	1.029091446	1.062	0.037
159	8	1.066247678	13	1.036498774	26	1.121773081	1.075	0.043
192	9	1.084796948	12	1.087200179	24	1.038155714	1.070	0.028
215	10	1.051650948	11	1.052902409	22	1.051466519	1.052	0.001

Source of Variation	SS	d.f.	MS	StDev	F	F-crit 95%	F-crit 99%
Between Units	0.013	9	0.001	0.012	1.401	2.393	3.457
Within Units	0.020	20	0.001	0.032			
Total	0.033	29					
Snedecor F-Test RESULTS	Differences between units statistically significant? (a=95%) :No						
	Differences between units statistically significant? (a=99%) :No						
Homogeneity Results	Weight.Avg.	Sbb	Sbb (%)	Swb	Swb (%)	Ubb*	Ubb* (%)
	1.073	0.012	1.1%	0.032	3.0%	0.010	1.0%

ANNEX 8: Reported results and Z scores, for Fipronil, Fipronil Sulfone & SUM in SAMPLE A & SAMPLE B

(#) Twenty Laboratories may have reported uncertainties in % (instead of µg/kg)

Lab code	SAMPLE A									SAMPLE B								
	Fipronil (F)			Fipronil Sulfone (FS)			F+FS			Fipronil (F)			Fipronil Sulfone (FS)			F+FS		
	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)
Unit	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg
1	8.97	-0.5	2.33	5.07	-1.0	0.91	13.8	-0.7	3.03	< 4.00			< 4.00			< 4.00		
2	0.01	-4.0	0.01	0.01	-4.0	0	0.02	-4.0	0.01	< 0.00			< 0.00			< 0.00		
3	9	-0.5	3	7	0.2	3	16	-0.2	0	< 2.00			< 5.00			< 5.00		
4 #	8	-0.9	50	6	-0.4	50	14	-0.7	50	< 1.00			< 2.50			< 2.00		
5	10.4	0.0	4.2	6.3	-0.3	2.5	16.5	-0.1	6.6	< 2.00			< 2.00			< 2.00		
6	9.2	-0.5	4.6	8.4	1.0	4.2	17.3	0.1	8.6	< 5.00			< 5.00			< 5.00		
7	9.6	-0.3	4.8	7.4	0.4	3.7	16.7	0.0	8.4	< 2.00			< 2.00			< 2.00		
8	9.59	-0.3	0.72	6.37	-0.2	0.93	15.74	-0.3	1.17	< 0.01			< 0.01			< 0.01		
9	9.2	-0.5	4.6	5.8	-0.6	2.9	14.8	-0.5	7.4	< 2.50			1		0	1		0
10 #	9.28	-0.4	50	5.41	-0.8	50	14.5	-0.6	50	< 1.00			1.05		50	1.01		50
11 #	8.3	-0.8	50	5.4	-0.8	50	13.5	-0.8	50	< 0.30			0.9		50	0.9		50
12	6.6	-1.5	2.2	12.6	-1.0	6.3	6.2	-0.3	3.1	< 5.00			< 3.00			< 5.00		
13 #	9.3	-0.4	25	7.1	0.2	25	16.1	-0.2	25				1.3		25	1.2		25
14	11	0.2	5.5	7.9	0.7	4	19	0.5	9.3	< 2.00			< 2.00			< 4.00		
15	9.31	-0.4	0.4	6.12	-0.4	0.4	15.22	-0.4	0.4				1.03		0.4	0.99		0.4
16	10.3	0.0		6.5	-0.1		16.6	-0.1		5		0	5		0	5		0
17	11	0.2	6	7	0.2	4	18	0.3	9				1		0	1		0
18	11	0.2	5.5	22	9.1	11	32.23	3.6	16.11	< 2.50			3.6		1.8	3.47		1.74

Lab code	SAMPLE A									SAMPLE B								
	Fipronil (F)			Fipronil Sulfone (FS)			F+FS			Fipronil (F)			Fipronil Sulfone (FS)			F+FS		
	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)
Unit	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg
19	8.8	-0.6	4.4	5.2	-0.9	2.6	13.9	-0.7	7	< 5.00			< 5.00			< 5.00		
20	12.12	0.7		6.3	-0.3		18.42	0.4		< 1.00			1.24		0	1.24		0
21 #	6	-1.7	50	12	3.1	50	18	0.3	50				11		50	11		50
22	8	-0.9		5	-1.0	0	12.82	-1.0	0	< 2.50			< 2.50			< 2.50		
23	11	0.2	5.5	6.7	0.0	3.4	17	0.0	8.5	< 2.00			< 2.00			< 5.00		
24	11.9	0.6	5.95	5.4	-0.8	2.7	17.1	0.1	8.55	< 5.00			< 5.00			< 5.00		
25 #	15.97	2.1	50	8.65	1.1	50	24.62	1.8	50	2.98		50	3.47		50	6.45		50
26	12	0.6		7	0.2		19	0.5		< 5.00			< 3.00			< 5.00		
27	11.4	0.4	5.7	7.39	0.4	3.7	18.6	0.4	9.3	< 1.00			1.21		0.6	1.17		0.59
28 #	10	-0.1	50	6	-0.4	50	16	-0.2	50	< 2.00			< 2.00			< 2.00		
29 #	9.22	-0.4	50	7.66	0.6	50	16.88	0.0	50	< 1.00			1.33		50	1.33		50
30	11.7	0.5	12	8.1	0.8	4	19.5	0.6	0	< 2.00						< 2.00		
31	10.1	-0.1	5.1	6.3	-0.3	3.2	16.2	-0.2	8.1	< 1.00			1.2		0.6	1.2		0.6
32	10.1	-0.1	1.51	5.84	-0.5	0.88	15.7	-0.3	1.75	< 1.00			1.11		0.17	1.06		0.17
33	9.1	-0.5	4.55	6.7	0.0	3.35	15.6	-0.3	7.8	< 5.00			< 5.00			< 5.00		
34 #	11.05	0.3	50	6.55	-0.1	50	17.34	0.1	50	< 2.50			< 2.50			< 2.50		
35	10	-0.1	1	7	0.2	0.7	17	0.0	1.7	< 2.40			< 2.40			< 2.40		
36	8.6	-0.7	4.3	5.2	-0.9	2.6	13.6	-0.8	6.8	< 2.50			< 2.50			< 5.00		
37	10.5	0.0		5.9	-0.5		16.19	-0.2		< 1.00			< 3.00			< 3.00		
38	9.6	-0.3	2.9	5.3	-0.9	1.6	14.7	-0.5	4.4	< 1.00			< 1.00			< 2.00		
39	10.37	0.0	6.22	6.11	-0.4	3.67	16.27	-0.1	9.76	< 5.00			< 5.00			< 5.00		
40	9.1	-0.5	4.6	5.7	-0.6	2.9	14.6	-0.5	7.3	< 2.50			< 2.50			< 5.00		
41 #	13	1.0	30	7	0.2	35	20	0.7	35	< 5.00			< 5.00			< 5.00		

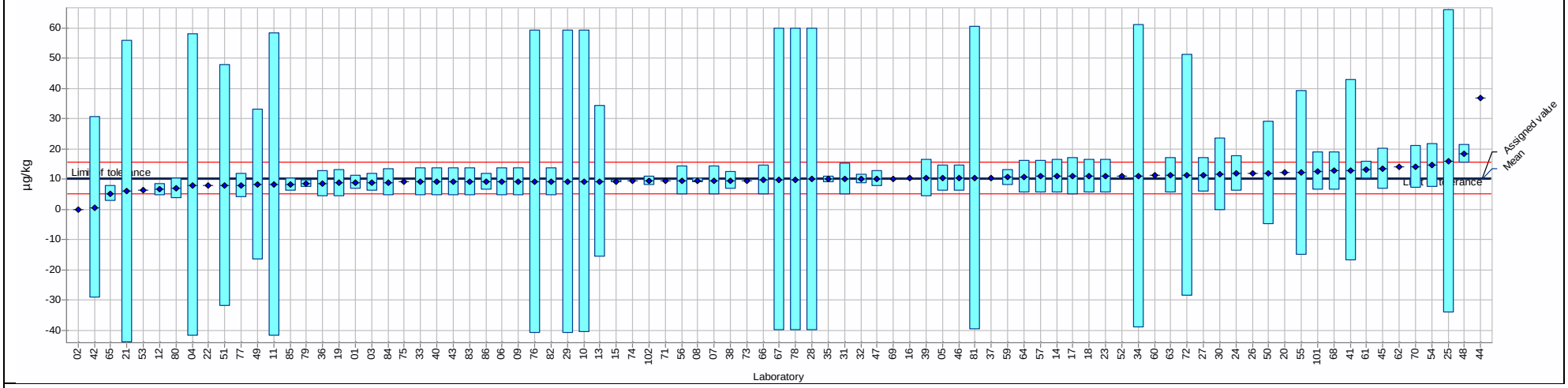
Lab code	SAMPLE A									SAMPLE B								
	Fipronil (F)			Fipronil Sulfone (FS)			F+FS			Fipronil (F)			Fipronil Sulfone (FS)			F+FS		
	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)
Unit	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg
42 #	0.69	-3.7	30	0.6	-3.6	30	1.27	-3.7	30	< 0.20			< 0.20			< 0.40		
43	9.1	-0.5	4.55	7.2	0.3	3.6	16	-0.2	8	< 5.00			< 5.00			< 5.00		
44	36.87	10.2		22.66	9.5		58.73	9.9		< 2.00			< 5.00			< 5.00		
45	13.5	1.2	6.8	8.2	0.9	4.1	21.3	1.0	10.7				1.4		0.7	1.4		0.7
46	10.41	0.0	4.3	6.72	0.0	6.2	16.89	0.0	5	< 5.00			< 5.00			< 10.00		
47	10.2	-0.1	2.5	6.7	0.0	1.7	16.7	0.0	4.2	< 1.00			< 1.00			< 1.00		
48	18.39	3.1	3	9.9	1.9	1.5	28.29	2.7	4.5	0.27		0.06	1.8		0.36	2.07		0.41
49 #	8.2	-0.8	25	5.3	-0.9	22	13.3	-0.8	25	< 3.00			< 3.00			< 3.00		
50 #	12	0.6	17	7.5	0.5	9	19.2	0.5					1.1		9	1		0
51 #	8	-0.9	40	5	-1.0	40	13	-0.9	40	< 2.00			< 2.00			< 4.00		
52	11	0.2		6	-0.4		17	0.0	8.5	< 2.00			< 2.00			< 4.00		
53	6.5	-1.5		7.1	0.2		13	-0.9	0.01	< 2.00			< 2.00			< 2.00		
54	14.6	1.6	7.3	8.4	1.0	4.2	23.3	1.5	11.7	< 1.00			2.9		1.4	3		1.5
55 #	12.2	0.7	27.2	7.5	0.5	27.2	19.4	0.6	27.2	< 2.50			< 2.50			< 5.00		
56	9.58	-0.3	4.79	6.56	-0.1	3.28	15.9	-0.2	7.95	< 5.00			< 5.00			< 5.00		
57	10.9	0.2	5.45	6.5	-0.1	3.25	17.4	0.1	8.7	< 2.50			< 2.50			< 2.50		
59	10.6	0.1	2.65	7.77	0.6	1.94	18.1	0.3	4.5	< 0.50			1.31		0.33	1.26		0.32
60	11.29	0.3		6.81	0.0		17.82	0.2		< 1.00			1.36		0	1.31		0
61	13.1	1.0	2.9	7.8	0.6	1.2	20.6	0.9	3.2	0.33		0.19	1.45		0.28	1.7		0.34
62	14	1.4	0	7.2	0.3	0	21.2	1.0	0				1		0	1		0
63	11.3	0.3	5.7	6.4	-0.2	3.2	17.5	0.1	8.8	< 2.00			< 2.00			< 4.00		
64	10.76	0.1	5.38	9.48	1.6	4.74	19.68	0.7	9.84	< 5.00			< 5.00			< 5.00		
65	5.32	-1.9	2.66	3.37	-2.0	1.68	8.57	-2.0	4.28	< 2.00			< 2.00			< 2.00		
66	9.8	-0.2	4.9	5.6	-0.7	2.8	15.2	-0.4	7.6	< 2.50			< 1.00			< 3.50		

Lab code	SAMPLE A									SAMPLE B								
	Fipronil (F)			Fipronil Sulfone (FS)			F+FS			Fipronil (F)			Fipronil Sulfone (FS)			F+FS		
	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)	Result	Z score	MU (abs.)
Unit	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg	µg/kg		µg/kg
67 #	9.9	-0.2	50	6.3	-0.3	50	15.9	-0.2	50				< 2.00			< 2.00		
68	12.7	0.9	6.3	7.7	0.6	3.8	20.1	0.8					1.5		0	1.4		0
69	10.23	-0.1		6.29	-0.3		16.52	-0.1		< 3.00			< 3.00			< 3.00		
70	14	1.4	7	6	-0.4	3	20	0.7	10	< 2.50			1.2		0.6	1.1		0.55
71	9.5	-0.3		6.4	-0.2		15.7	-0.3		< 1.00			1.1		0	1.1		0
72 #	11.33	0.4	40	8.76	1.2	40	19.99	0.7		< 3.00			< 3.00			< 5.00		
73	9.61	-0.3		8.7	1.2		17.96	0.3		< 3.00			< 3.00			< 3.00		
74	9.4	-0.4	0.06	5.6	-0.7	0.05	14.8	-0.5	0.1	< 0.50			1		0.05	0.95		0.1
75	9.02	-0.5		6.39	-0.2		15.19	-0.4		< 2.00			< 2.00			< 2.00		
76 #	9.2	-0.5	50	5.4	-0.8	50	14.4	-0.6		< 1.00			< 1.00			< 1.00		
77	8	-0.9	4	5	-1.0	2.5	13	-0.9	6.5	< 5.00			< 5.00			< 5.00		
78 #	9.92	-0.2	50	nd			nd											
79	8.53	-0.7	1.28	5.5	-0.7	0.66	13.81	-0.7	1.91	< 1.00			1.43		0.17	1.37		0.16
80	7	-1.3	3.5	6.3	-0.3	3.2	13	-0.9	6.5	< 2.00			< 2.00			< 4.00		
81 #	10.45	0.0	50	5.89	-0.5	50	16.13	-0.2	50	< 2.00			< 3.00			< 5.00		
82	9.2	-0.5	4.6	6	-0.4	3	15	-0.4	7.5	< 2.00			< 2.00			< 2.00		
83	9.1	-0.5	4.6	6.1	-0.4	3.1	15	-0.4	7.5	< 2.50			< 2.50			< 5.00		
84	9	-0.5	4.5	6.95	0.1	3.48	15.7	-0.3	7.85	< 2.50			< 2.50			< 5.00		
85	8.32	-0.8	2.08	5.32	-0.8	1.33	13.42	-0.8	3.36	< 1.00			1.14		0.29	1.09		0.27
86	9.18	-0.5	2.75	4.82	-1.1	1.45	13.83	-0.7	4.15	< 1.50			< 1.00			< 2.50		
101	12.6	0.8	6.3	6.3	-0.3	3.2	18.6	0.4	9.3				1.3		0.6	1.3		0.6
102	9.5	-0.3	1.43	6.45	-0.2	0.77	15.69	-0.3	2.17	< 1.00			1.33		0.16	1.27		0.15

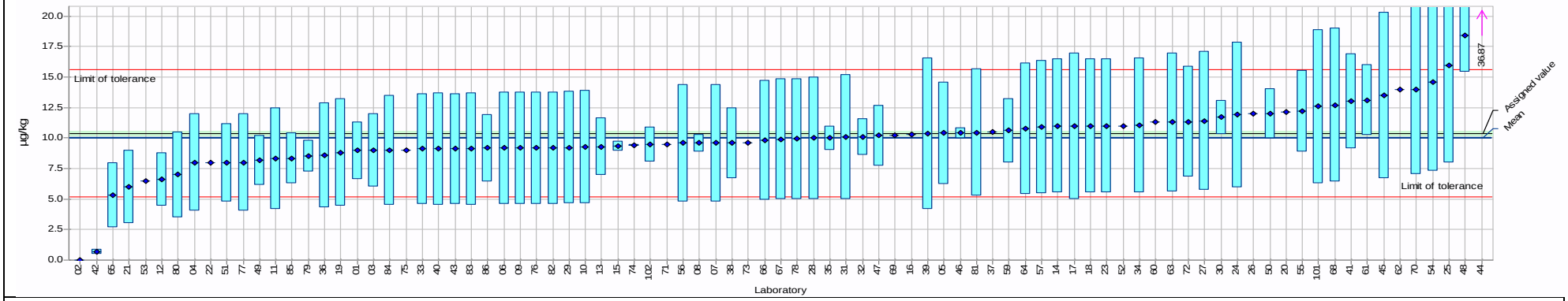
ANNEX 9: Graphs of data reported by participants

Reported results for Fipronil (F) content in SAMPLE A

Sample:	**SAMPLE A**	**Assigned value:**	**10.39 µg/kg (Reference value)**
Measurand	**F**	**Mean value:**	**10.05 µg/kg**
Method:	**ISO 5725-5 (Alg. A+S)**	**Rel. reproducibility s.d.:**	**17.24 %**
Number of laboratories in calculation:	**87**	**Rel. target s.d.:**	**25.02 % (Reference value)**

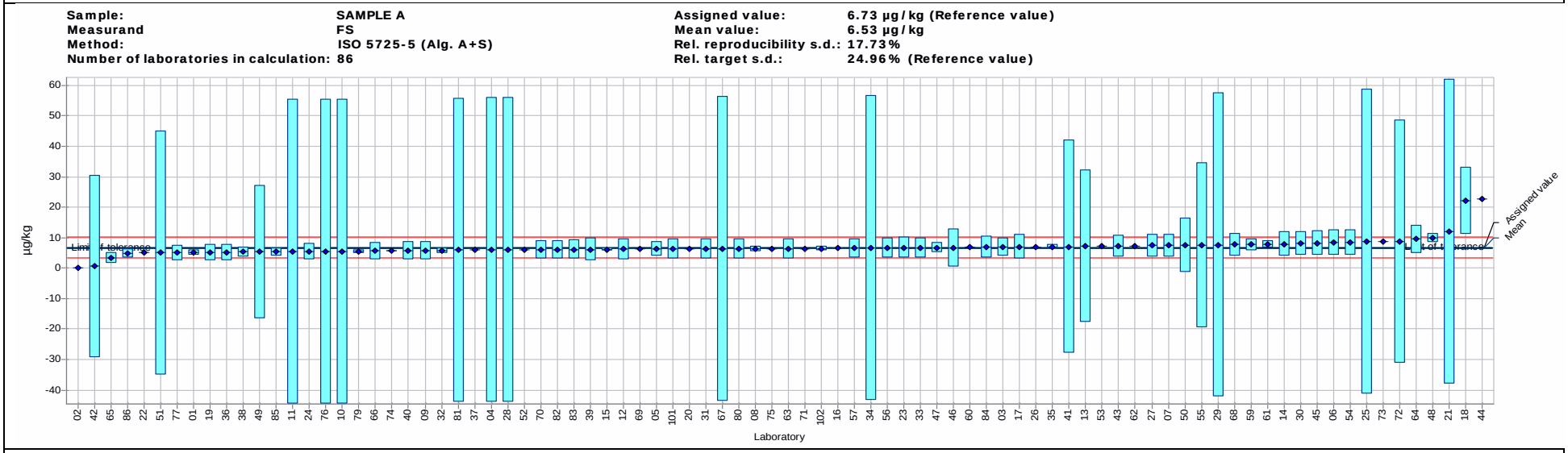


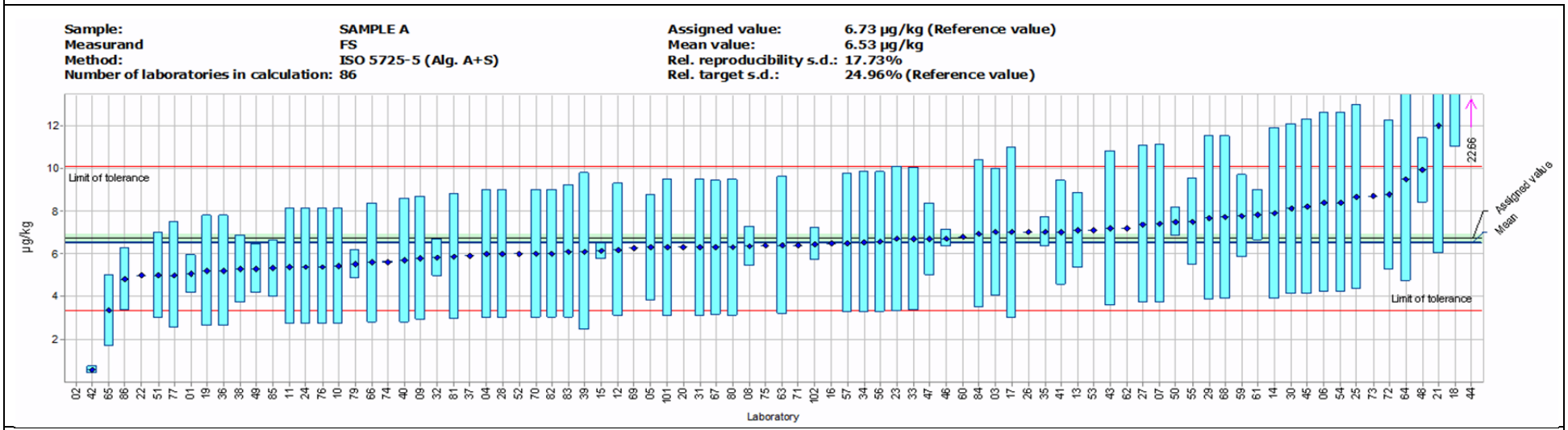
Sample:	**SAMPLE A**	**Assigned value:**	**10.39 µg/kg (Reference value)**
Measurand	**F**	**Mean value:**	**10.05 µg/kg**
Method:	**ISO 5725-5 (Alg. A+S)**	**Rel. reproducibility s.d.:**	**17.24 %**
Number of laboratories in calculation:	**87**	**Rel. target s.d.:**	**25.02 % (Reference value)**



blue rhombus: individual results, blue box: reported expanded measurement uncertainty (k=2),
green line: assigned value, green area around assigned value: expanded uncertainty of the assigned value (k=2),
red lines: lower and upper limit of satisfactory z score range; green band: confidence interval of the assigned value

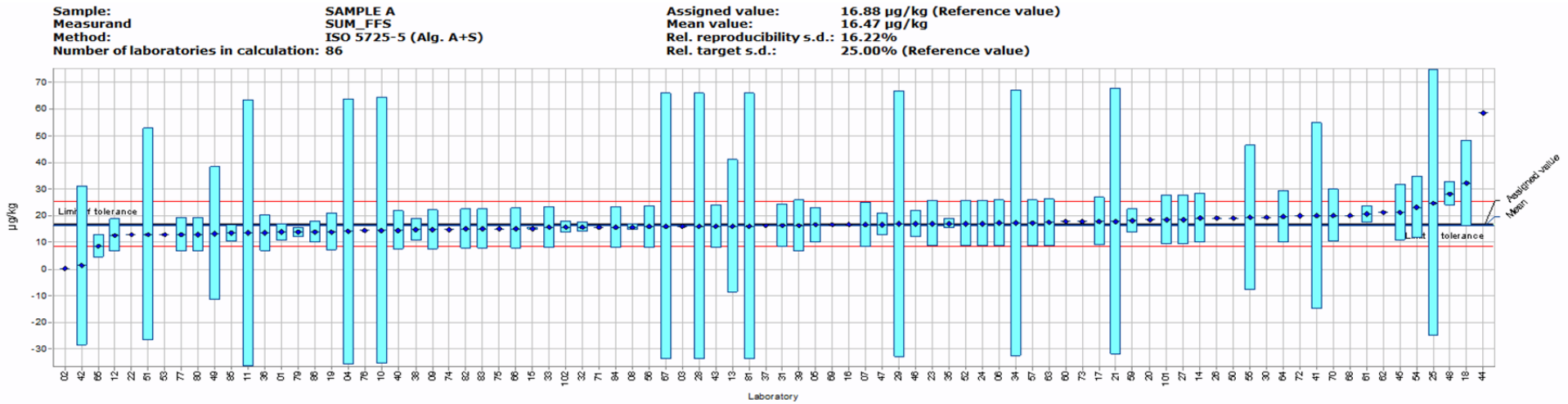
Reported results for Fipronil Sulfone (FS) content in SAMPLE A

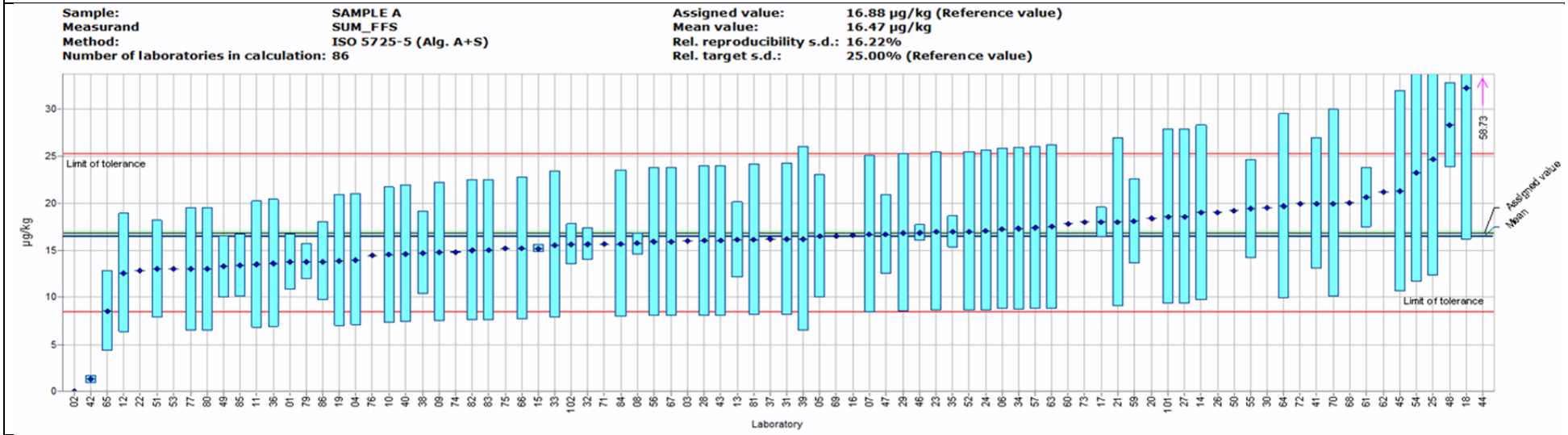




blue rhombus: individual results, blue box: reported expanded measurement uncertainty (k=2),
green line: assigned value, green area around assigned value: expanded uncertainty of the assigned value (k=2),
red lines: lower and upper limit of satisfactory z score range; green band: confidence interval of the assigned value

Reported results for the SUM of Fipronil and Fipronil Sulfone (FFS) content in SAMPLE A

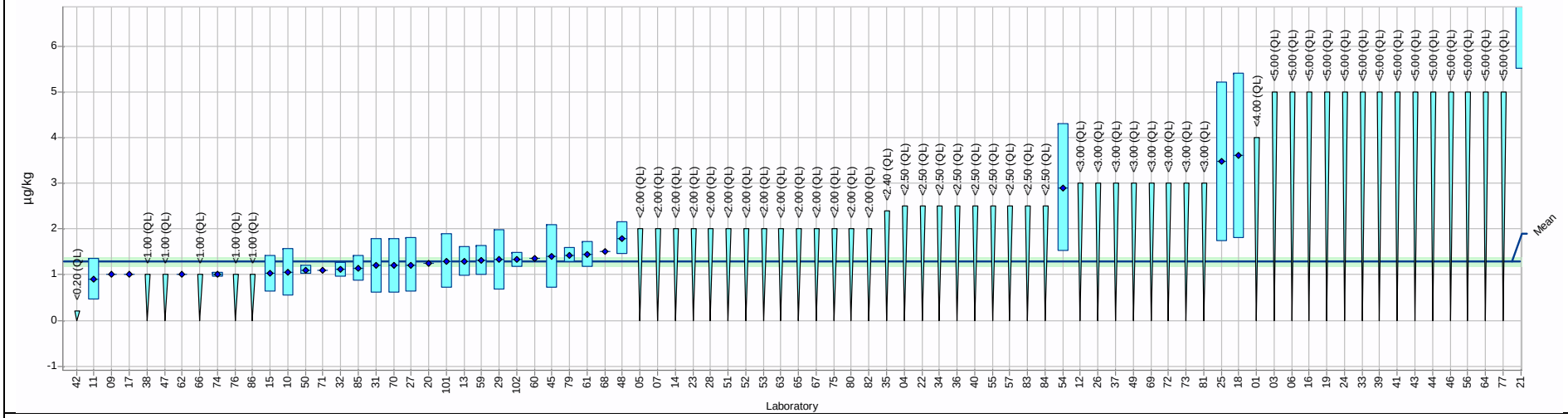




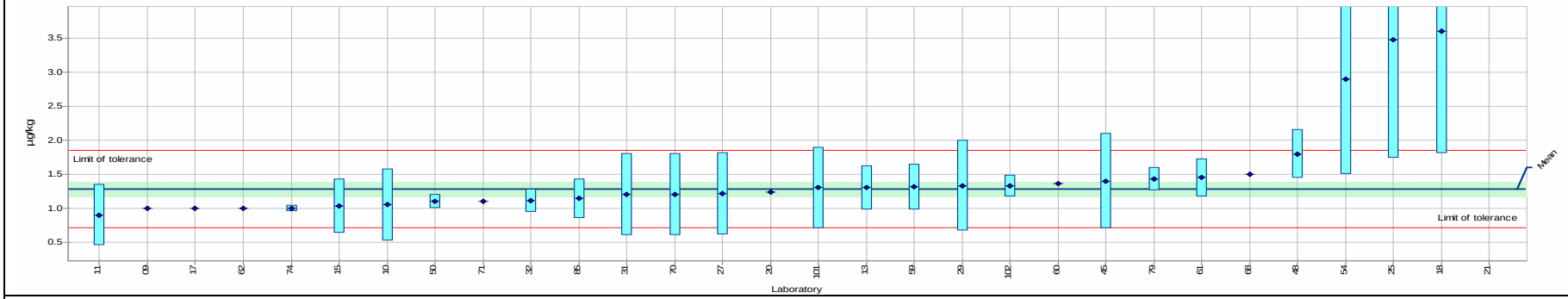
blue rhombus: individual results, blue box: reported expanded measurement uncertainty (k=2),
green line: assigned value, green area around assigned value: expanded uncertainty of the assigned value (k=2),
red lines: lower and upper limit of satisfactory z score range; green band: confidence interval of the assigned value

Reported results for the Fipronil Sulfone (FS) content in SAMPLE B

Sample:	**SAMPLE B**	**Assigned value:**	**1.28 µg/kg (Empirical value)**
Measurand	**FS**	**Mean value:**	**1.28 µg/kg**
Method:	**ISO 5725-5 (Alg. A+S)**	**Rel. reproducibility s.d.:**	**21.28%**
Number of laboratories in calculation:	**30**	**Rel. target s.d.:**	**22.00%**



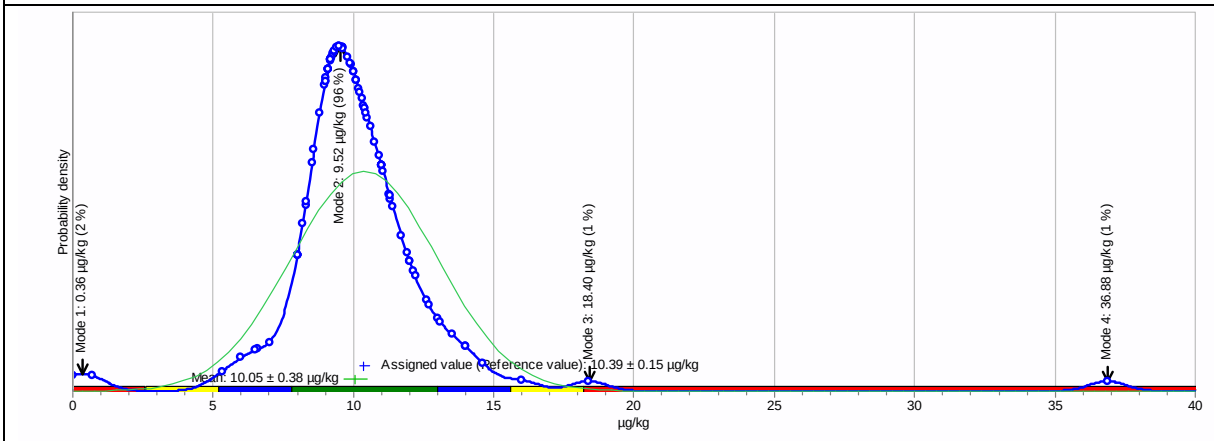
Sample:	**SAMPLE B**	**Assigned value:**	**1.28 µg/kg (Empirical value)**
Measurand	**FS**	**Mean value:**	**1.28 µg/kg**
Method:	**ISO 5725-5 (Alg. A+S)**	**Rel. reproducibility s.d.:**	**21.28%**
Number of laboratories in calculation:	**30**	**Rel. target s.d.:**	**22.00%**



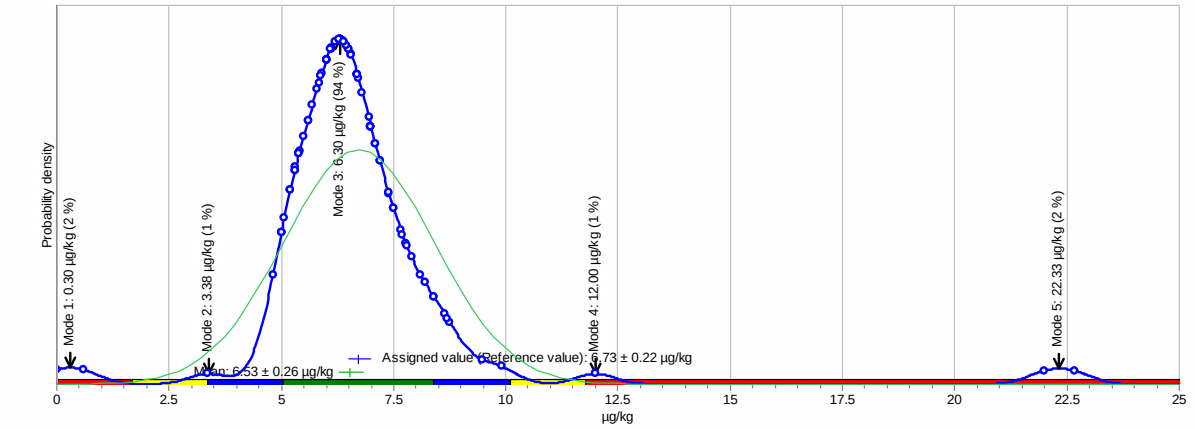
blue rhombus:individual results, blue box: reported expanded measurement uncertainty (k=2),
green line: assigned value, green area around assigned value: expanded uncertainty of the assigned value (k=2),
red lines: lower and upper limit of satisfactory z score range; green band: confidence interval of the assigned value

ANNEX 10: Kernel density plots of the data reported by participants for SAMPLE A

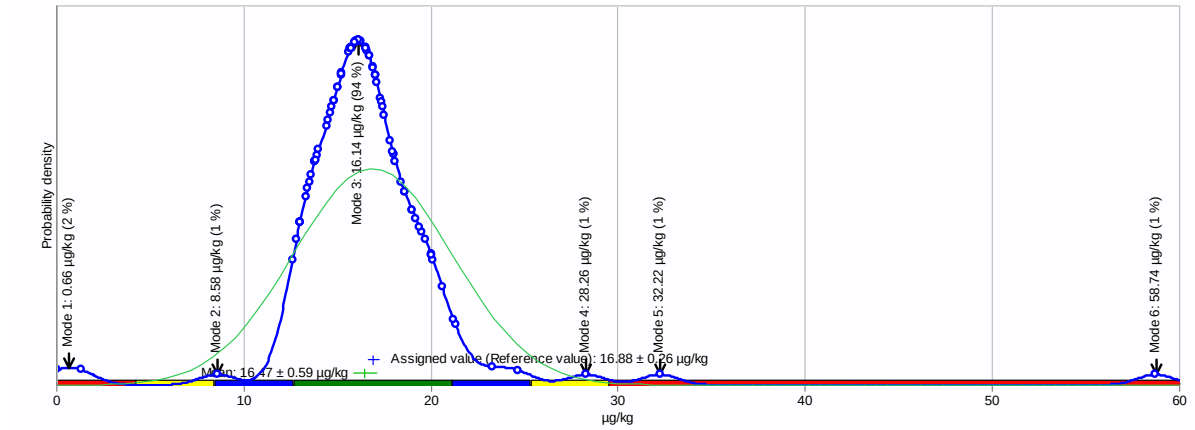
Fipronil content in SAMPLE A



Fipronil Sulfone content in SAMPLE A



SUM of Fipronil and Fipronil Sulfone content in SAMPLE A



Blue dots and lines – Kernel density distribution; green lines – normal distribution

KJ-NA-28806-EN-N

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