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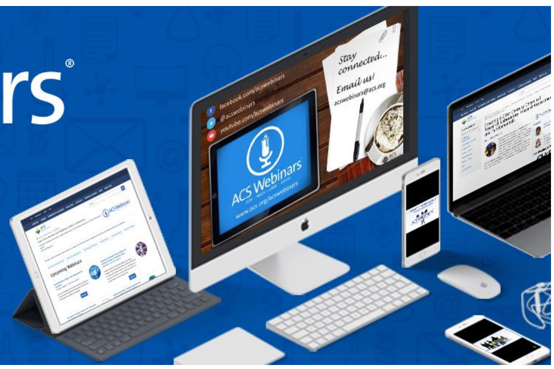

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www.acs.org/heroes

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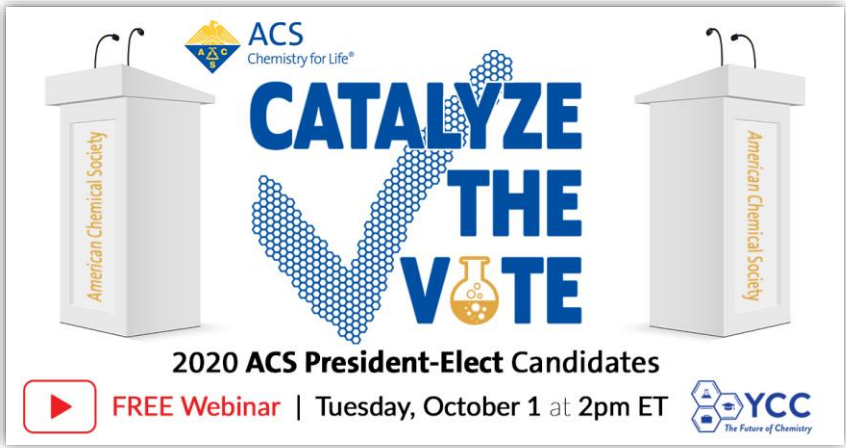

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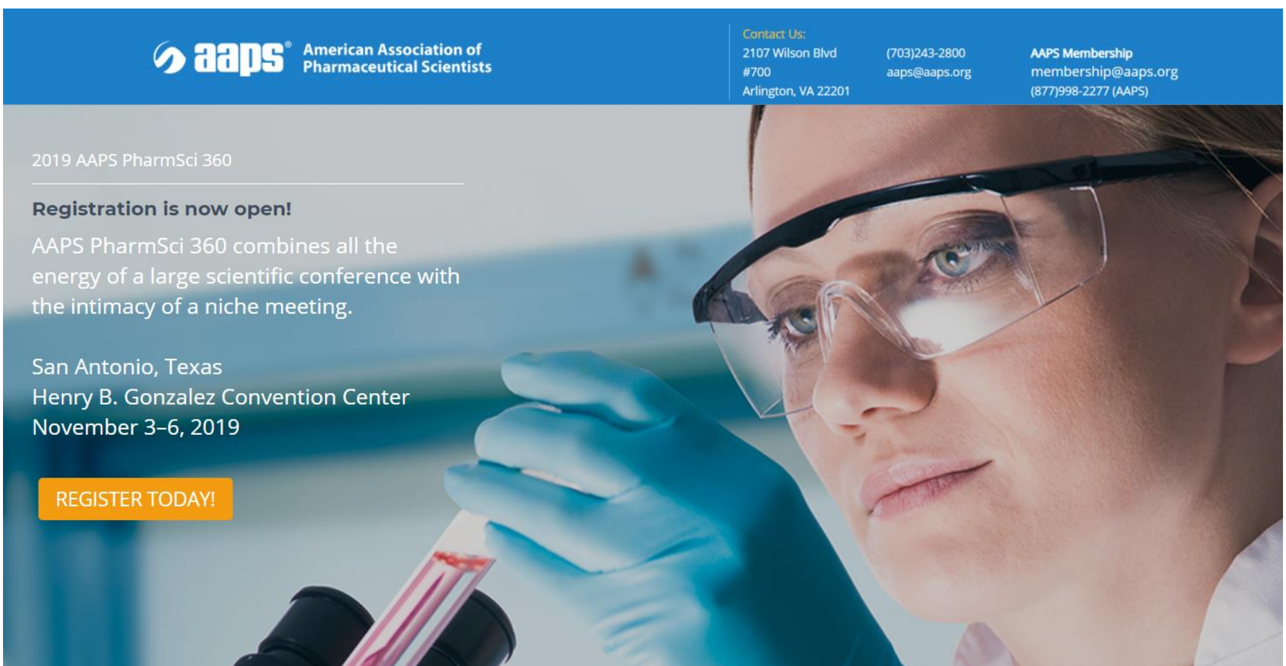

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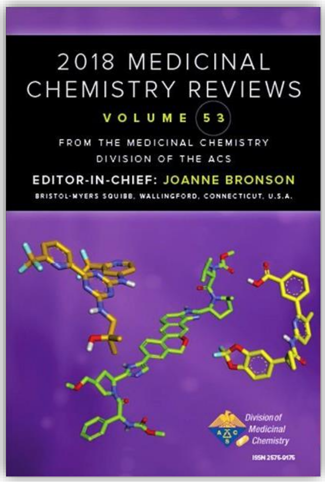

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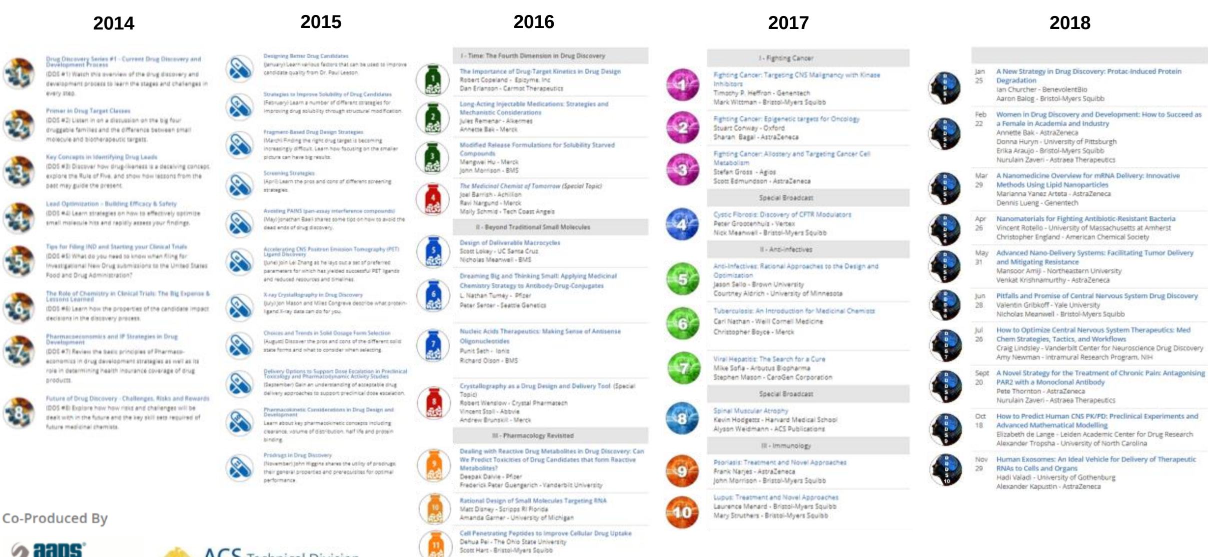

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2019 Drug Design and Delivery Symposium

http://bit.ly/2019DDDS

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Jan 31	**How to Succeed in Drug Discovery: Insight from Medicinal Chemists** (1.5 hrs.)	John Lowe III - JL3 Pharm; Mark Murcko - Relay Therapeutics; Ann Weber - Kallyope; William Greenlee - MedChem Discovery Consulting
Feb 28	**Cosolvent Molecular Dynamics: Mapping Protein Surfaces to Discover Allosteric Sites**	Heather Carlson - University of Michigan; Rommie Amaro - UC San Diego
Mar 28	**Women at the Interface of Computational Chemistry and Drug Discovery** (1.5 hrs)	Zoe Cournia - Biomedical Research Foundation and *JCIM*; Kate Holloway - Gfree Bio; Yvonne C. Martin - Previously of Abbott Laboratories; Shana Posy - Bristol-Myers Squibb
Apr 18	**Effective Exploration of Chemical Space in Hit-Finding**	Hanneke Jansen - Novartis Institutes for BioMedical Research; Zoe Cournia - Biomedical Research Foundation and *JCIM*
May 30	**Widening the Therapeutic Window: Kinetic Selectivity and Target Vulnerability**	Peter Tonge - Stony Brook University and *ACS Infectious Diseases*; Stewart Fisher - C4 Therapeutics
Jun 27	**Precision Control of CRISPR-Cas9**	Amit Choudhary - Broad Institute of Harvard and MIT; Venkat Krishnamurthy - AstraZeneca
Aug 8	**Transformation of Recombinant Cells to FDA Approved Products: Clinical Development to Marketplace** (New Date)	Rodney Ho - University of Washington; Venkat Krishnamurthy - AstraZeneca
Aug 22	**The Evolving Outsourcing Landscape in Pharma R&D: Pros and Cons of Different Models**	Bart DeCorte - MercachemSyncom; Allen Reitz - Fox Chase Chemical Diversity Center
Sep 19	**Thinking Outside the Pillbox: Lead Generation and Optimization in Crop Protection Research**	Fides Benfatti - Syngenta
Oct 24	**Treating Diabetes: Designing the Once-Weekly and Oral GLP-1 Semaglutide**	Jesper Lau - Novo Nordisk A/S
Nov 28	**Prodrugs**	Jarkko Rautio - University of Eastern Finland

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Meet the Organizers

- Annette Bak, AstraZeneca, AAPS
- Nicholas Meanwell, Bristol-Myers Squibb, ACS MEDI
- Janice Silverman, American Chemical Society
- John Morrison, Bristol-Myers Squibb, AAPS

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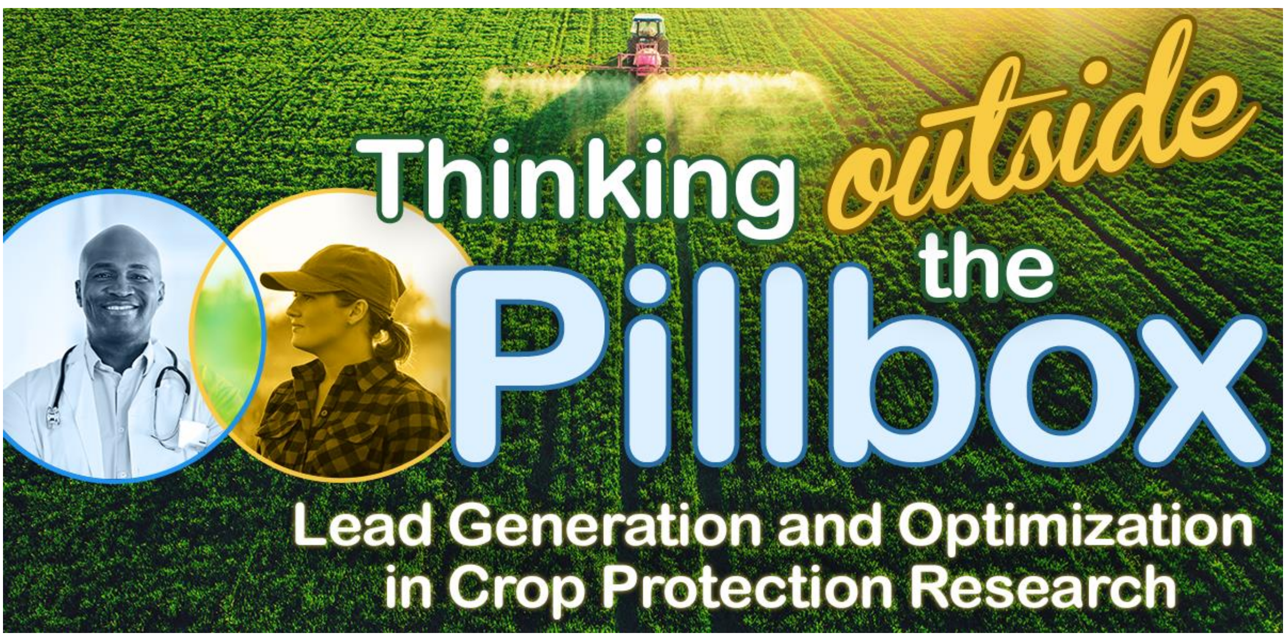

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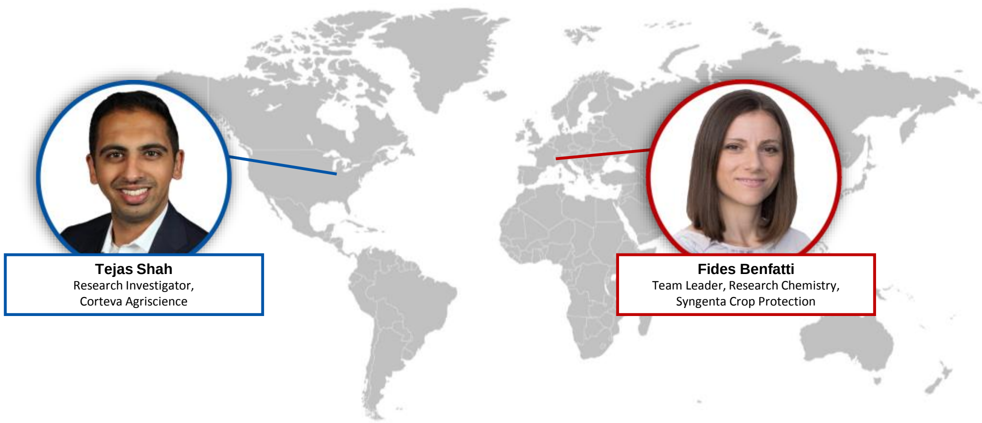
16

Thinking Outside the Pillbox: Lead Generation and Optimization in Crop Protection Research



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Thinking Outside the Pillbox: Lead Generation and Optimization in Crop Protection Research

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Fides Benfatti
Senior Team Leader Chemical Research

Syngenta Crop Protection
fides.benfatti@syngenta.com

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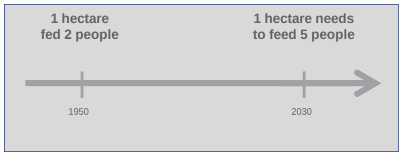
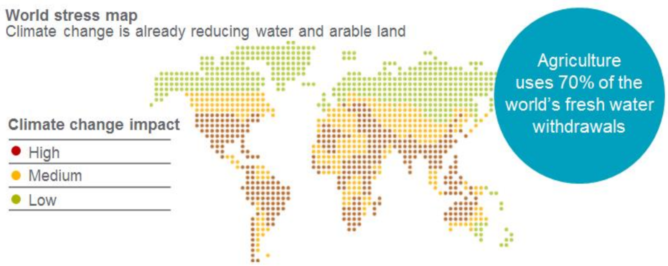
Why do we need new Crop Protection products?

- **Growing more with less**



Source: UNEP, Cline, Syngenta

- **Climate change**



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Why do we need new Crop Protection products?

- **Resistance development** and **shifting pest populations**

- **Changing regulatory landscape**

Source: www.plantmanagementnetwork.org

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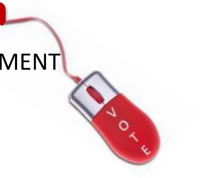
Audience Survey Question

ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT



Approximately how much was lost in billons (USD) **to plant diseases and infestations from 2005 to 2015?**

- 1 billion
- 3.5 billion
- 5 billion
- 7 billion
- 9.5 billion

** If your answer differs greatly from the choices above tell us in the chat!*

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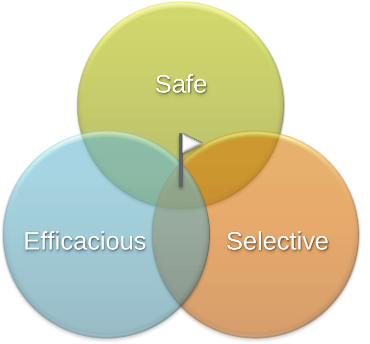
How are crop protection products discovered and optimized

Outline

- ❖ Discovery of insecticidal spiroindolines
- ❖ Bioavailability-guided design of new aphicides
- ❖ Modern crop protection products targeting ion channels



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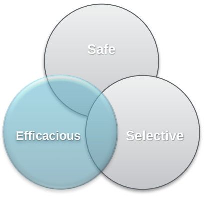
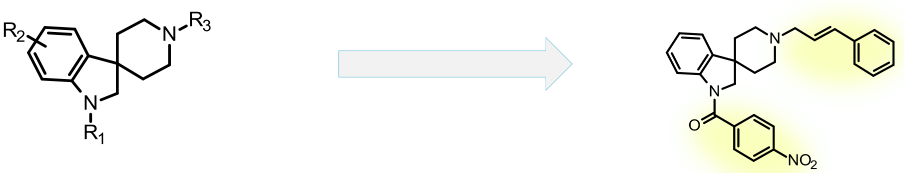
Discovery of insecticidal spiroindolines





Spiro[indoline-3,4'-piperidine]

Insecticidal Hit!

- Screening of a chemical library (obtained from Oxford Asymmetry, now Evotec)

- Insecticidal hit compound identified by high throughput screening
- Activity on *Drosophila melanogaster*, *Plutella xylostella* and *Heliothis virescens* at 1000 ppm
- Only compounds possessing a cinnamyl group displayed insecticidal activity

Hughes, D. J.; Worthington, P. A.; Russell, C. A.; Clarke, E. D.; Peace, J. E.; Ashton, M. R.; Coulter, T. S.; Roberts, R. S.; Molleyres, L.-P.; Cederbaum, F.; Cassayre, J.; Maienfisch, P. **WO2003106457**

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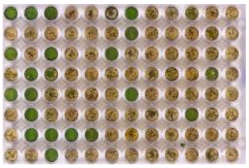
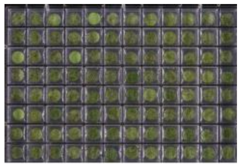
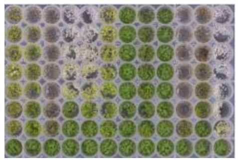
Agrochemical research: screening cascade

- *In vivo* on-target test from day 1


Insecticide HTS


Fungicide HTS


Herbicide HTS

- Increase in tests size parallel to project stage (HTS >>> field)

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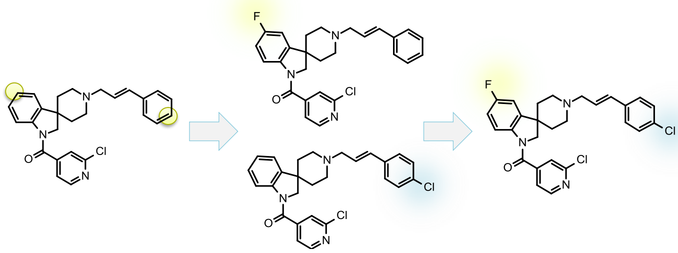
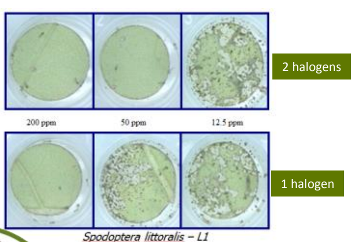
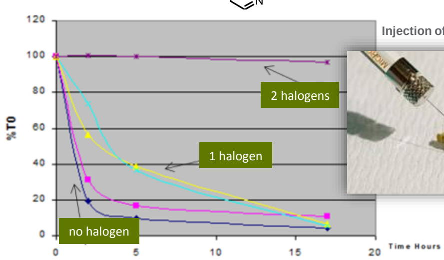
Discovery of insecticidal spiroindolines: Hit-to-Lead Optimization

Block metabolically weak positions
→ improved potency
→ cumulative effect







Injection of compounds in *Heliothis virescens*

Hughes, D. J.; Worthington, P. A.; Russell, C. A.; Clarke, E. D.; Peace, J. E.; Ashton, M. R.; Coulter, T. S.; Roberts, R. S.; Molleyres, L.-P.; Cederbaum, F.; Cassayre, J.; Maienfisch, P. **WO2003106457**

Sluder A.; Shah, S.; Cassayre, J.; Clover R.; Maienfisch, P.; Molleyres, L.-P. et al *PLoS One* **2012**, 7(5), e34712

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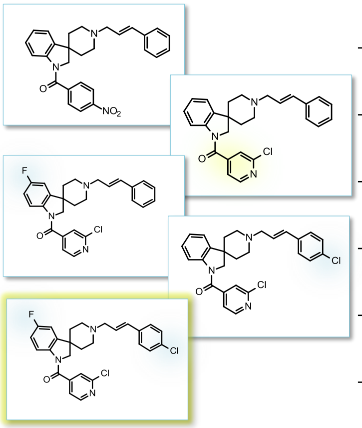
Discovery of insecticidal spiroindolines: Hit-to-Lead Optimization

- Cumulative effect observed cross lepidopteran target species
- Lead shows promising activity at low dose

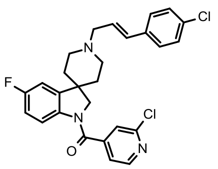


Spodoptera littoralis L1	Heliothis virescens L1	Plutella xylostella L2
> 500	> 500	>500
200	200	200
200	50	200
50	≤12	<50
12	0.8	3

Primary screening [EC80-100 in ppm (mg/L)]

Properties and insecticidal activity of spiroindoline lead

Spiroindoline Lead



Melting Point (M.p.)	168-170°C
Aqueous solubility	5 ppm@pH 6.5
Log P	5.94
pKa	7.88
PhotostabilityT_{50}	114 mins
Rat acute toxicity	MLD_{50} > 200 mg/Kg

Lepidopteran control (activity given as effective concentration EC_{80} in ppm (mg / L)

effective concentration EC_{80} in ppm (mg / L)	*Spodoptera littoralis L1*	*Heliothis virescens L1*	*Plutella xylostella L3*	*Cydia pomonella L1*
Spiroindoline 6b	**12**	**0.8**	**6**	**3**
Spinosad	0.8	0.8	0. 2	12
Indoxacarb	3	3	3	12

Check

Spinosad 10 g / hl*

Spirodindoline 8 g / hl*

Diamondback moth, *Plutella xylostella*, on cabbage

Symptomology – insights into the mode of action

- Exploiting genetic model systems – *C. elegans* and *Drosophila*

Decades of academic work linking <u>phenotype</u> (visible effects) to genetic disfunction

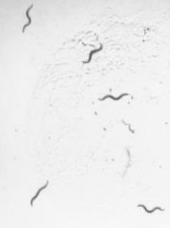
- Comparing chemical symptoms to genetic phenotypes identifies candidate target proteins e.g. Spiroindolines

For a recent extensive analysis of *C. elegans* behavorial phenotype:
A database of *Caenorhabditis elegans* behavioral phenotypes, E. Yemini *et al* *Nature Methods,* **2013**, *10*, 877-879

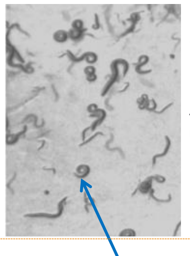
Symptomology in *C. elegans*

- Spiroindolines induces characteristic symptoms in *C. elegans*

C. elegans untreated



+ spiroindoline



Genetic mutation in acetylcholine signalling discovered in 1993

(Alfonso *et al.* The Caenorhabditis elegans unc-17 gene: a putative vesicular acetylcholine transporter. Science, **1993**, *261*, 617-9.)

Photo: Syngenta

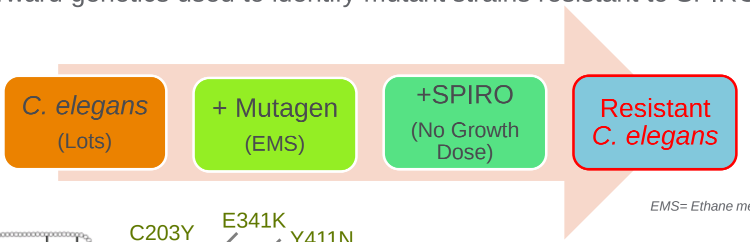
Symptom resembles phenotype

Hypothesis: Spiroindolines affect cholinergic signaling

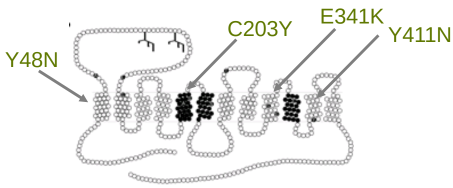
PLoS ONE **2012**, *7*, e34712.

Confirming the hypothesis

- Forward genetics used to identify mutant strains resistant to SPIRO



EMS= Ethane methyl sulfonate



Locate resistance mutations using genetic mapping and sequencing

- Resistance mutations locate to **vesicular acetylcholine transporter** (VAChT)
- Binding of spiroindoline to VAChT can now be confirmed using standard biochemical approaches *in vitro*

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Bioavailability-guided design of new aphicides

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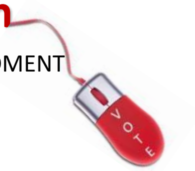
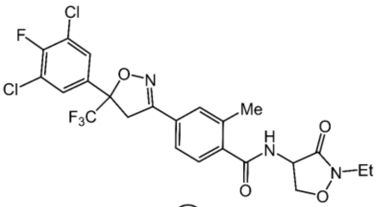
Audience Survey Question

ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT

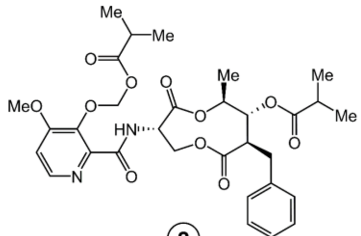


Which of these molecules is an or are agrochemical(s)?

(Select all that apply)



1



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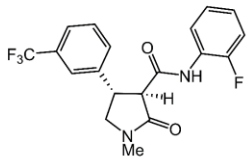

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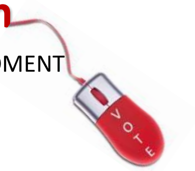
** If your answer differs greatly from the choices above tell us in the chat!*

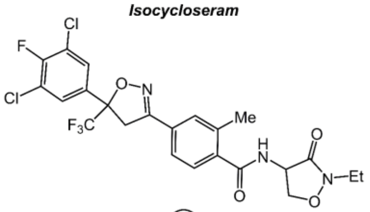
Audience Survey Question

ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT



Which of these molecules is an or are agrochemical(s)?

Isocycloseram



1

Insecticide

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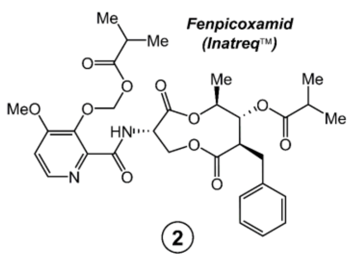
Fenpicoxamid (Inatreq™)

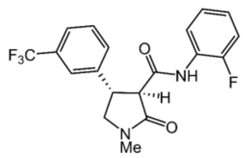


2

Fungicide

CORTEVA™ agriscience

Tetflupyrolimet



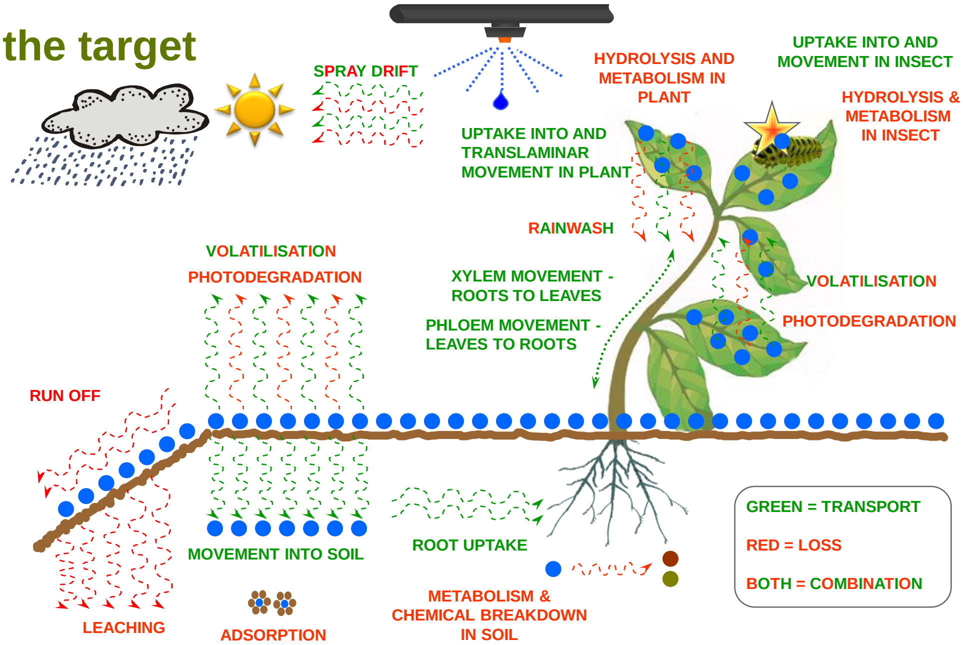
3

Herbicide

FMC

** If your answer differs greatly from the choices above tell us in the chat!*

How to reach the target



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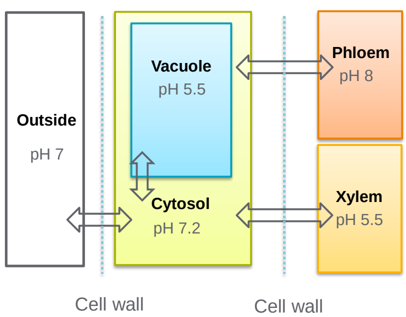
Influence of physical chemical properties on the uptake into plants



The **logP** and **pka** of agrochemicals dictate their distribution in plants

- Weak acids with intermediate lipophilicity get trapped in phloem
- Basic molecules get trapped in vacuoles
- Caveat: carrier proteins, channels can provide active transport

Trapp S. 2009 in J. Devillers, Ed.: Ecotoxicology Modeling. Springer, Dordrecht
Buchholz, A.; O'Sullivan, A. C.; Trapp, S. In *Discovery and Synthesis of Crop Protection Products*; American Chemical Society: 2015; Vol. 1204, p 93.

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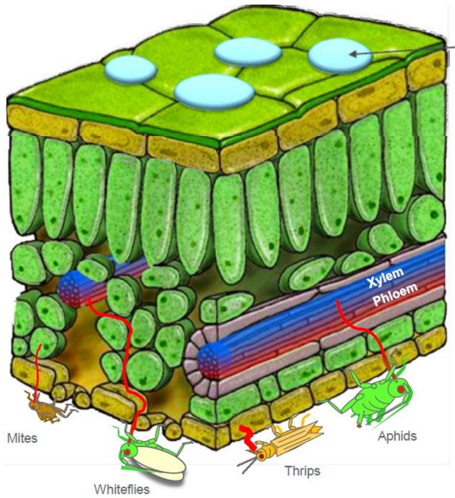
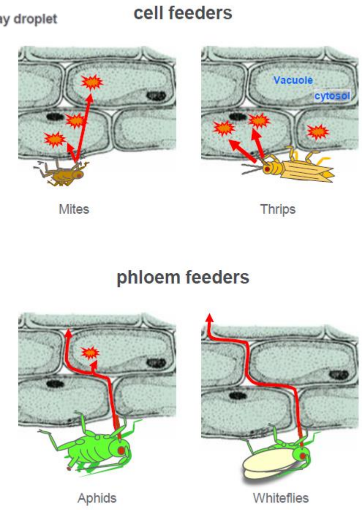
Feeding behaviour of insect pests





leaf modified after 'Digital frog international ©'

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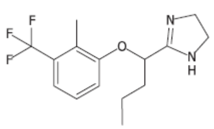
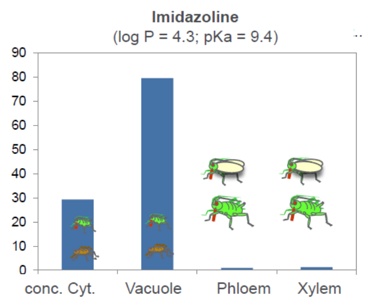
Which compound won't be efficacious for aphids control?

Imidazoline



Basic



Remember:

- Aphids are phloem and xylem feeders
- Weak acids get trapped in phloem
- Basic molecules get trapped in vacuoles (i.e. cells)

Buchholz, A., Trapp, S., Pest Manag. Sci. (2016); 72: 929–939

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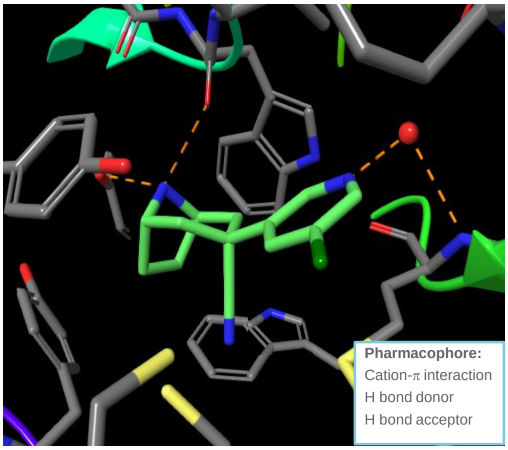
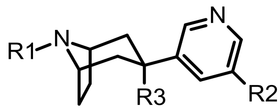
nAChR agonists for aphids control



AChBP co-crystal structure with lead



Our target
- Low lipophilicity (logP < 2.5)
- ✓ Low molecular volume
- Non-basic

Proc. Brighton Crop Protection Conf: Pest and Diseases **2002**, *1*, 145 – 152.

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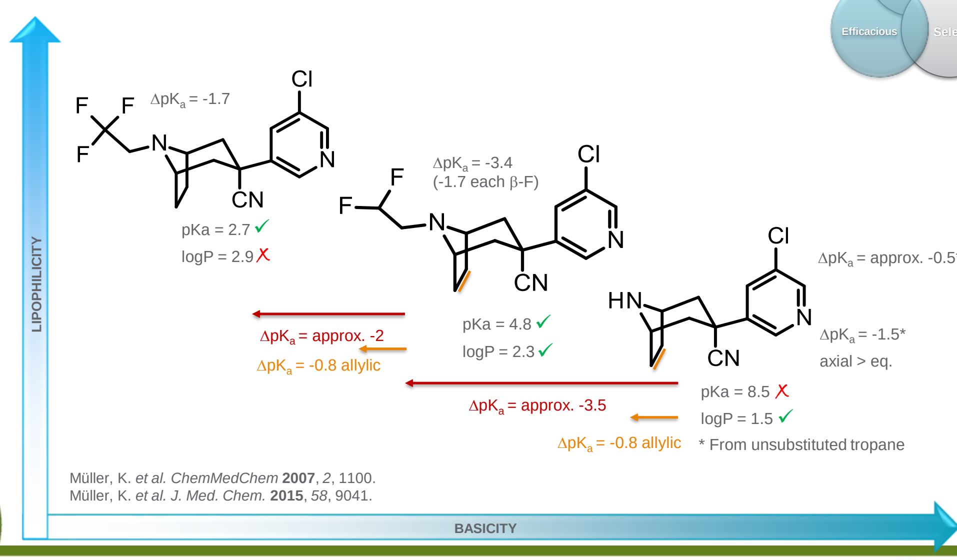
Tuning physicochemical properties



Müller, K. *et al. ChemMedChem* **2007**, *2*, 1100.
Müller, K. *et al. J. Med. Chem.* **2015**, *58*, 9041.

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Modern crop protection products targeting ion channels

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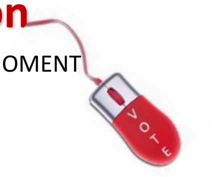
Audience Survey Question

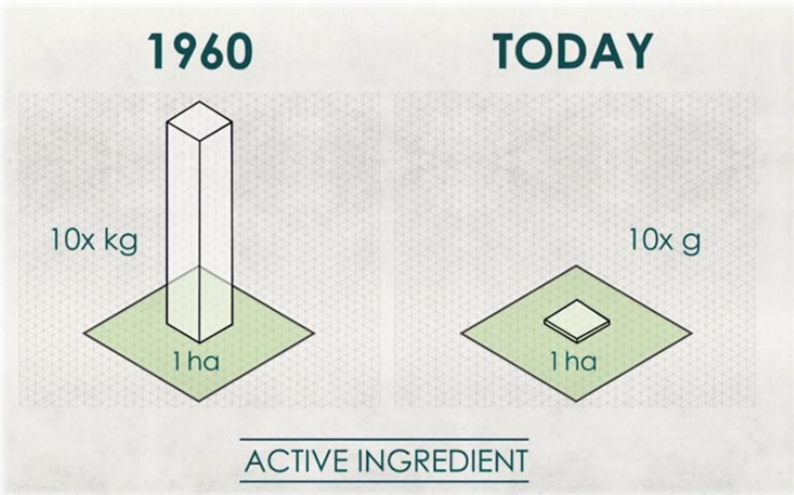
ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT



The crop science in the 1960's used 10 Kgs of active ingredient per hectare to protect crops (1 hectare is almost 2.5 acres). **How much active ingredient is used today in comparison?**

- 1 kilogram
- 500 grams
- 100 grams
- 10 grams
- 1 gram

** If your answer differs greatly from the choices above tell us in the chat!*



https://croplife.org/crop-protection/innovation-in-crop-protection-products/

43

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Product Safety – Who is being protected?

44

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Product safety: studies per stage

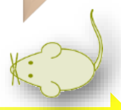
Lead Generation → Lead Exploration → Lead Optimisation → Candidate Confirmation

Setting Direction
Mode of action alerts
Structure activity alerts
Competitor analysis

In silico, in vitro

Compound design and selection
Acute toxicity (oral/dermal/irritation)
Skin sensitization
14/28 day rat and mouse
Preliminary Pharmacokinetics (PK)
Genotoxicity screening
Dermal absorption

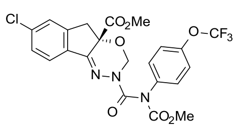
ADME
28 day dog
90 day rat and mouse
Preliminary development toxicity
Genotoxicity evaluation
Preliminary risk assessments

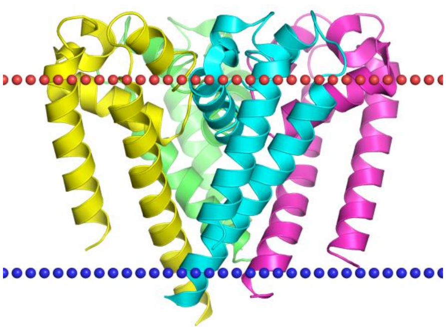


In vivo

Predicting Regulatory Outcome
Acute toxicity
Genotoxicity
Carcinogenicity/Chronic 2 year study
Reproductive toxicity
Developmental toxicity
Neurotoxicity
90 day & 1 year dog
Dermal absorption
ADME – metabolites
Metabolite testing
Manufacturing intermediates
Formulation toxicity
Definitive risk assessments

Efficacious, safe and selective: Indoxacarb





Spodoptera litura (Oriental leafworm moth) feeding on a cabbage leaf

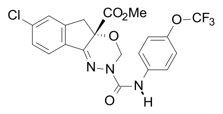
- Indoxacarb (DuPont) is an insecticide that exerts its mode of action by targeting the Voltage-gated Sodium channel

Channels **2008**, *2*, 100.
Bioorg. Med. Chem. Lett. **2014**, *24*, 3690.
J. Med. Chem. **2015**, *58*, 7093–7118

- It is active against Lepidoptera (moths)

Mammalian safety of Indoxacarb

Differential binding

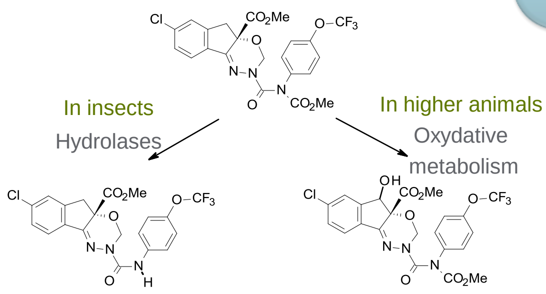


DCJW   Active ingredient

IC_{50} = **1000** nM on rat Na_V 1.4
IC_{50} = **25** nM on Bg Na_V 1-1a
(Bg = Blattella Germanica)

Pestic. Biochem. Physiol. **2010**, *97*, 87.

Selective metabolism



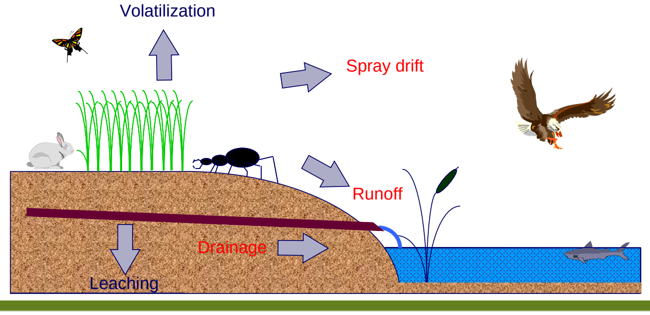
DCJW

+ other metabolites

Crop Protection **2000**, *19*, 537.
Indoxacarb, *JMPR* Evaluation report 2005.

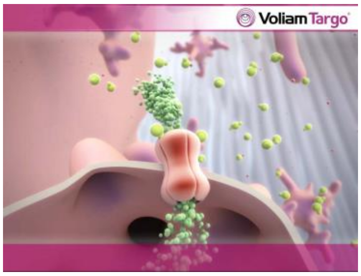
Environmental Safety – What is being protected?

- **Groundwater** – Drinking water (human exposure), irrigation water, the aquifer itself as an entity
- **Surface Water** – Drinking water, irrigation water, aquatic organisms (fish and aquatic plants)
- **Soil** – Persistence in soil, topsoil erosion, carry-over into follow on crops
- **Non target insects, plants and the organisms**
  - Bees, beneficial insects, worms, off target plant species, birds, field dwelling mammals
- **Air** – long range transport, atmospheric degradation, vapour movement



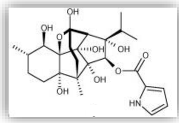
Efficacious, safe and selective: Anthranilic amides

Anthranilic amides bind to the insect ryanodine receptor in muscle cells



Intrinsic target-based selectivity:
Insect vs. Mammal

	EC50 (nM)
Cockroach	36
Fruit Fly	48
Mouse	14'000
Rat	> 100'000



Ryanodine (plant metabolite)

VOLIAM TARGO | **Untreated**

Active in the field at rates as low as **5 g/ha**!
(typical rate for organophosphates = **1 Kg/ha**)

Crop: Cauliflower
Source: Taiwan, Syngenta trials 2007

49

Pest Manage. Sci. **2013**, *69*, 7.; *Invert. Neurosci.* **2008**, *8*, 107-19; *Biochemistry*, **2009**, *48*, 10342

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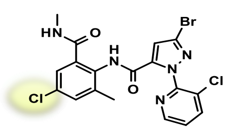
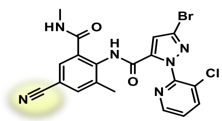
Modulation of soil persistance



Chlorantraniliprole (DuPont)

M.p. (°C) 208-210
logP 2.76
pKa (acid) 10.8

Water solubility (mg/l, 20-25 °C) 1.0
Water DT50 10 d (pH 9, 25 °C)
Soil DT50 < 2–12 mo



Cyantraniliprole (DuPont/Syngenta)

M.p. (°C) 224
logP 1.94
pKa (acid) 8.8

Water solubility (mg/l, 20-25 °C) 14.24
Water DT50 < 1 d (pH 9, 25 °C)
Soil DT50 average 32 d

Improved plant mobility
Increased spectrum of insect control (aphids and leafhoppers)

(Data from BCPC Pesticide Manual)

50

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Product Safety
Steve Hadfield
Caroline Winn
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***C. elegans* biology**
Anthony Flemming
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* University of Manchester
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Bringing plant potential to life

Bringing plant potential to life

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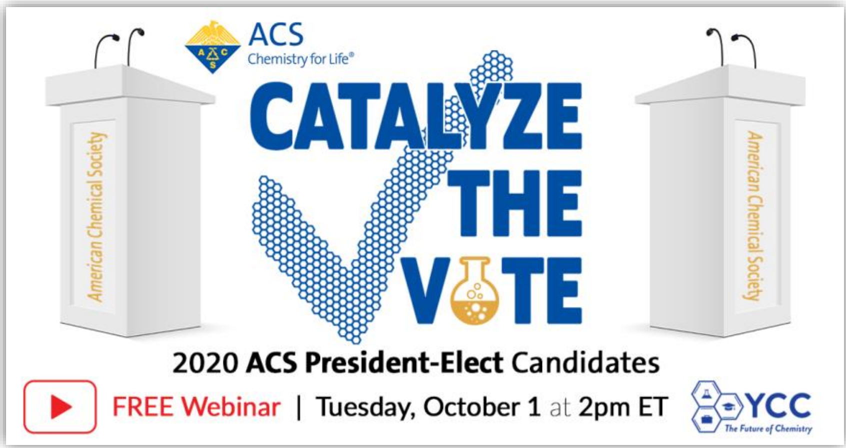

https://www.acs.org/content/acs/en/acs-webinars/popular-chemistry/catalyze-the-vote-2020.html

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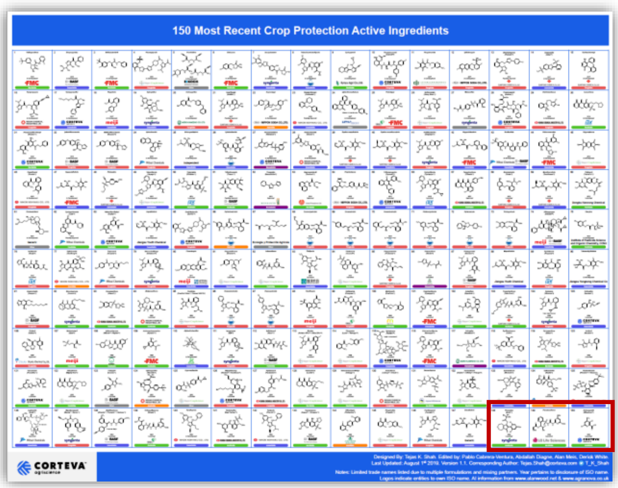
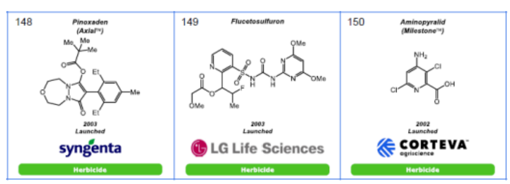
Webinar Additional Resource!

150 Most Recent Crop Protection Active Ingredients





https://www.acs.org/content/dam/acsorg/events/drug-discovery/slides/2019-09-19-recent-crop-protection-poster.pdf

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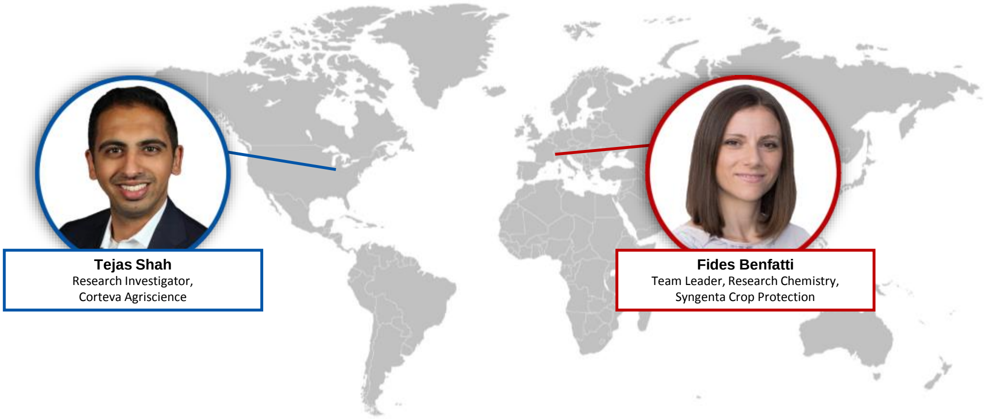

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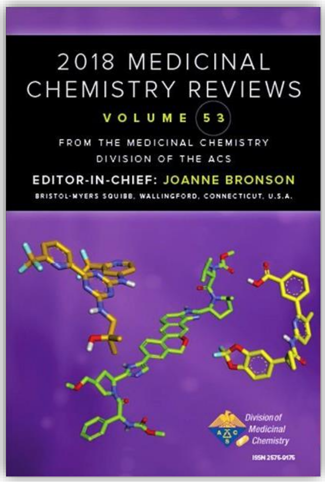

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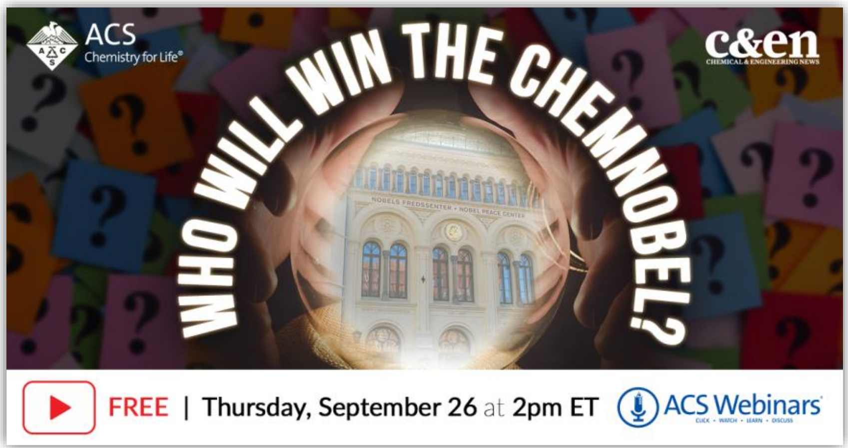

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