

Table 14. The limit of detection (LOD), the limit of quantification (LOQ), and r^2 values for ELISA standard curves for five antibiotics tested in this study.

Antibiotics	The limit of detection (μgL^{-1})	The limit of quantification (μgL^{-1})	r^2
Monensin	0.49	1.18	>0.99
Sulfamethazine	0.11	0.33	0.97~0.99
Tylosin	1.48	1.53	0.97~1
Virginiamycin	0.47	0.75	0.97~0.98
Tetracycline	0.05	0.35	0.93~0.97

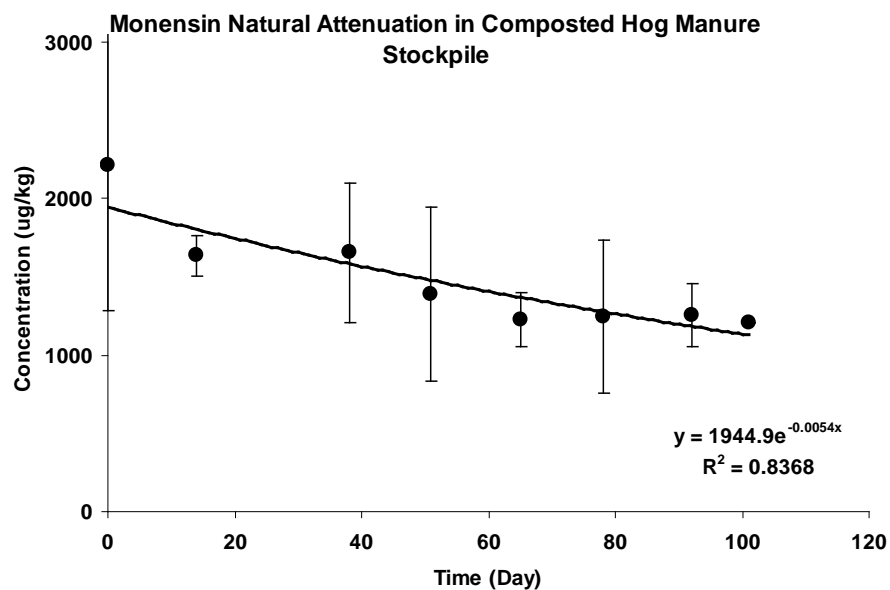


Figure 13. Monensin degradation during composting of hog manure with wood shavings.

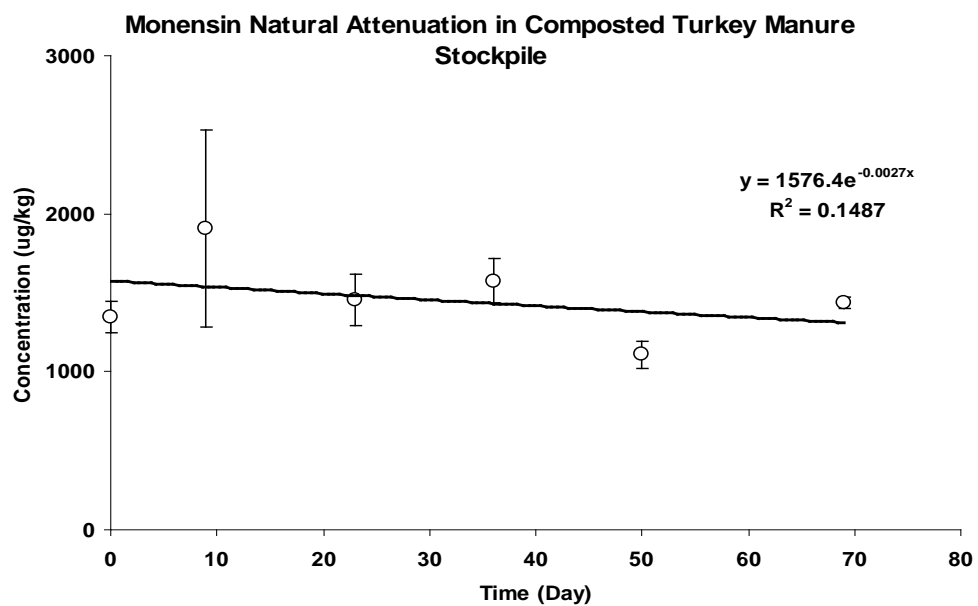


Figure 14: Monensin degradation during composting of turkey manure.

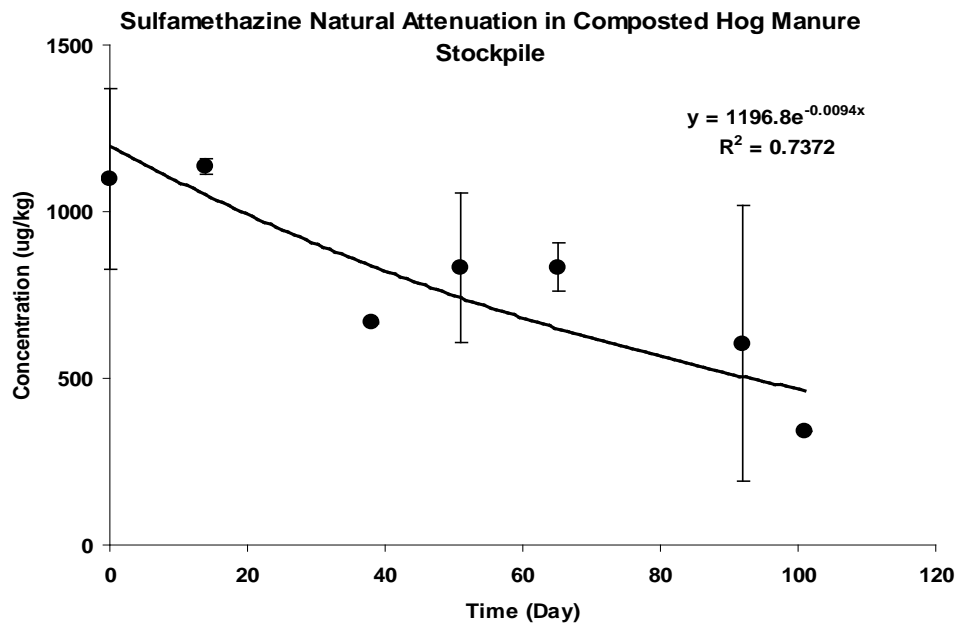


Figure 15: Sulfamethazine degradation during composting of hog manure with wood shavings.

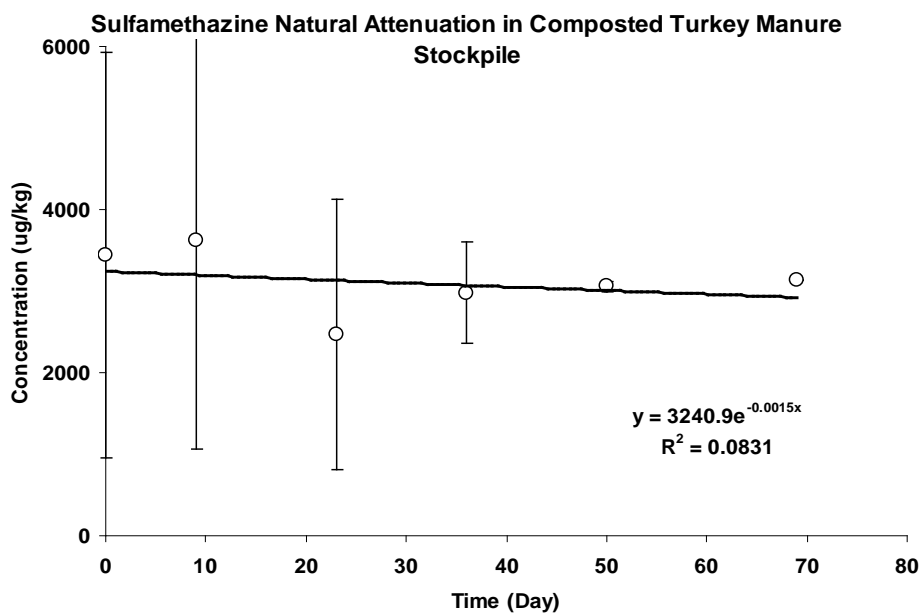


Figure 16: Sulfamethazine degradation during composting of turkey manure.

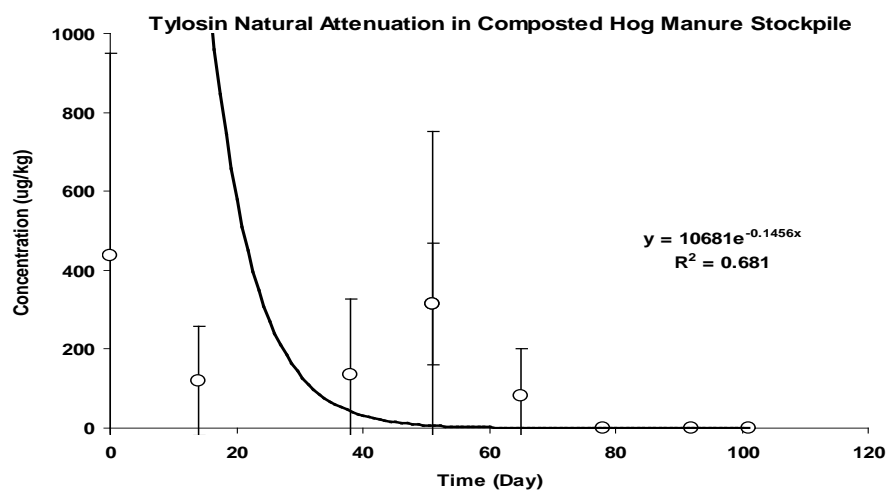


Figure 17: Tylosin degradation during composting of hog manure with wood shavings.

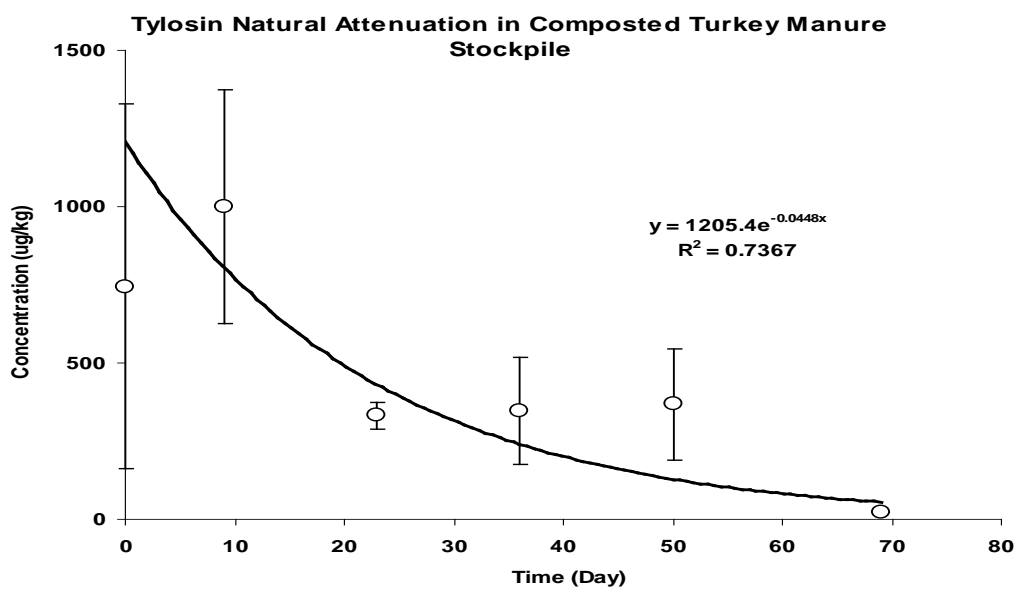


Figure 18: Tylosin degradation during composting of turkey manure.

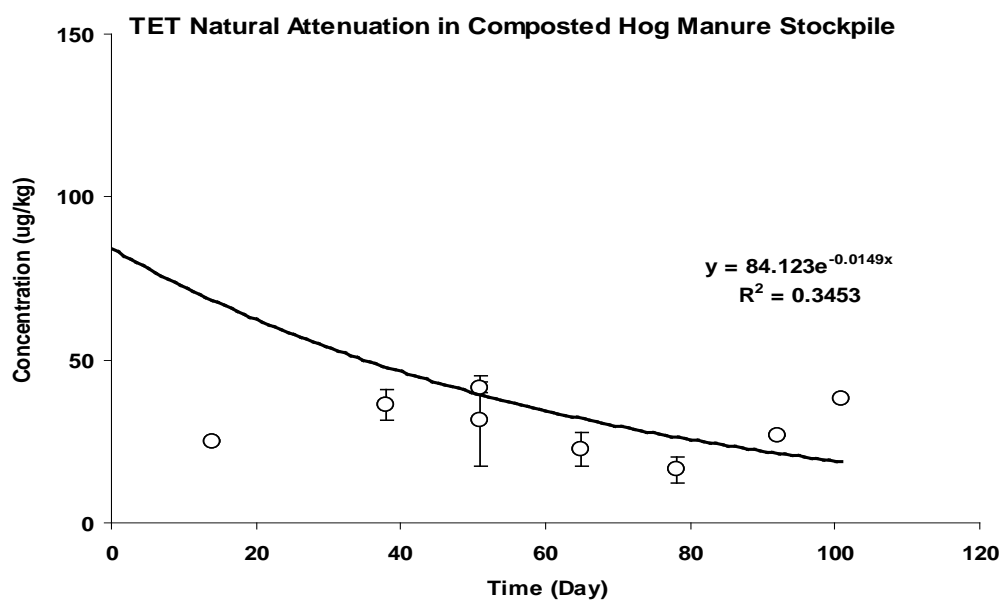


Figure 19: Chlortetracycline degradation during composting of hog manure with wood shavings.

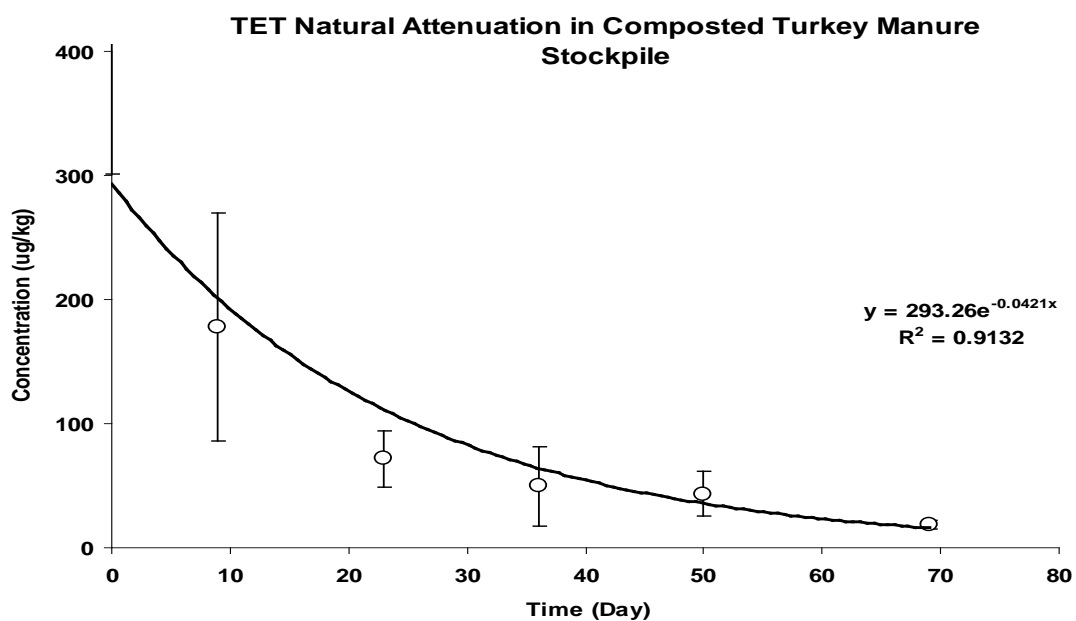


Figure 20: Chlortetracycline degradation during composting of turkey manure.

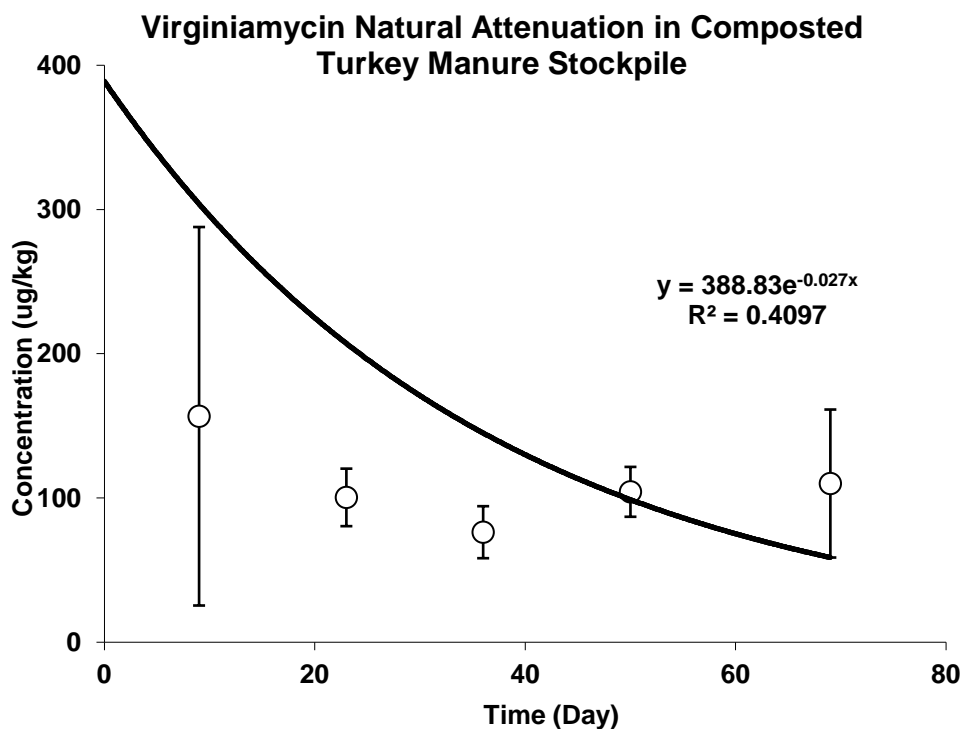


Figure 21: Virginiamycin degradation during composting of turkey manure.

Table 15: First order degradation rate constant ($-k$) and half-life ($t_{1/2}$) of monensin, sulfamethazine, tylosin, virginiamycin, and tetracycline during composting of hog and turkey manures.

Antibiotic	Hog Manure		Turkey Manure	
	μ, d^{-1}	$t_{1/2}, d$	μ, d^{-1}	$t_{1/2}, d$
Monensin	-0.0054	128.4	-0.0027	256.7
Sulfamethazine	-0.0094	73.7	-0.0015	462.1
Tylosin	-0.1456	4.8	-0.0448	15.5
Virginiamycin	N/A	N/A	-0.0274	25.3
Chlortetracycline	-0.0149	46.2	-0.0421	16.5

Table 16. Antibiotics concentrations in fresh and composted hog and turkey manures at the time of their application.

Antibiotics	Raw Hog $\mu\text{g L}^{-1}$	Composted Hog $\mu\text{g kg}^{-1}$	Raw Turkey $\mu\text{g kg}^{-1}$	Composted Turkey $\mu\text{g kg}^{-1}$
Monensin	3240.2	1205.0	1249.3	1432.8
Sulfamethazine	2546.6	341.1	4353.5	3128.3
Tylosin	171.8	0.0	291.2	22.4
Virginiamycin	N.A. [§]	N.A.	27.9	109.9
Chlortetracycline	122.8	38.2	13.9	18.8

[§]N.A. -Not added

Table 17: Water contents of plant parts and soil at the time of vegetable harvest. Vegetable moisture content is based on wet weight basis whereas soil moisture content is based on dry weight basis.

Vegetable	Date of Harvest		Plant/soil	Moisture Content (%)	
	Waseca	Staples		Waseca	Staples
Radish	6/11/2009	7/6/2009	Root	96.3	95.1
			Skin	94.7	93.7
			Soil	38.9	8.9
Spinach	6/15/2009	7/6/2009	Leaf	89.0	92.1
			Soil	30.6	6.1
Lettuce	6/22/2009	7/9/2009	Leaf	95.5	94.4
			Soil	28.8	9.2
Carrot	7/24/2009	8/25/2009	Root	90.5	88.4
			Skin	90.5	89.8
			Soil	24.6	10.2
Garlic	8/4/2009	8/25/2009	Bulb	N.A	61.6
			Soil	29.7	13.6
	6/29/2009	6/29/2009	Scape	81.4	76.6
Cabbage	8/14/2009	8/11/2009	Leaf	92.7	91.7
			Soil	N.A	11.5
Onion	8/14/2009	8/11/2009	Bulb	93.9	94.6
			Soil	23.8	13.8
Pepper	8/15/2009	8/11/2009	Fruit	94.5	94.5
Potato	8/19/2009	8/25/2009	Tuber	66.4	77.5
			Skin	81.9	83.6
			Soil	23.4	11.9
Tomato	9/1/2009	8/25/2009	Tomato	95.2	94.7
			Soil	23.6	10.6
Corn	9/1/2009	9/10/2009	Corn kernels	74.3	74.1
			Soil	26.5	12.0

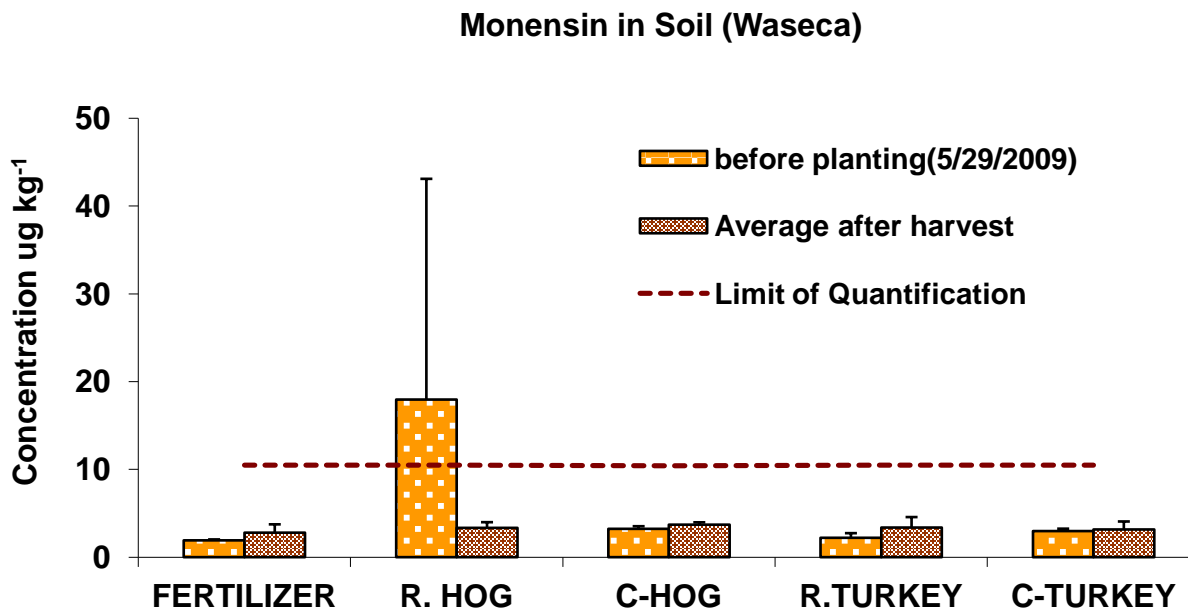


Figure 22. Monensin concentration in manure applied soils before planting and after harvest at Waseca, MN. Prefix R and C refer to raw and composted hog or turkey manure.

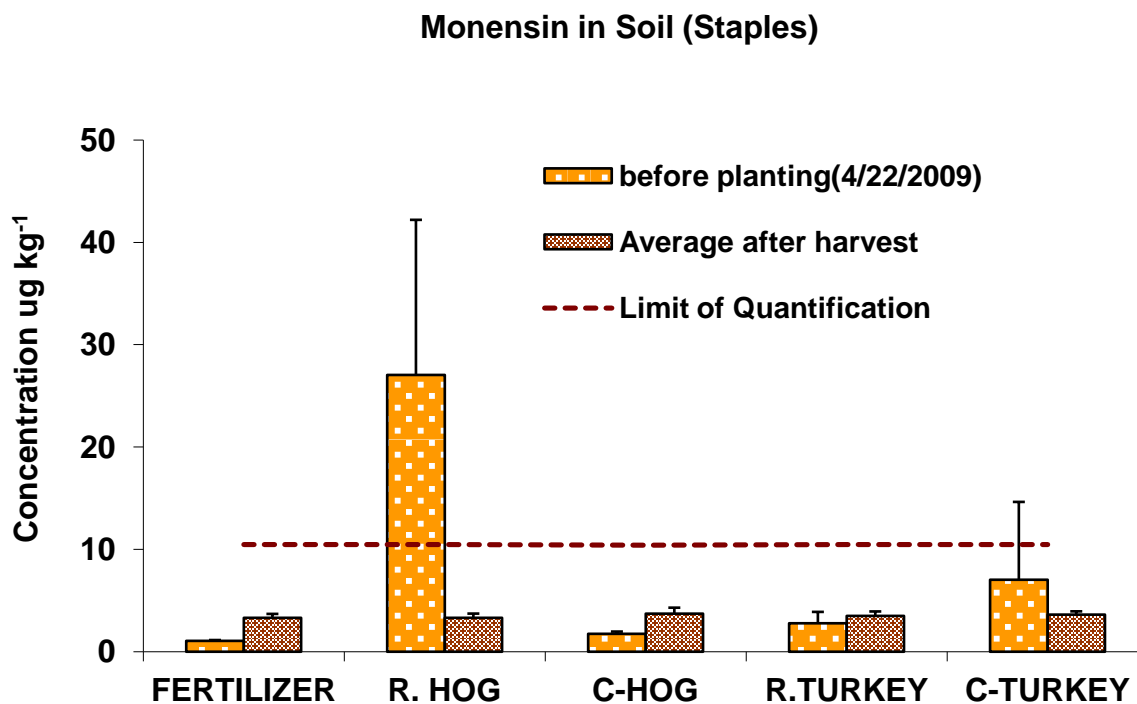


Figure 23. Monensin concentration in manure applied soils before planting and after harvest at Staples, MN. Prefix R and C refer to raw and composted hog or turkey manure.

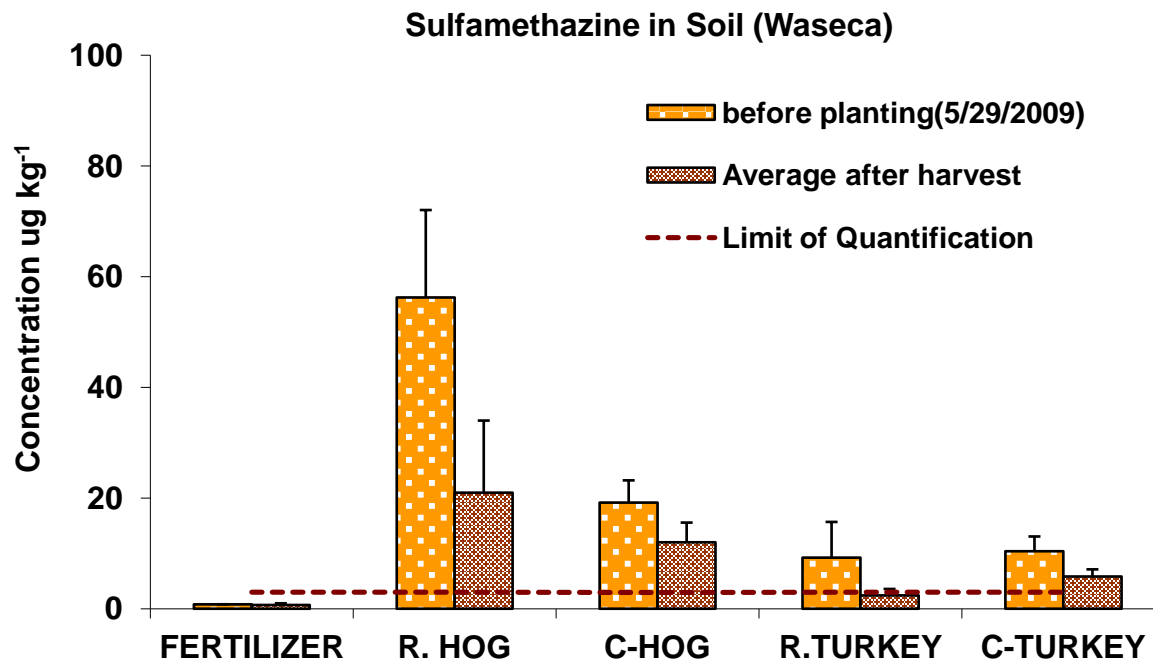


Figure 24. Sulfamethazine concentration in manure applied soils before planting and after harvest at Waseca. Prefix R and C refer to raw and composted hog or turkey manure.

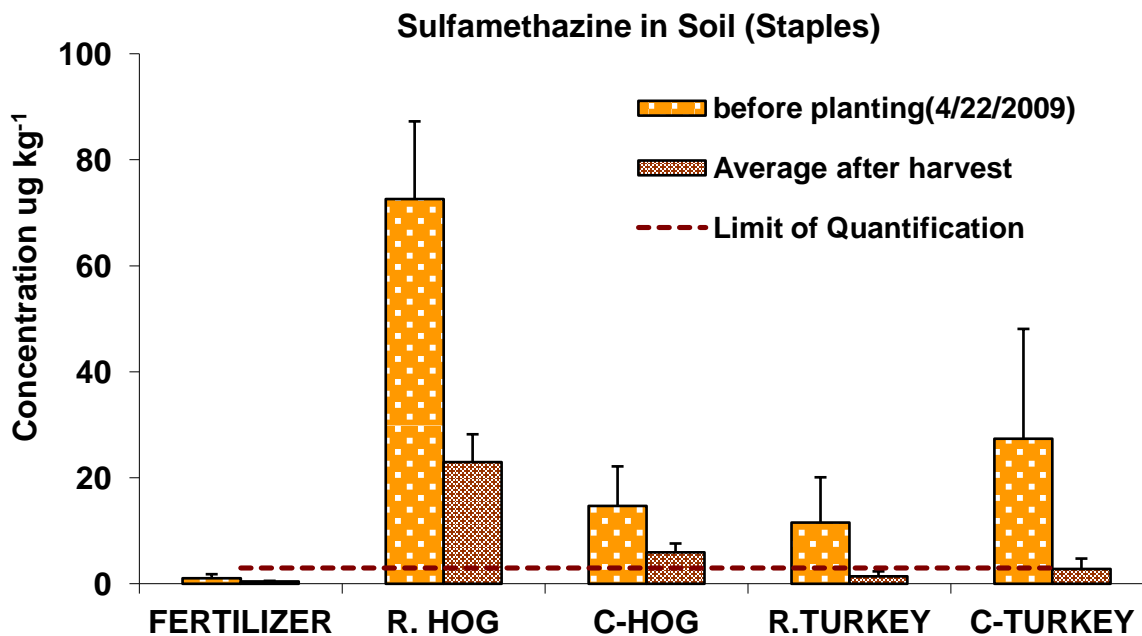


Figure 25 Sulfamethazine concentration in manure applied soils before planting and after harvest at Staples. Prefix R and C refer to raw and composted hog or turkey manure.

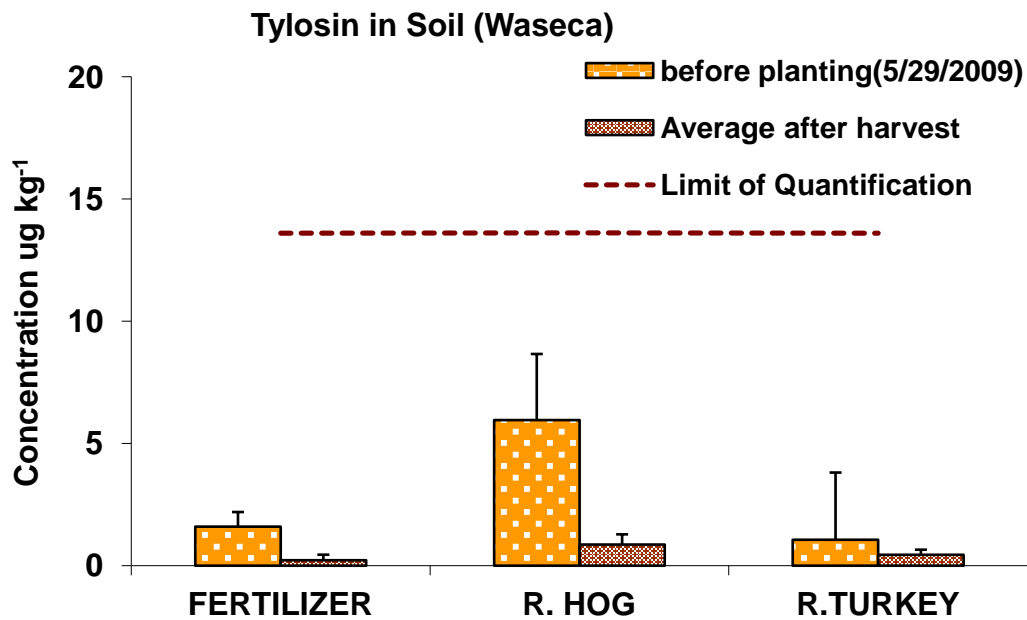


Figure 26. Tylosin concentration in manure applied soils before planting and after harvest at Waseca. Prefix R and C refer to raw and composted hog or turkey manure.

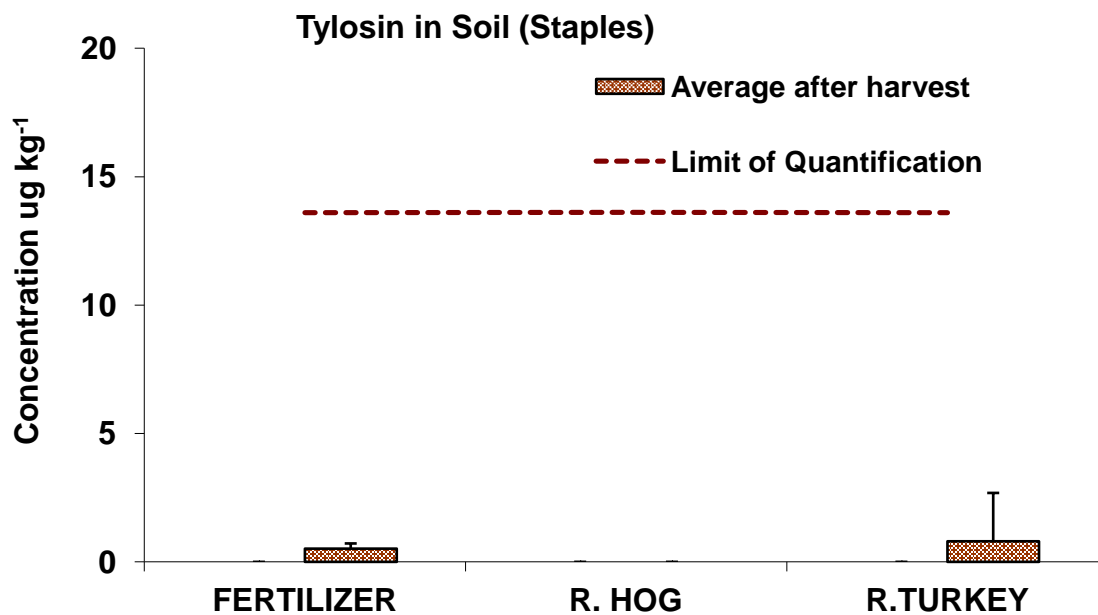


Figure 27. Tylosin concentration in manure applied soils before planting and after harvest at Staples. Prefix R refers to raw hog or turkey manure.

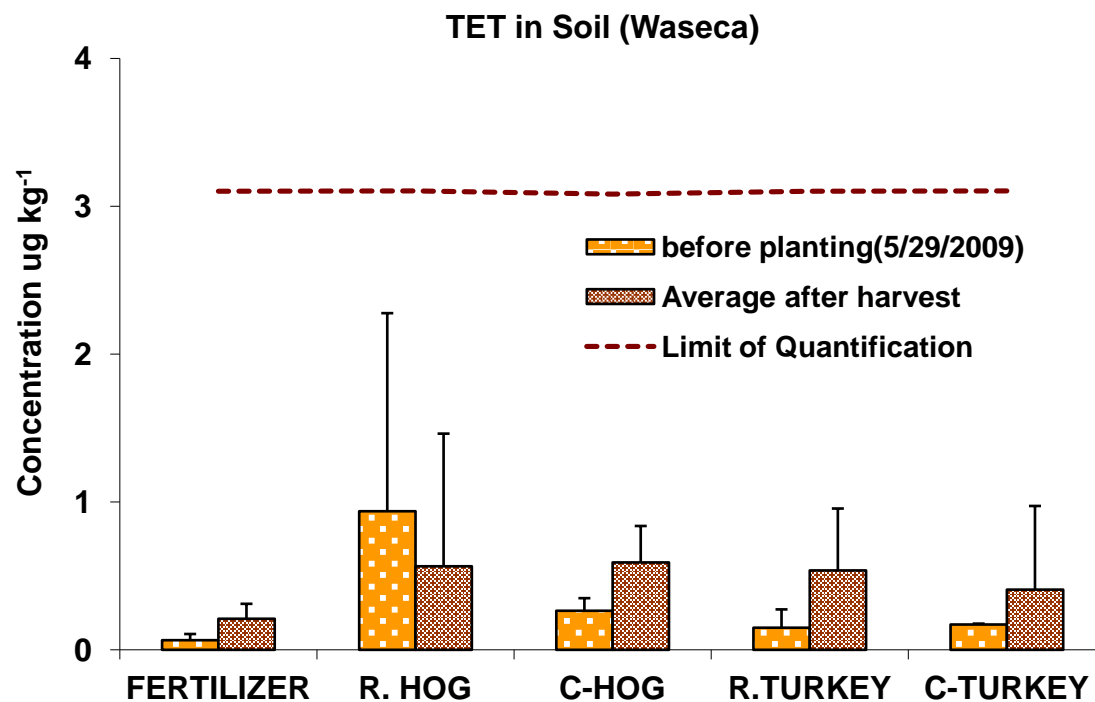


Figure 28. Chlortetracycline concentrations in manure applied soils before planting and after harvest at Waseca. Prefix R and C refer to raw and composted hog or turkey manure.

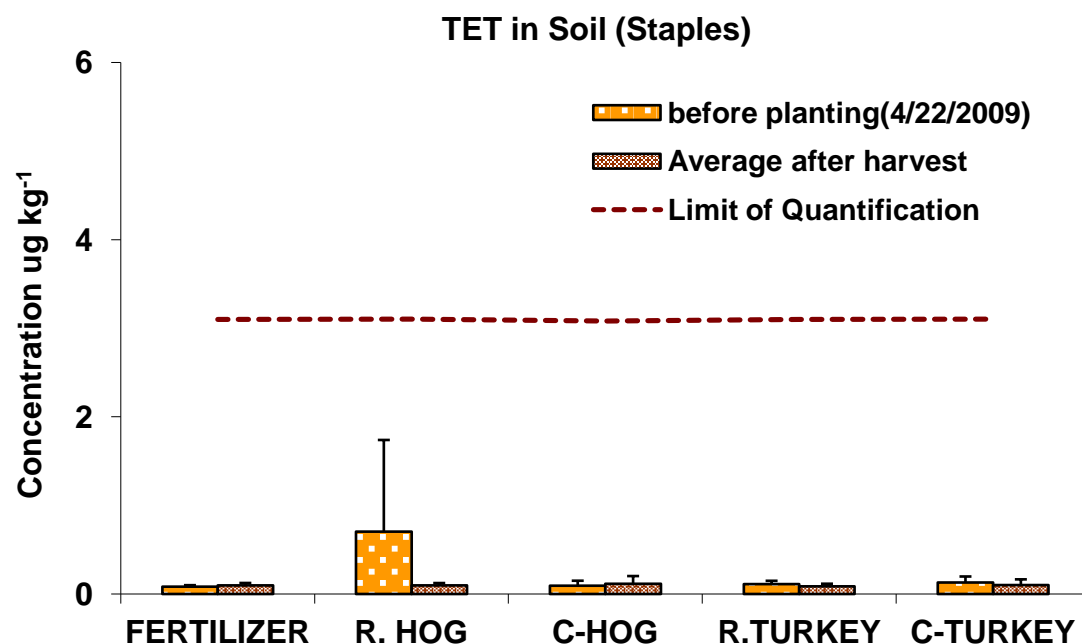


Figure 29. Chlortetracycline concentrations in manure applied soils before planting and after harvest at Staples. Prefix R and C refer to raw and composted hog or turkey manure.

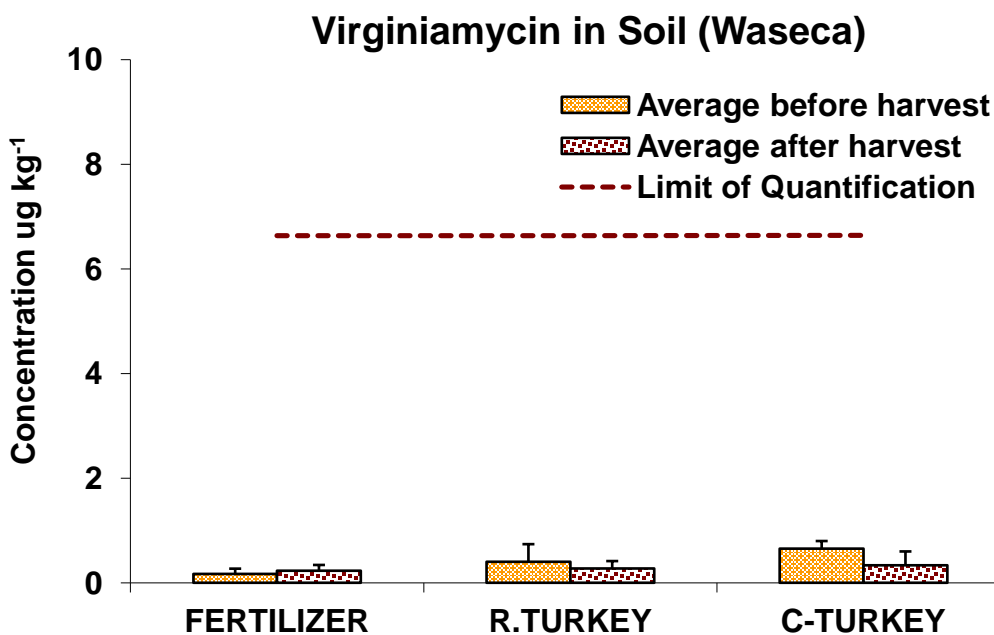


Figure 30. Virginiamycin concentration in manure applied soils before planting and after harvest at Waseca. Prefix R and C refer to raw and composted hog or turkey manure.

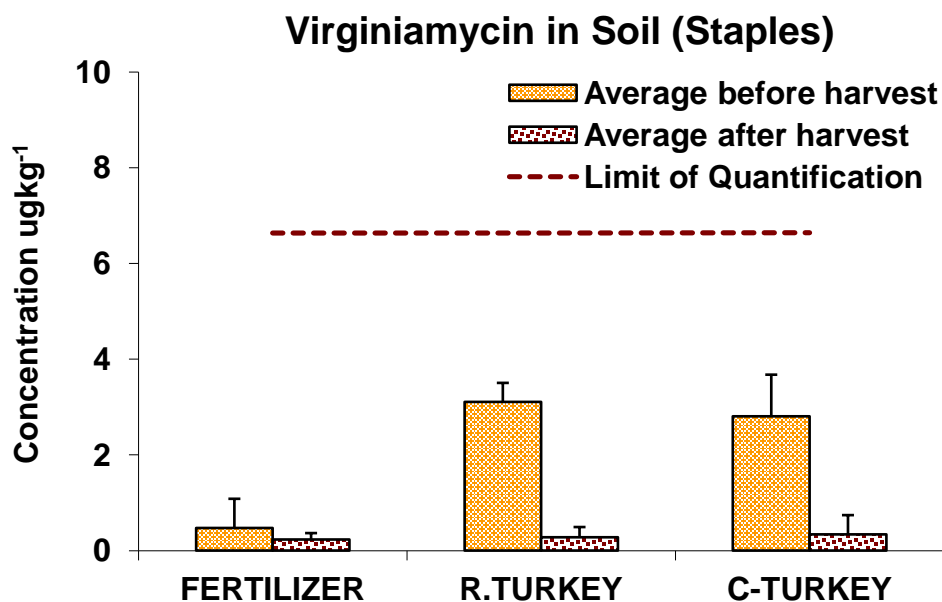


Figure 31. Virginiamycin concentration in manure applied soils before planting and after harvest at Staples. Prefix R and C refer to raw and composted hog or turkey manure.

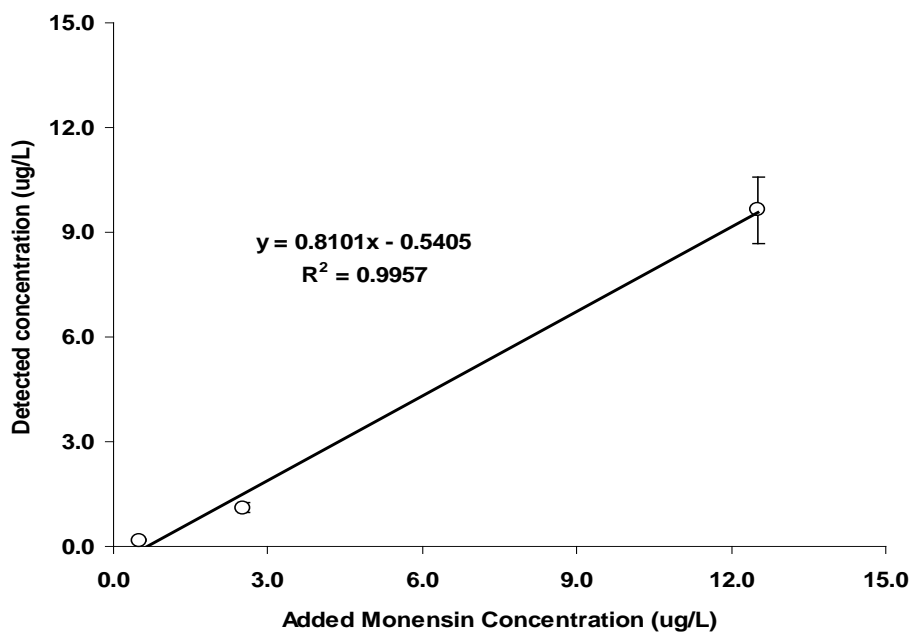


Figure 32. Relationship between added monensin into pepper extractant and measured ELISA monensin concentrations.

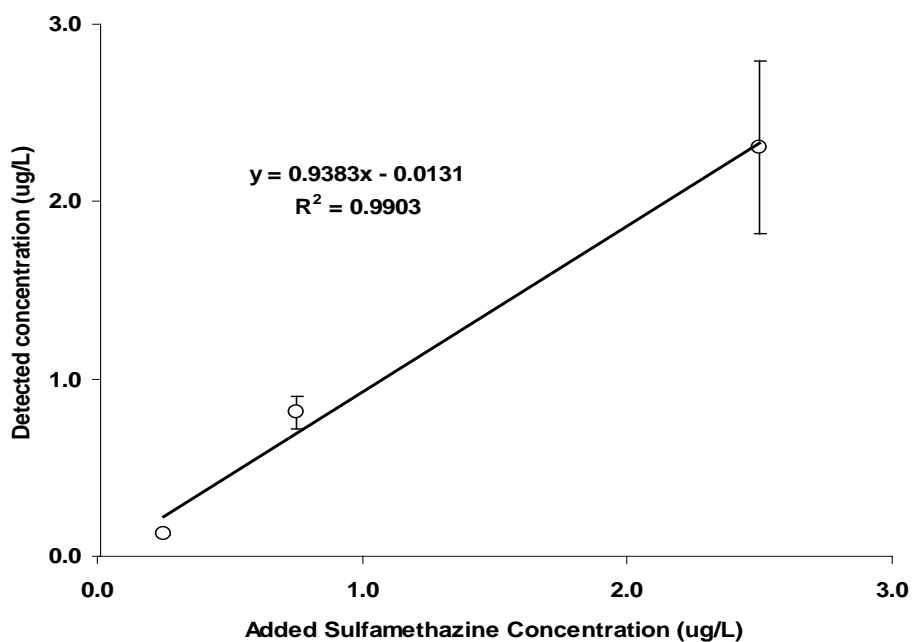


Figure 33. Relationship between added sulfamethazine into pepper extractant and measured ELISA sulfamethazine concentration.

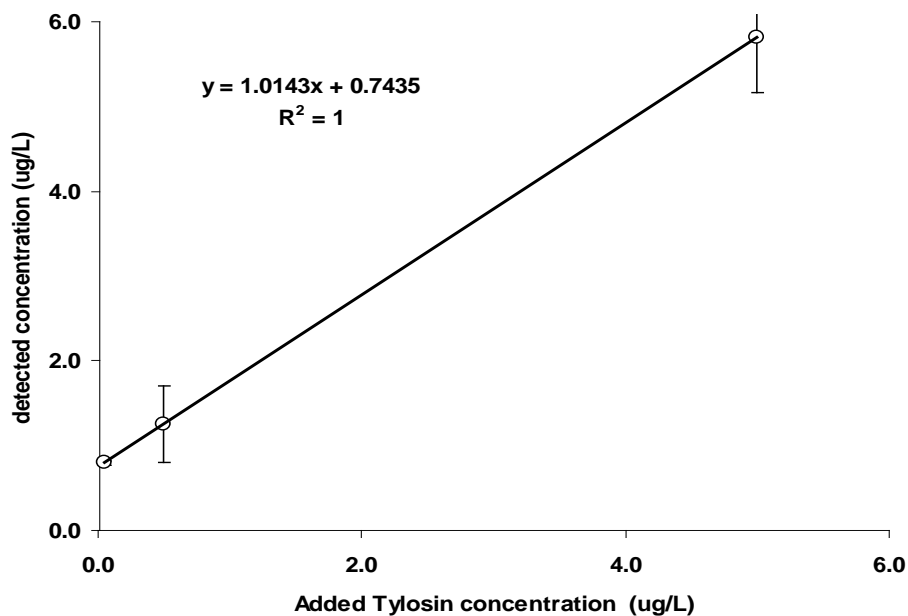


Figure 34. Relationship between added tylosin into pepper extractant and measured ELISA tylosin concentration.

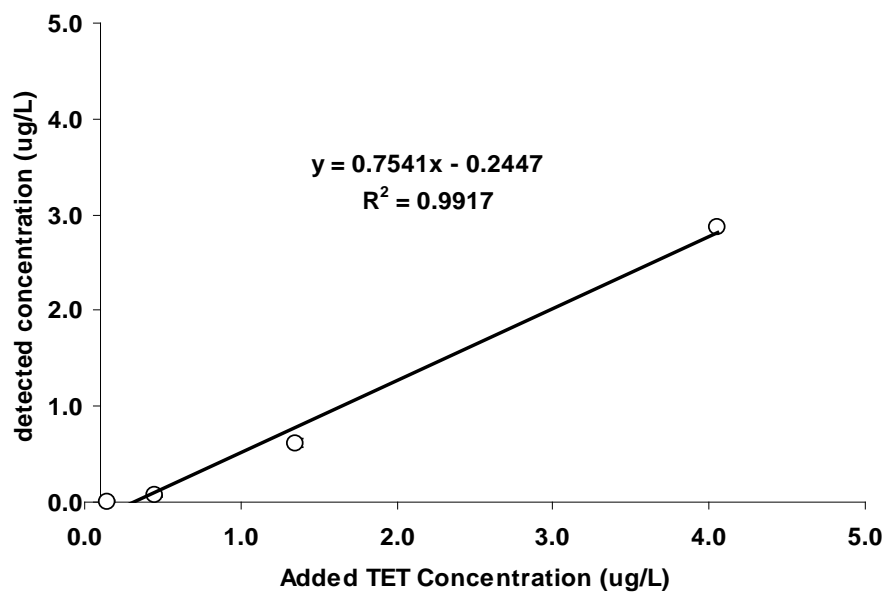


Figure 35. Relationship between added chlortetracycline into pepper extractant and measured ELISA chlortetracycline concentration.

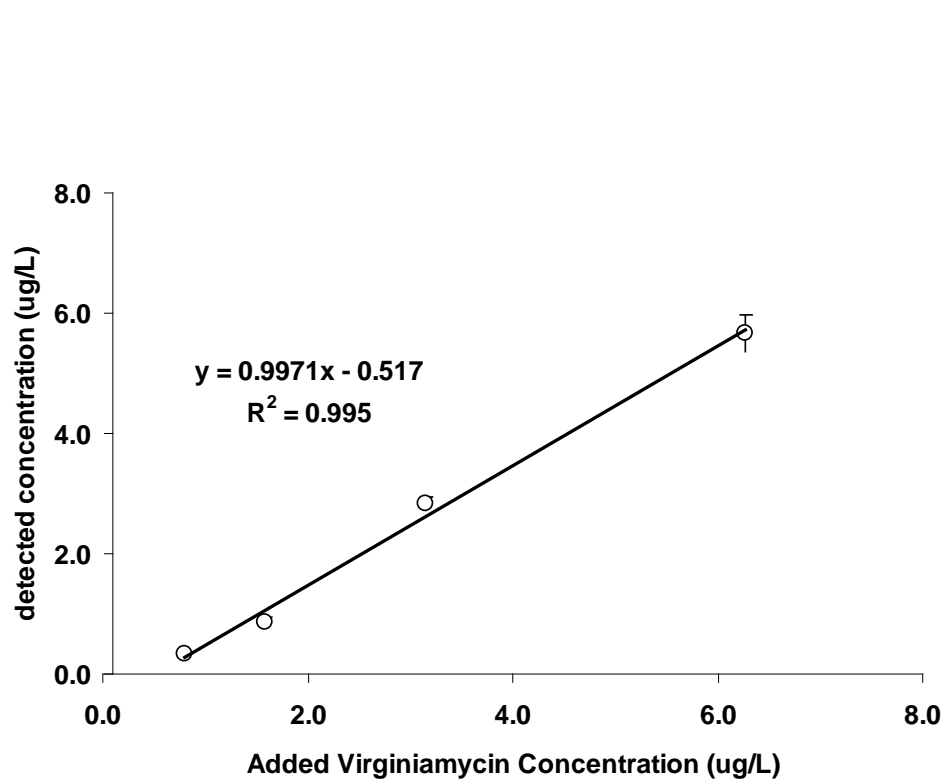


Figure 36. Relationship between added virginiamycin into pepper extractant and measured ELISA virginiamycin concentration.

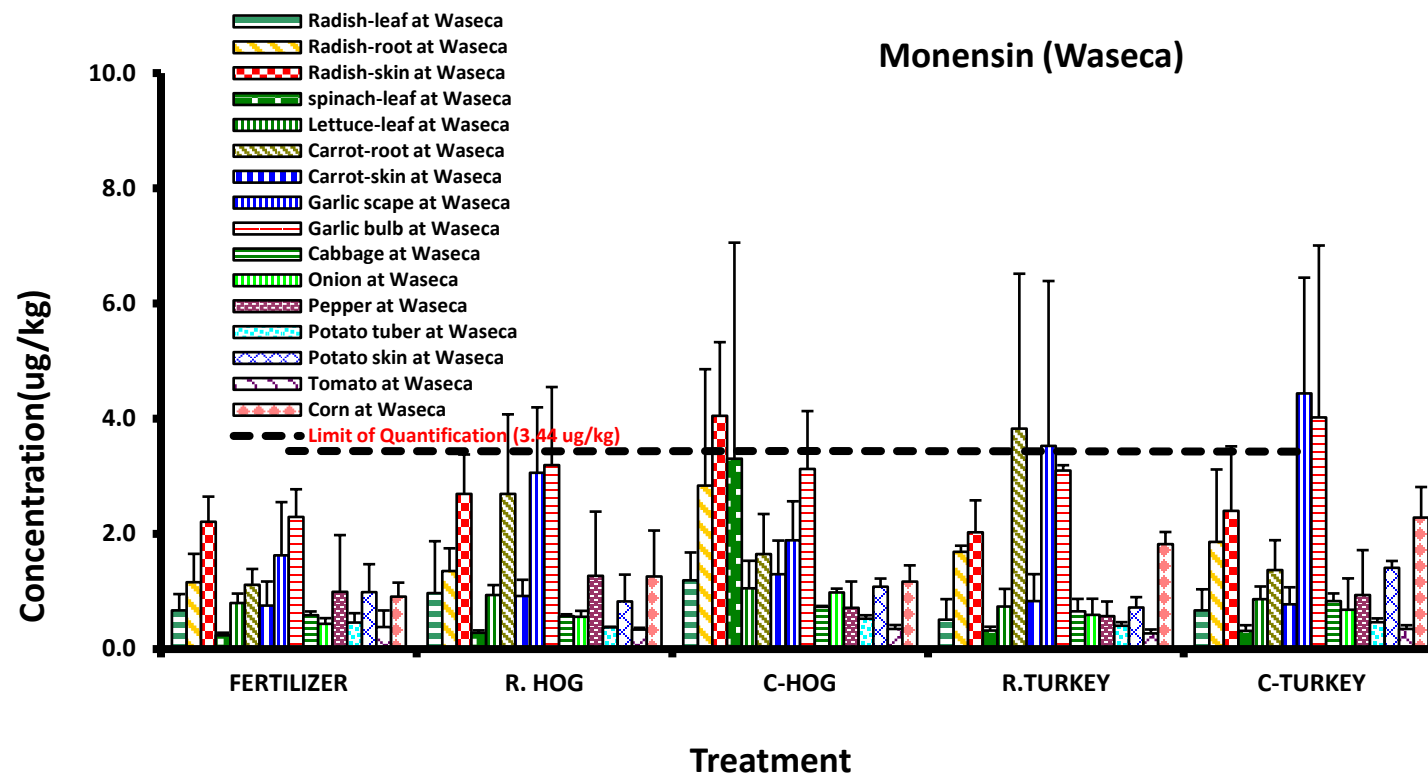


Figure 37. Concentration of monensin in plant tissue of different vegetable crops for various nutrient source treatments at Waseca, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

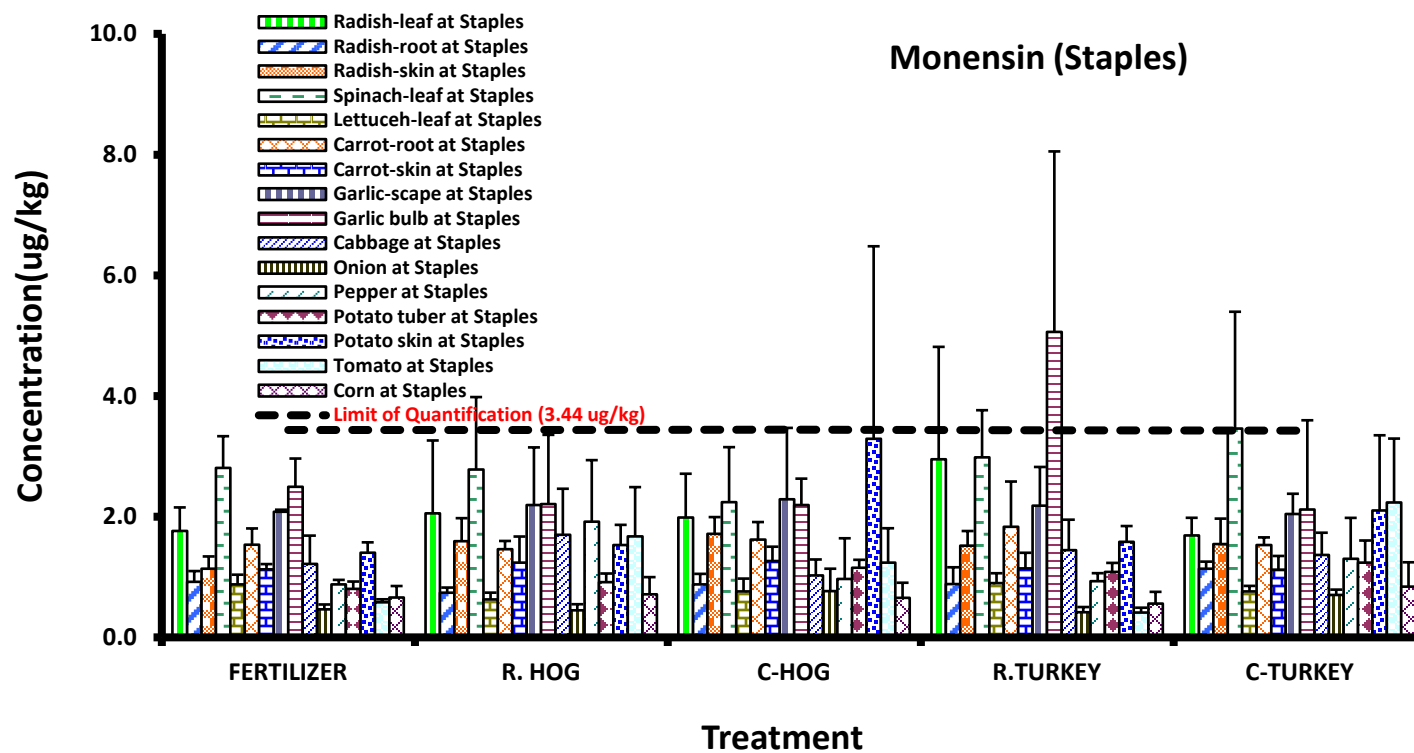


Figure 38. Concentration of monensin in plant tissue of different vegetable crops for various nutrient source treatments at Staples, MN. Error bar indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

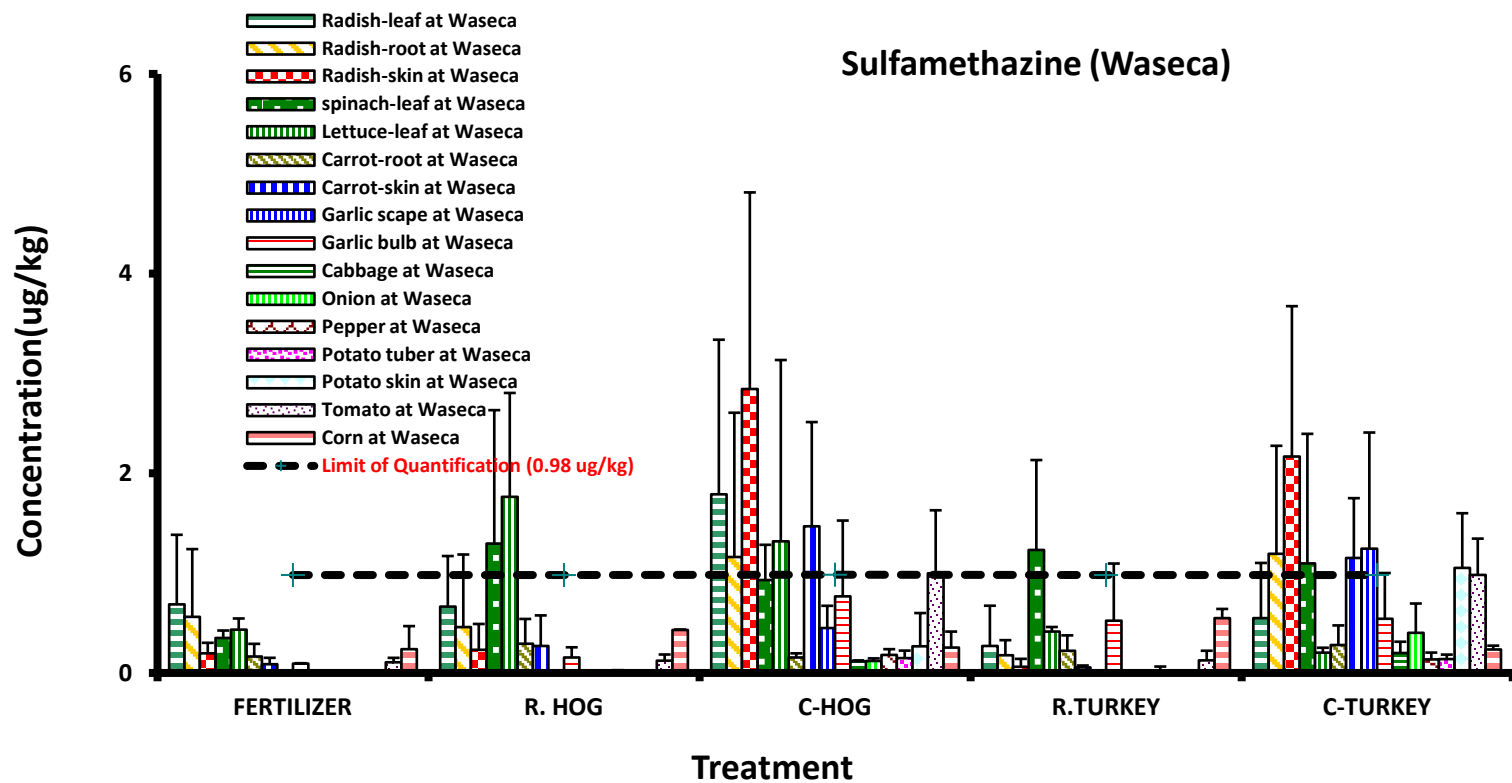


Figure 39. Concentration of sulfamethazine in plant tissue of different vegetable crops for various nutrient source treatments at Waseca, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

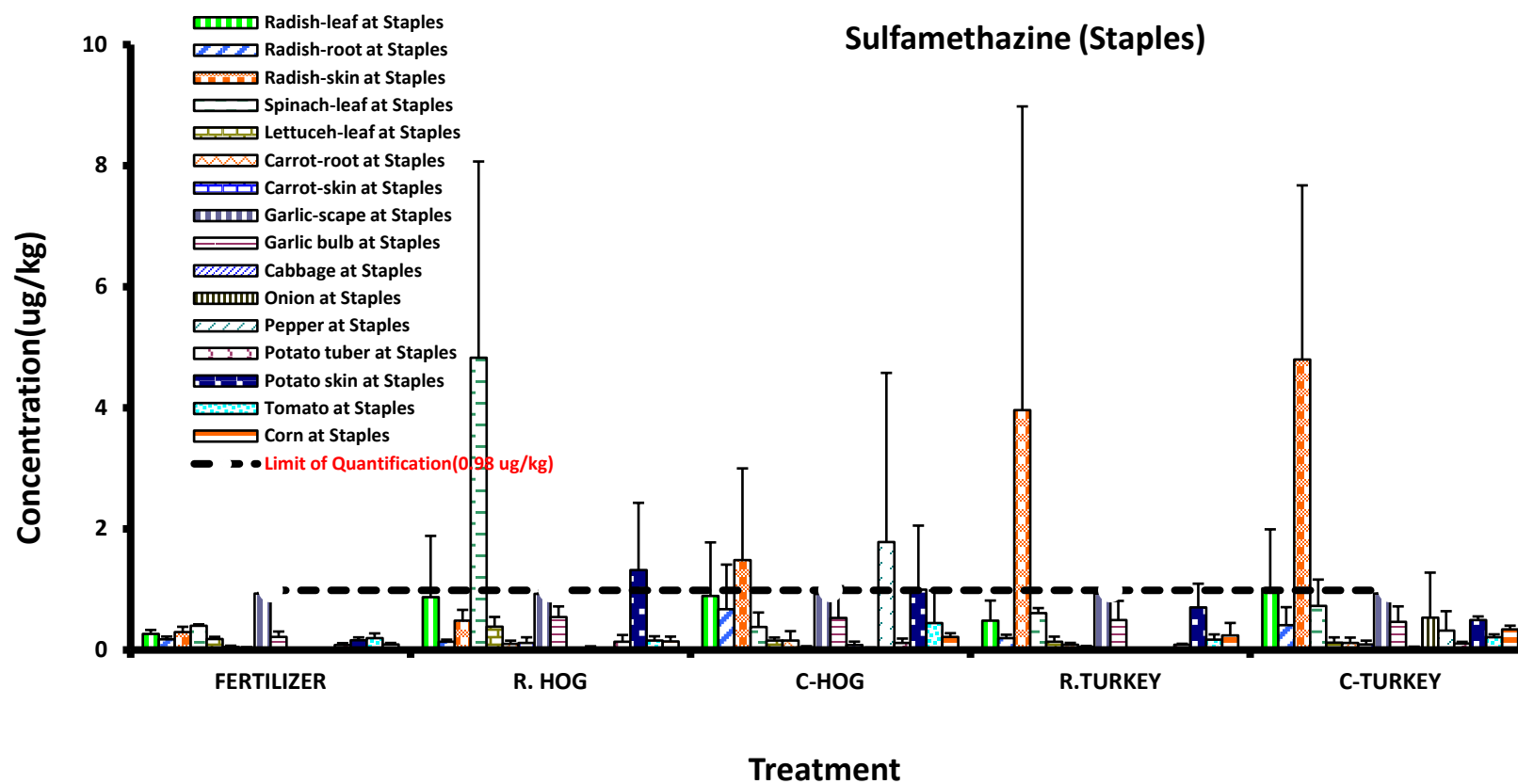


Figure 40. Concentration of sulfamethazine in plant tissue of different vegetable crops for various nutrient source treatments at Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

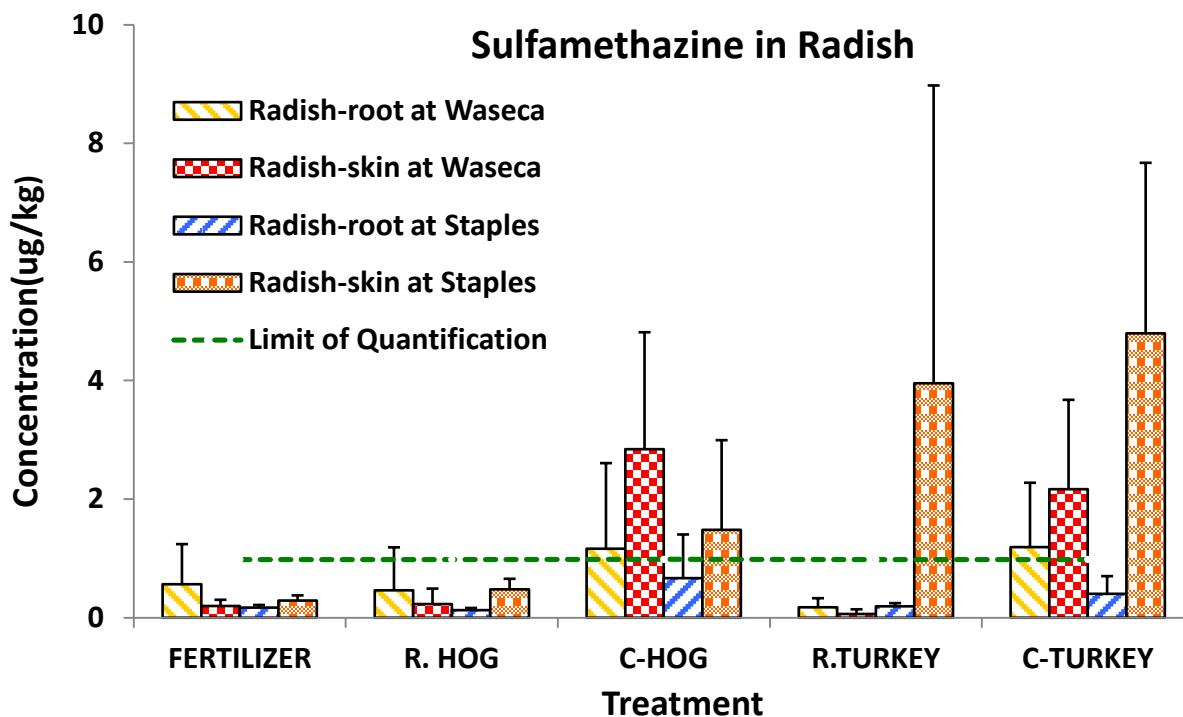


Figure 41. Concentration of sulfamethazine in radish skin and root for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

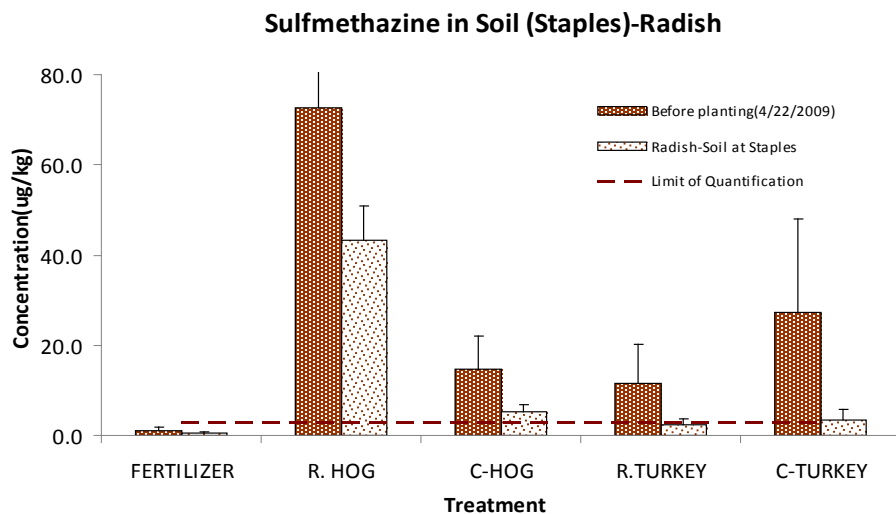


Figure 42. Sulfamethazine concentration in soils from radish plots before planting and at harvest for various nutrient source treatments at Staples. Prefix R and C refer to raw and composted hog or turkey manure.

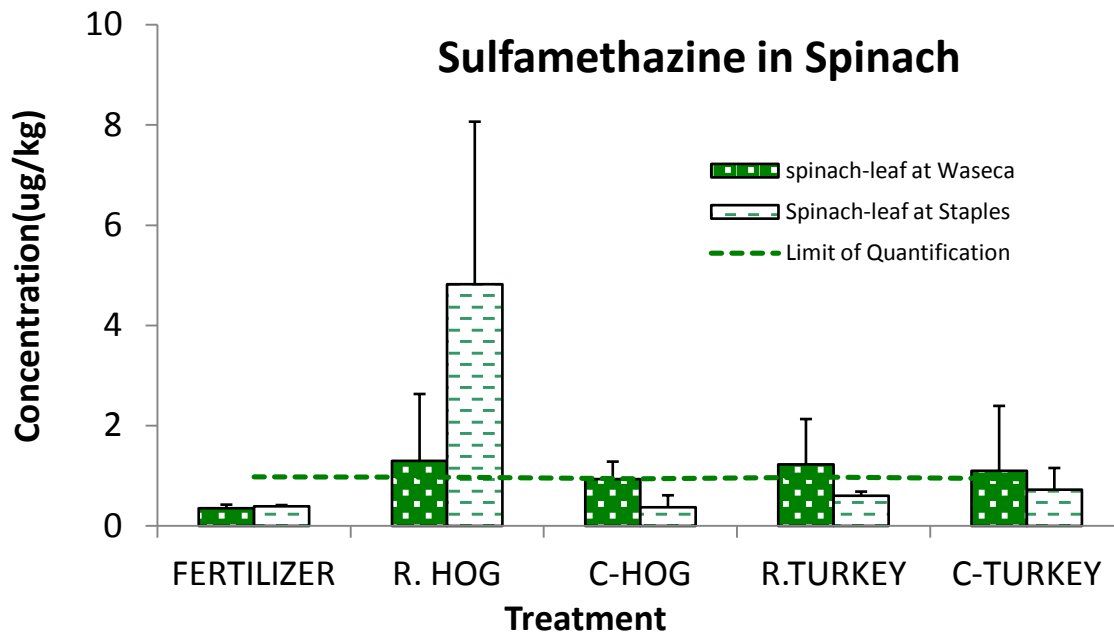


Figure 43. Concentration of sulfamethazine in spinach for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

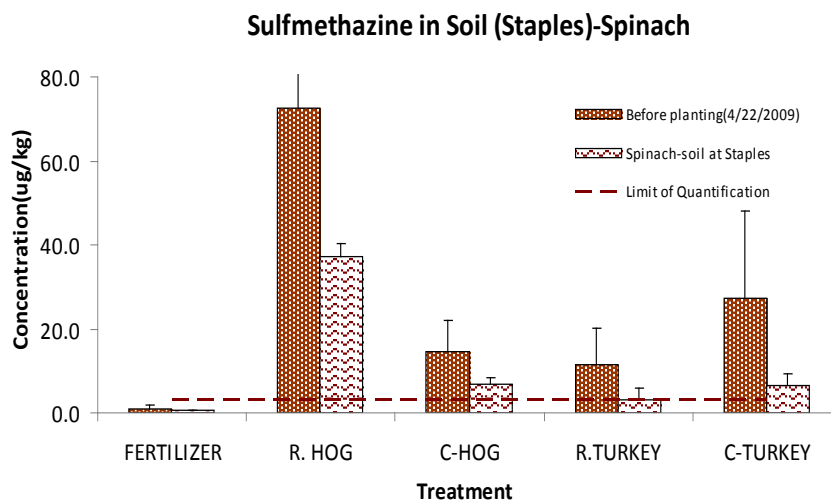


Figure 44. Sulfamethazine concentration in soils from spinach plots before planting and at harvest for various nutrient source treatments at Staples. Prefix R and C refer to raw and composted hog or turkey manure.

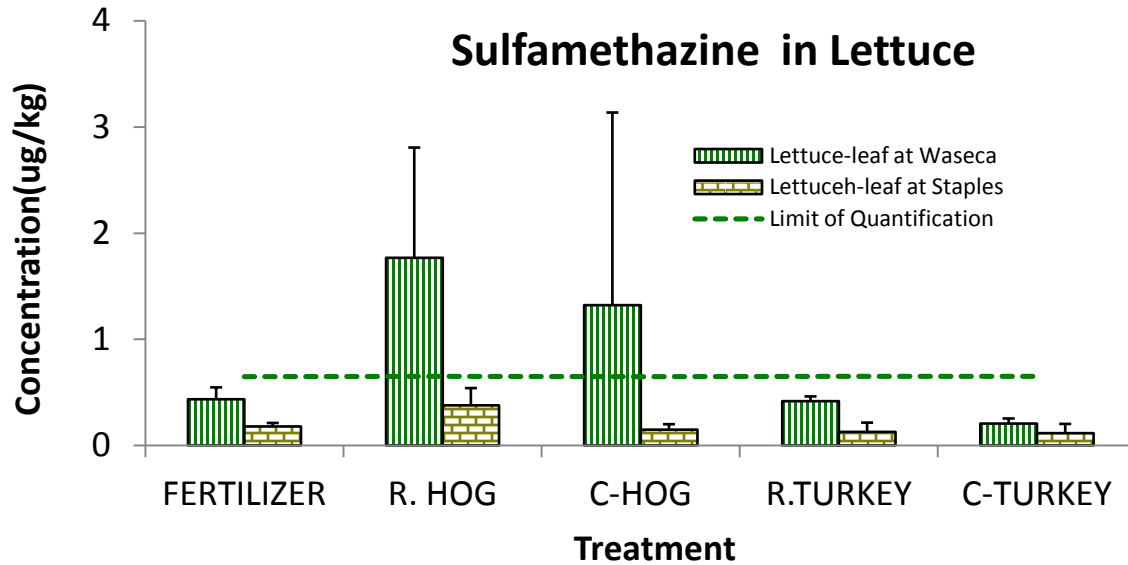


Figure 45. Concentration of sulfamethazine in lettuce for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

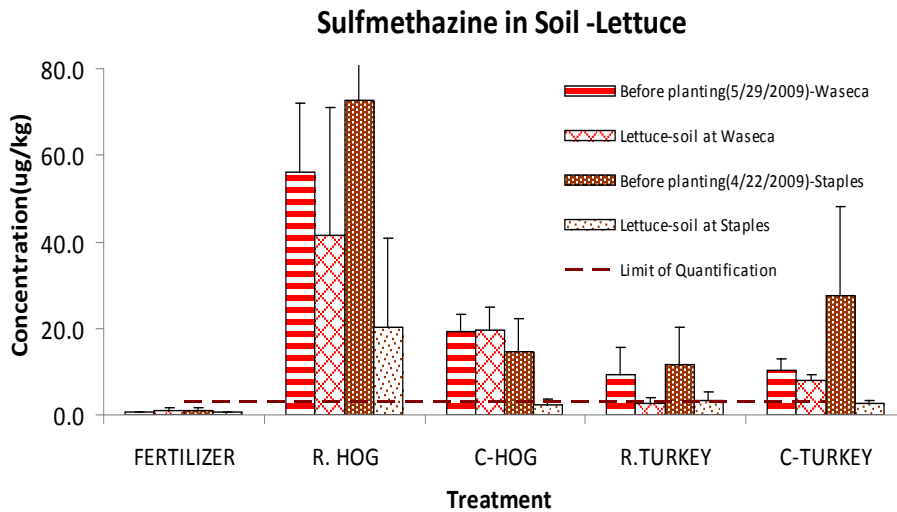


Figure 46. Sulfamethazine concentration in soils from lettuce plots before planting and at harvest for various nutrient source treatments at both Waseca and Staples. Prefix R and C refer to raw and composted hog or turkey manure.

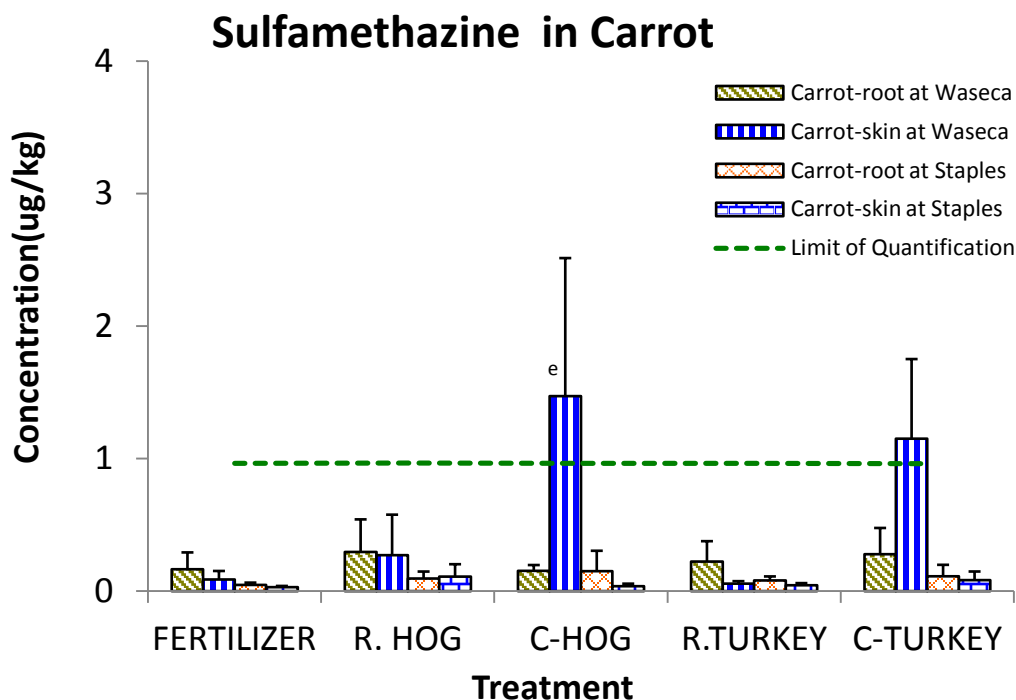


Figure 47. Concentration of sulfamethazine in carrot for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

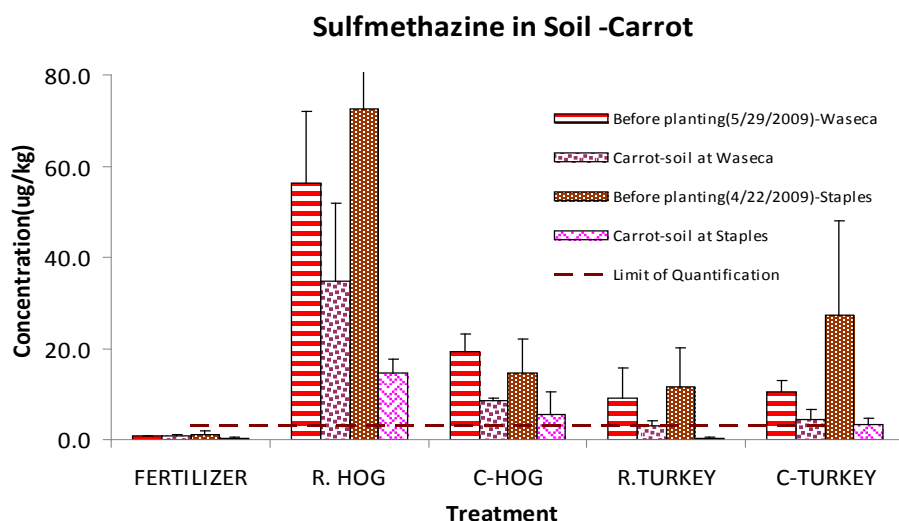


Figure 48. Sulfamethazine concentration in soils from carrot plots before planting and at harvest for various nutrient source treatments at both Waseca and Staples. Prefix R and C refer to raw and composted hog or turkey manure.

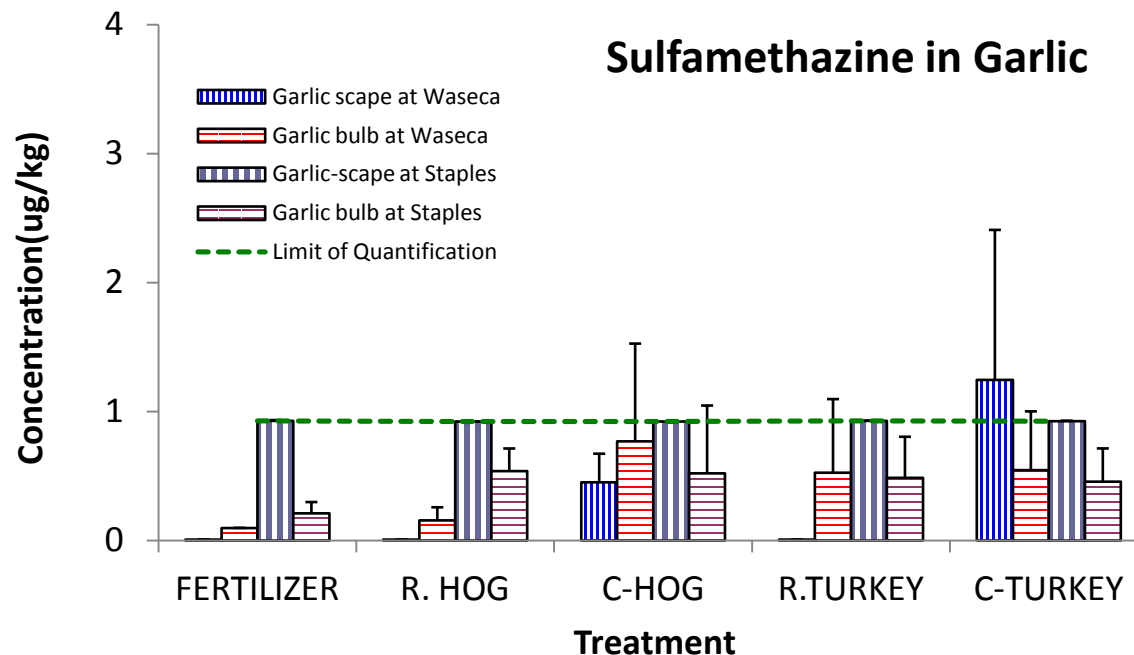


Figure 49. Concentration of sulfamethazine in garlic for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

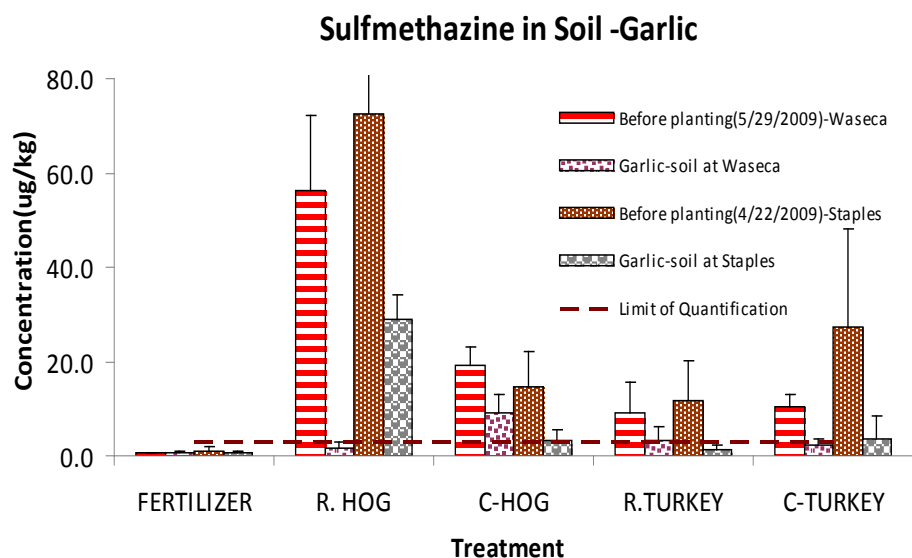


Figure 50. Sulfamethazine concentration in soils from garlic plots before planting and at harvest for various nutrient source treatments at both Waseca and Staples. Prefix R and C refer to raw and composted hog or turkey manure.

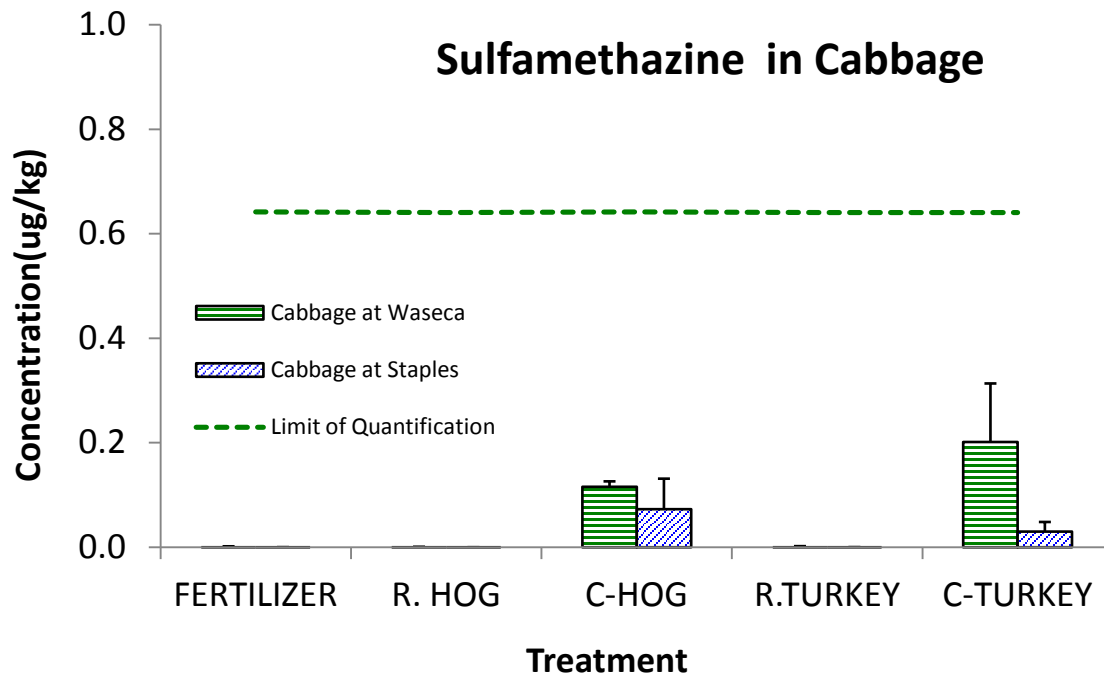


Figure 51. Concentration of sulfamethazine in cabbage for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

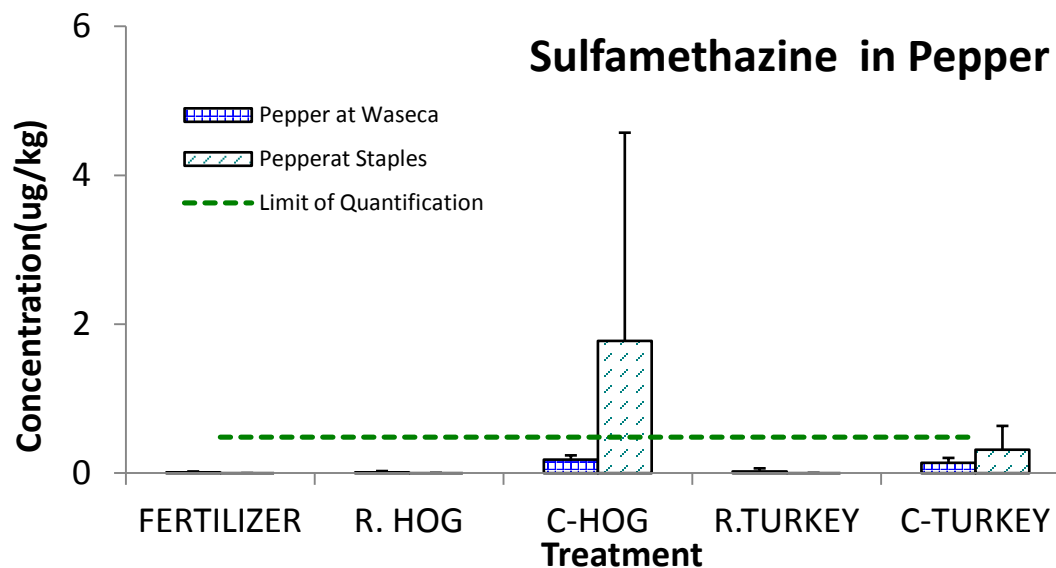


Figure 52. Concentration of sulfamethazine in pepper for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

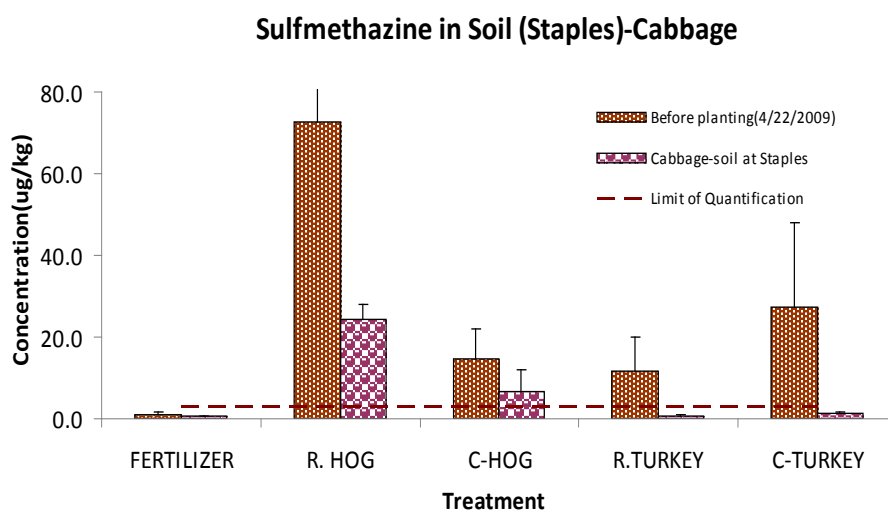


Figure 53. Sulfamethazine concentration in soils from cabbage plots before planting and at harvest for various nutrient source treatments at Staples. Prefix R and C refer to raw and composted hog or turkey manure.

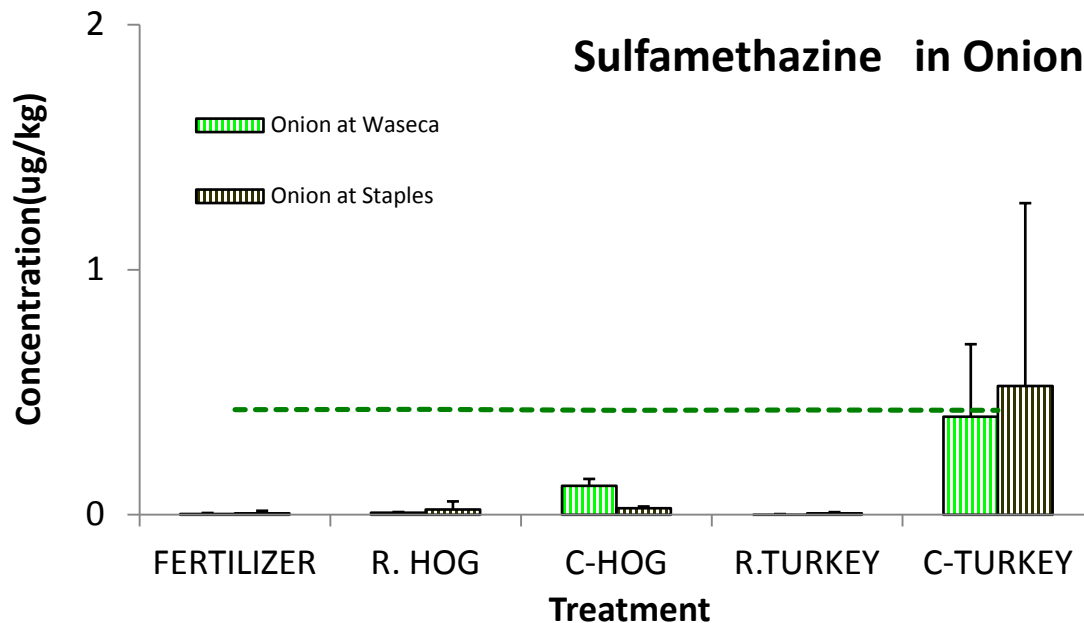


Figure 54. Concentration of sulfamethazine in onions for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

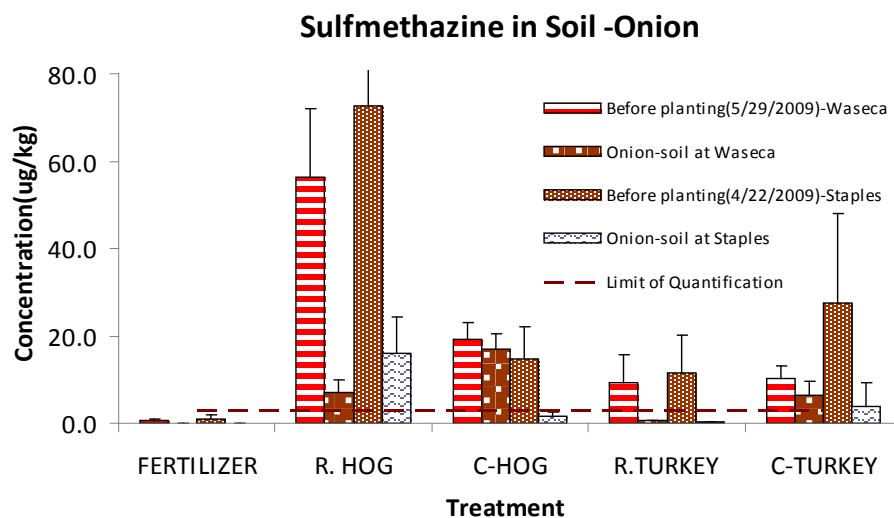


Figure 55. Sulfamethazine concentration in soils from the onion plots before planting and at harvest at Waseca and Staples for various nutrient source treatments. Prefix R and C refer to raw and composted hog or turkey manure.

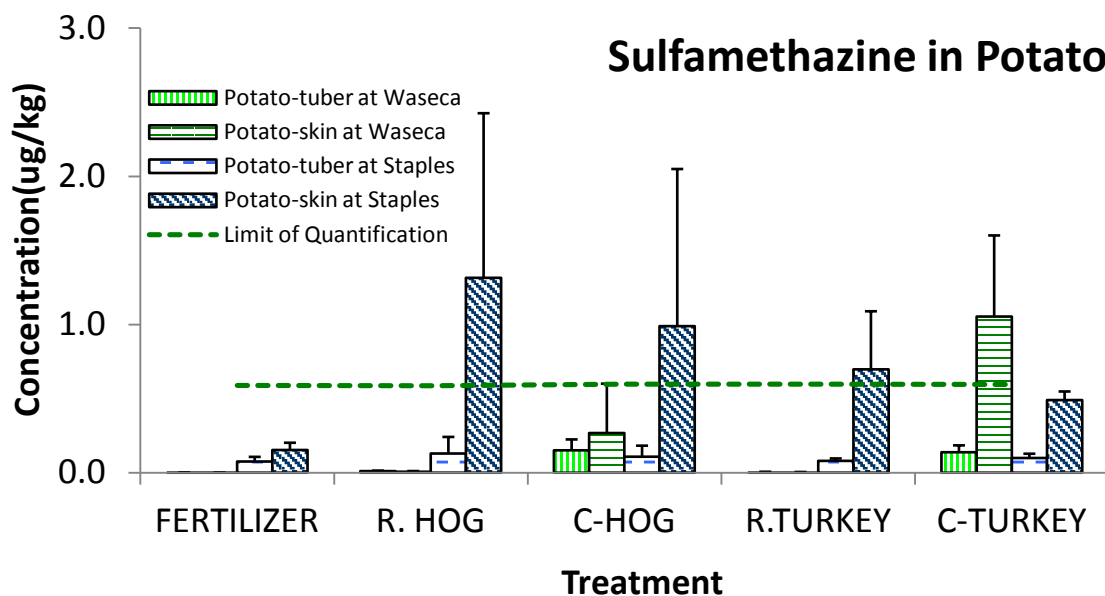


Figure 56. Concentration of sulfamethazine in potatoes for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

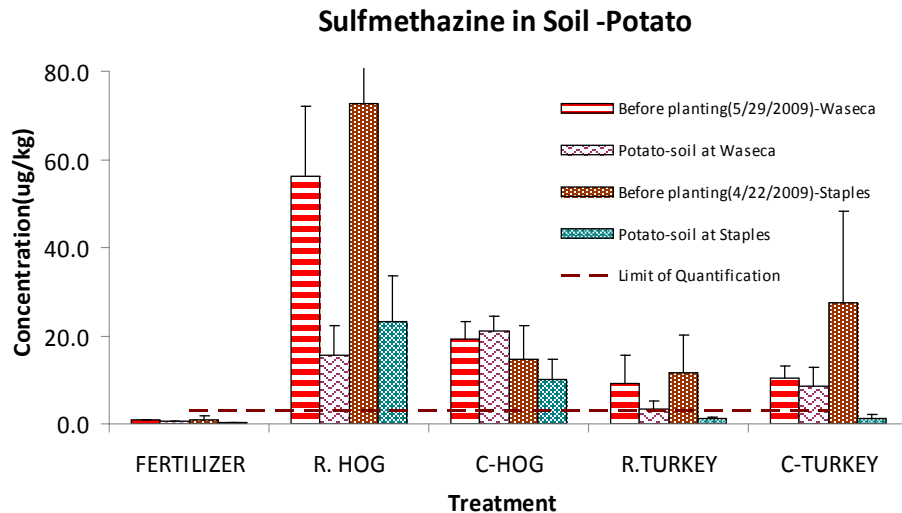


Figure 57. Sulfamethazine concentration in soils from potato plots before planting and at harvest for various nutrient source treatments at Waseca and Staples. Prefix R and C refer to raw and composted hog or turkey manure.

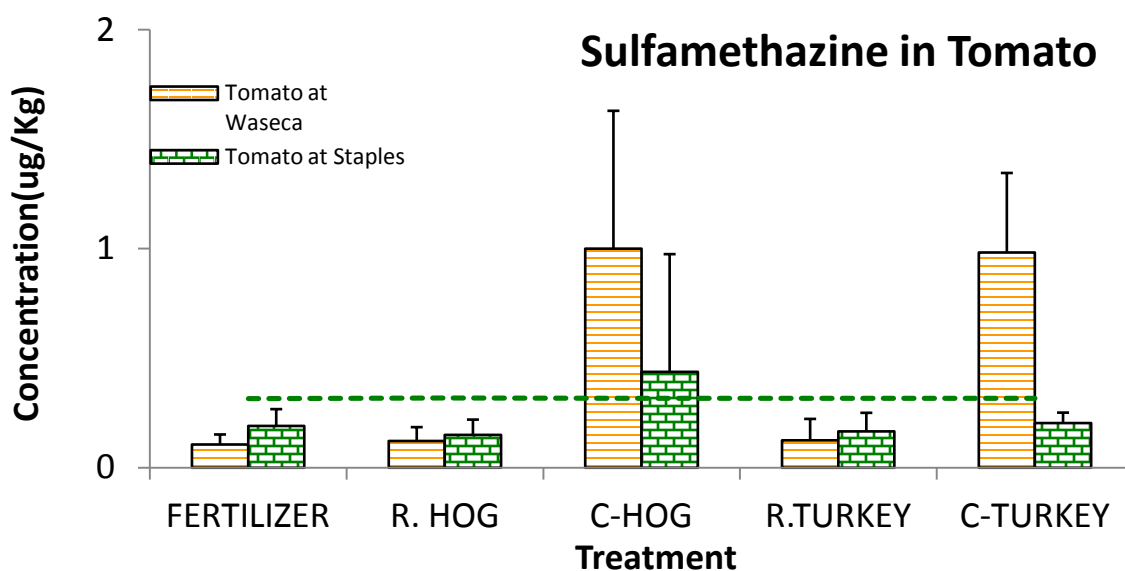


Figure 58. Concentration of sulfamethazine in tomatoes for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

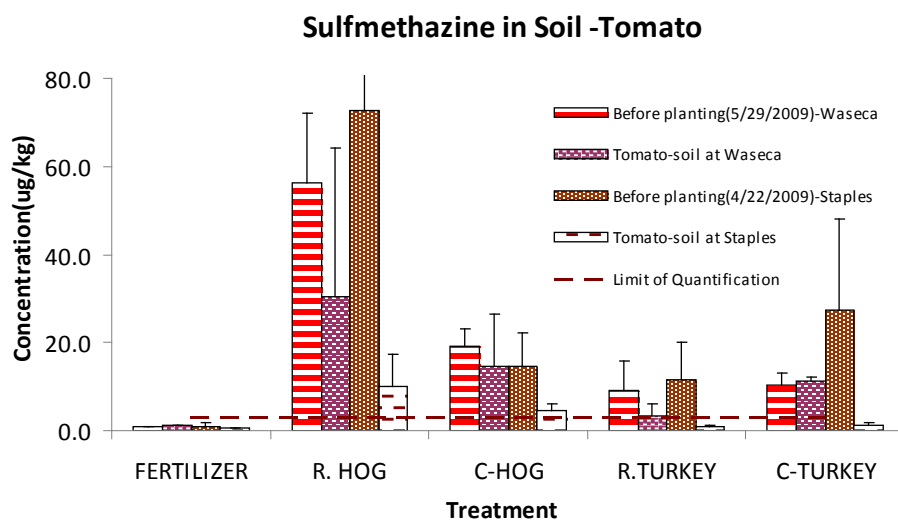


Figure 59. Sulfamethazine concentration in soils from tomato plots before planting and at harvest for various nutrient source treatments at Waseca and Staples. Prefix R and C refer to raw and composted hog or turkey manure.

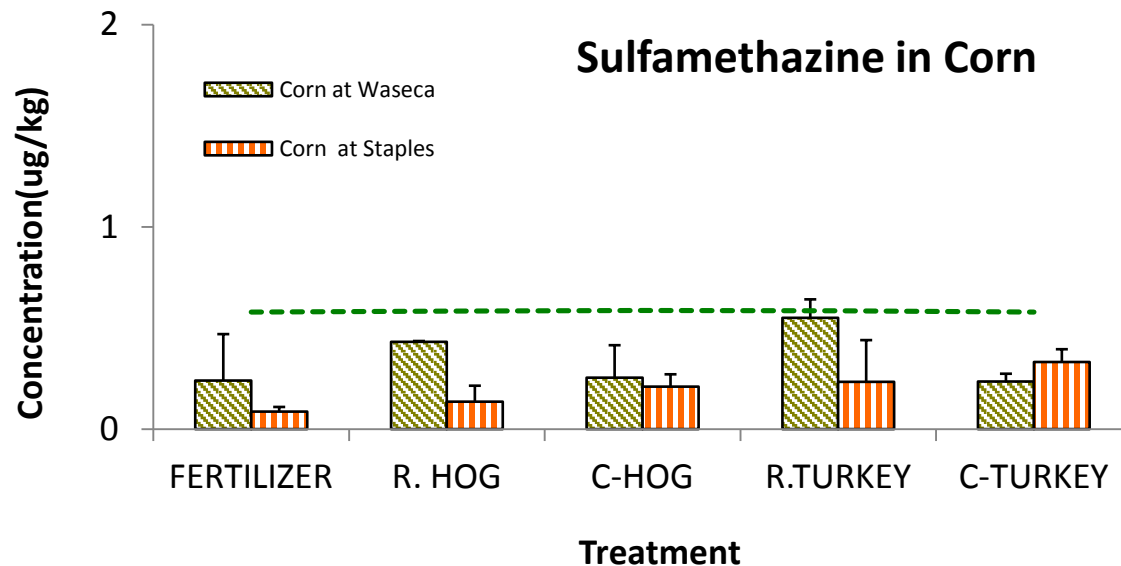


Figure 60. Concentration of sulfamethazine in sweet corn for various nutrient source treatments at Waseca and Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

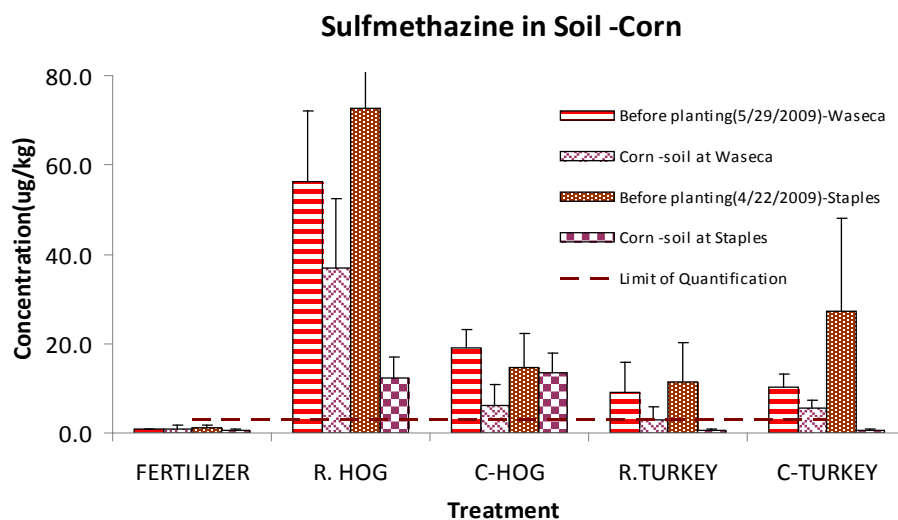


Figure 61. Sulfamethazine concentration in soil from sweet corn plots before planting and at harvest for various nutrient source treatments at Waseca and Staples. Prefix R and C refer to raw and composted hog or turkey manure.

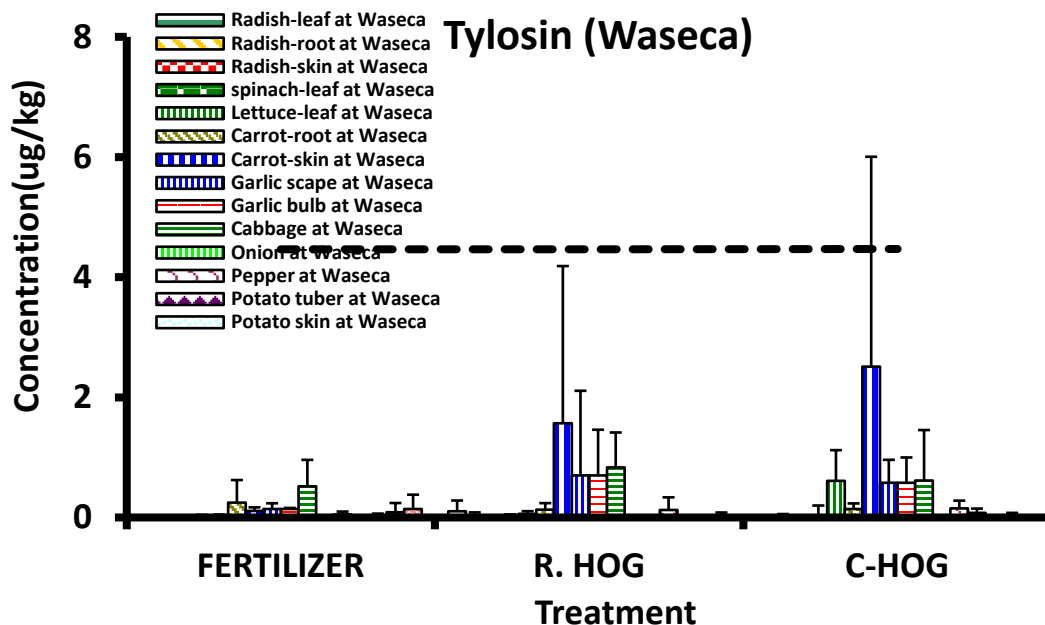


Figure 62. Concentration of tylosin in plant tissue of different vegetable crops for various nutrient source treatments at Waseca, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

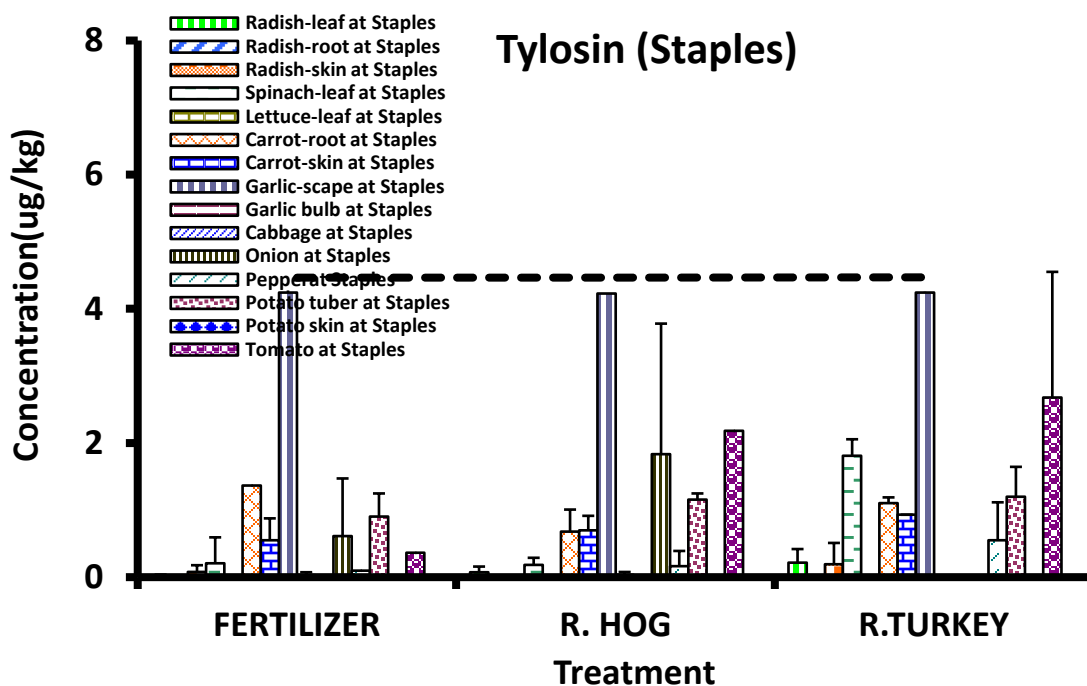


Figure 63. Concentration of tylosin in plant tissue of different vegetable crops for various nutrient source treatments at Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

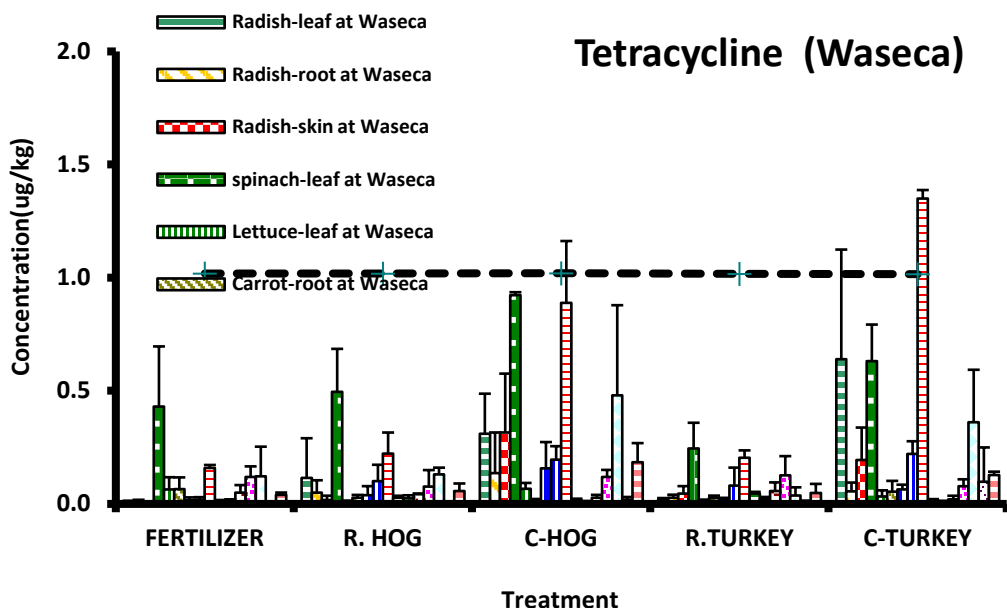


Figure 64. Concentration of tetracycline in plant tissue of different vegetable crops for various nutrient source treatments at Waseca, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

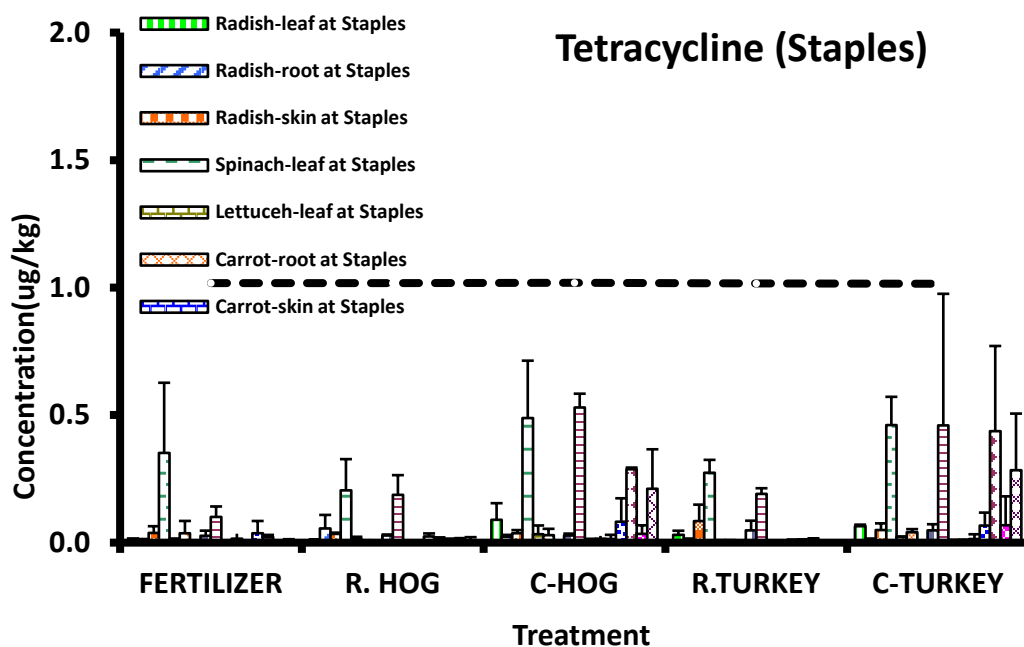


Figure 65. Concentration of tetracycline in plant tissue of different vegetable crops for various nutrient source treatments at Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

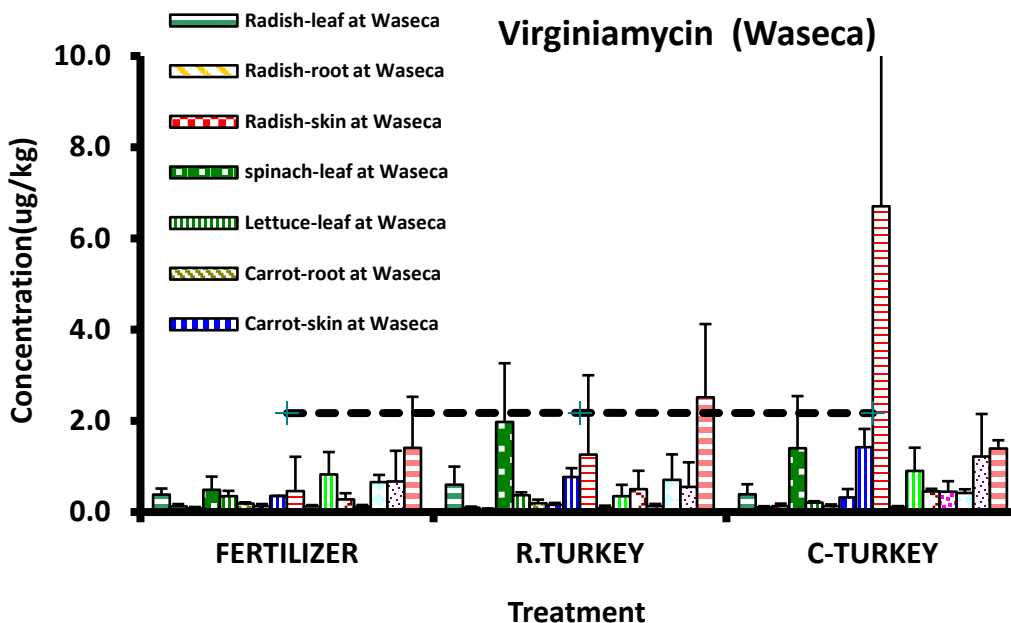


Figure 66. Concentration of virginiamycin in plant tissue of different vegetable crops for various nutrient source treatments at Waseca, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

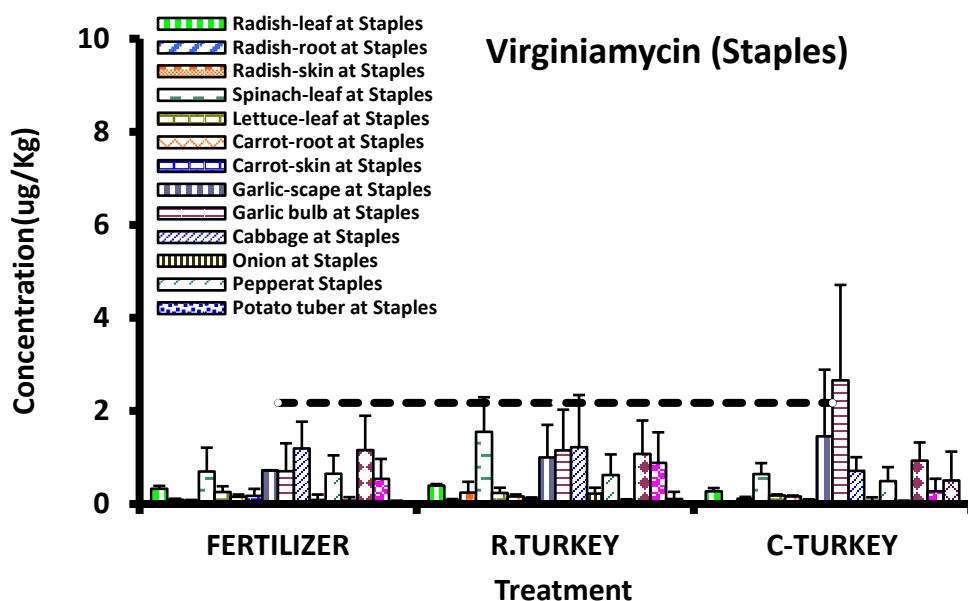


Figure 67. Concentration of virginiamycin in plant tissue of different vegetable crops for various nutrient source treatments at Staples, MN. Error bars indicate standard deviation of the mean. Prefix R and C refer to raw and composted hog or turkey manure.

Table 18: Characteristics of various farms that participated in the antibiotic uptake by vegetables survey.

Farm	Certified	Description
A	Yes	15 acres with about 400 chickens. Used poultry manure in vegetable crops
B	Yes	30 acres, used legume (hairy vetch) and composted dairy manure
C	Yes	200 acres, horse and sheep manure
D	No	Turkey manure
E	Yes	Composted cow/calf manure mixed with organic hay and straw from cow yard
F	Yes	3 acres, composted pig and cattle manure added to soil and potting mix
G	No	Dairy manure, Occasionally used Rumensen (monensin), Aureomycin (CTC), Penicillin, and excenel (ceftiofur)

Table 19. Antibiotic concentrations in manure from various certified organic and non-organic grower's farms in our survey.

	FARMS				
Antibiotics	A¹	B²	D³	E⁴	G⁵
	-----Antibiotic concentration, $\mu\text{g kg}^{-1}$ -----				
Monensin	8.6	14.6	411.8	5.4	20.8
Sulfamethazine	11.3	27.8	7.8	7.1	6.0
Virginiamycin	15.6	14.5	49.9	12.1	19.2
Tetracycline	8.6	0.5	2.9	0.5	4.3

¹ Manure was only applied to corn

² Dairy manure compost & Legume (hairy vetch)

³ Turkey manure

⁴ Composted cow manure

⁵ Dairy manure

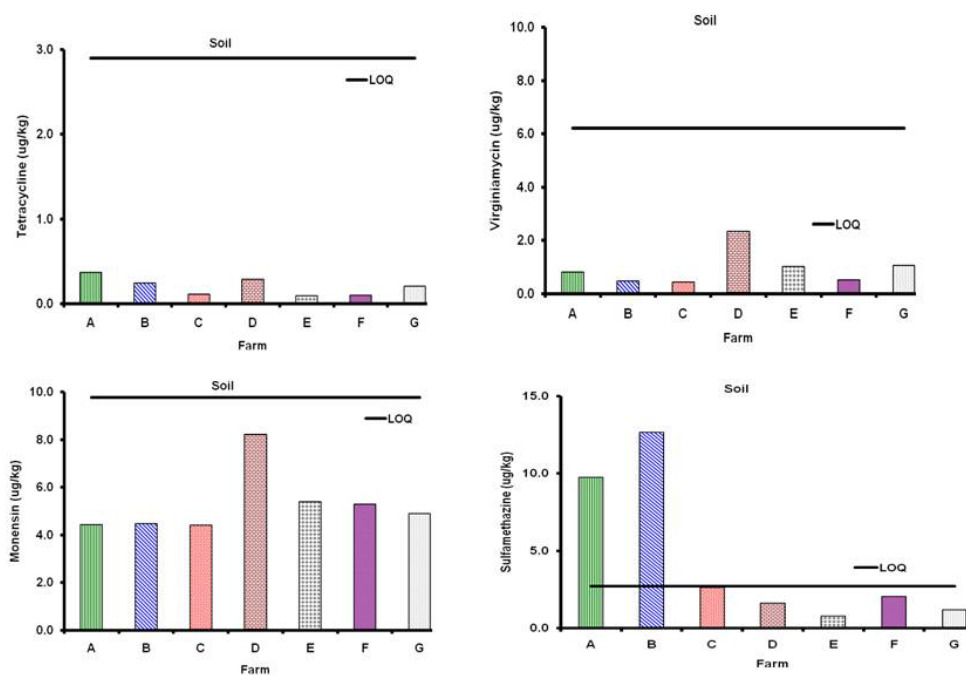


Figure 68. Antibiotic concentrations in soil collected from organic and conventional vegetable growers participating in our survey of antibiotic uptake by vegetables crops.

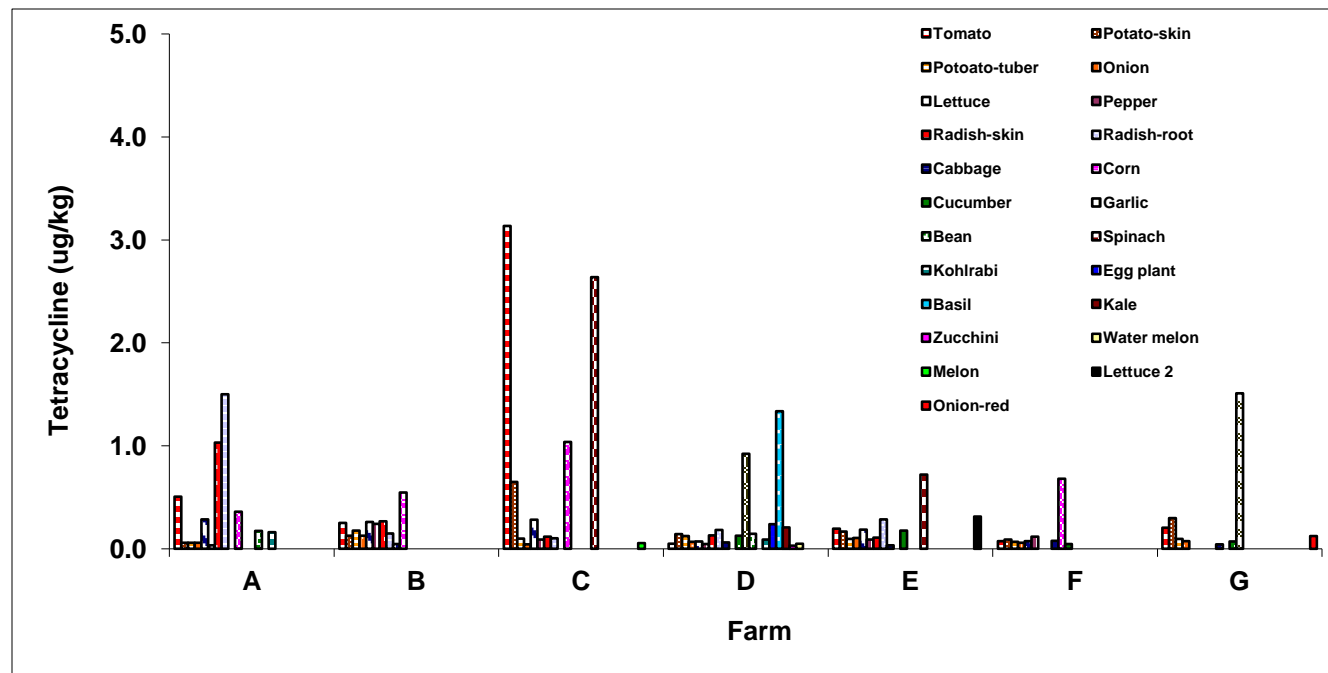


Figure 69: Tetracycline concentration in various vegetables from certified organic and conventional farms using manures. The LOQ for tetracycline was 0.83 $\mu\text{g/kg}$. Farms A, B, C, E, and F were certified organic farms whereas farms D and G were conventional farms.

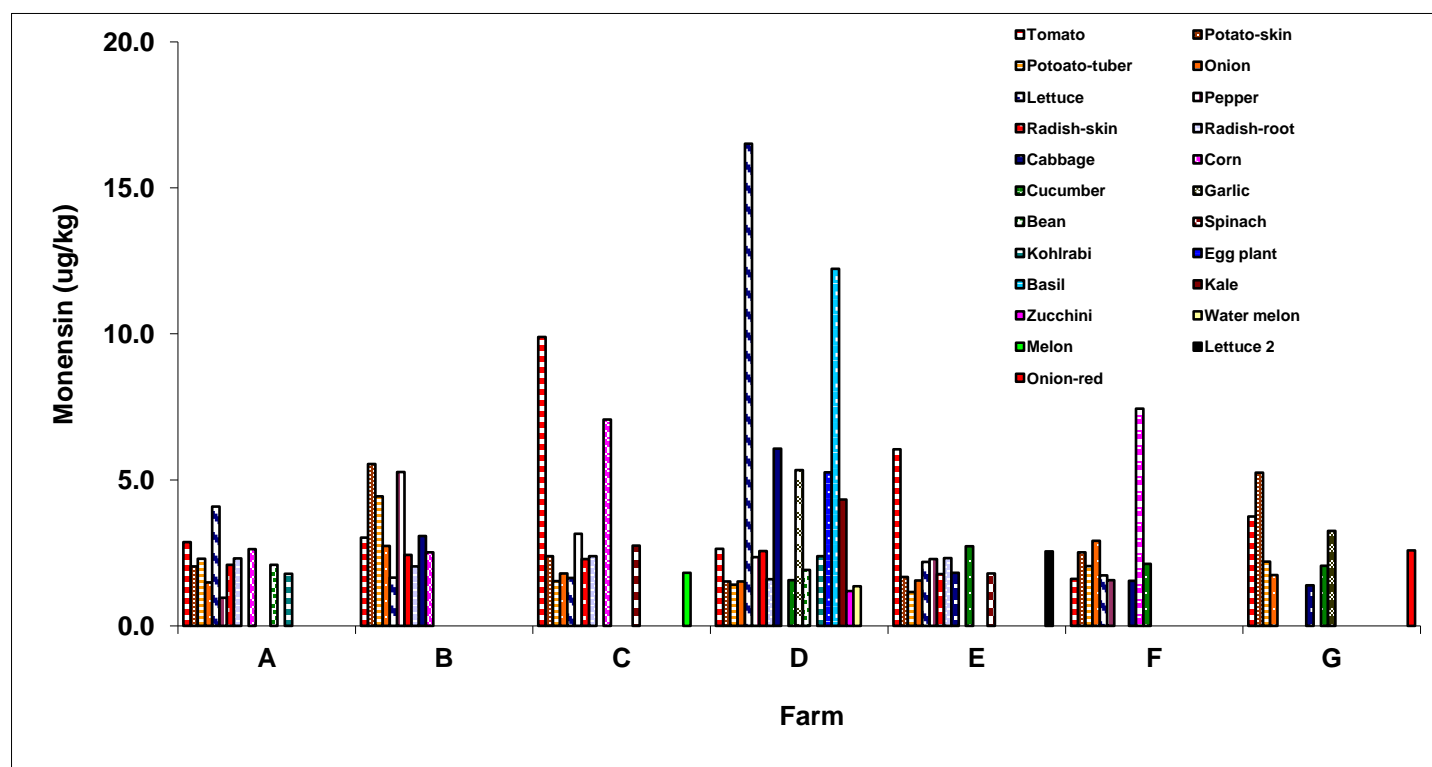


Figure 70: Monensin concentration in various vegetables from certified organic and conventional farms using manures. The LOQ for monensin was 2.17 $\mu\text{g/kg}$. Farms A, B, C, E, and F were certified organic farms whereas farms D and G were conventional farms.

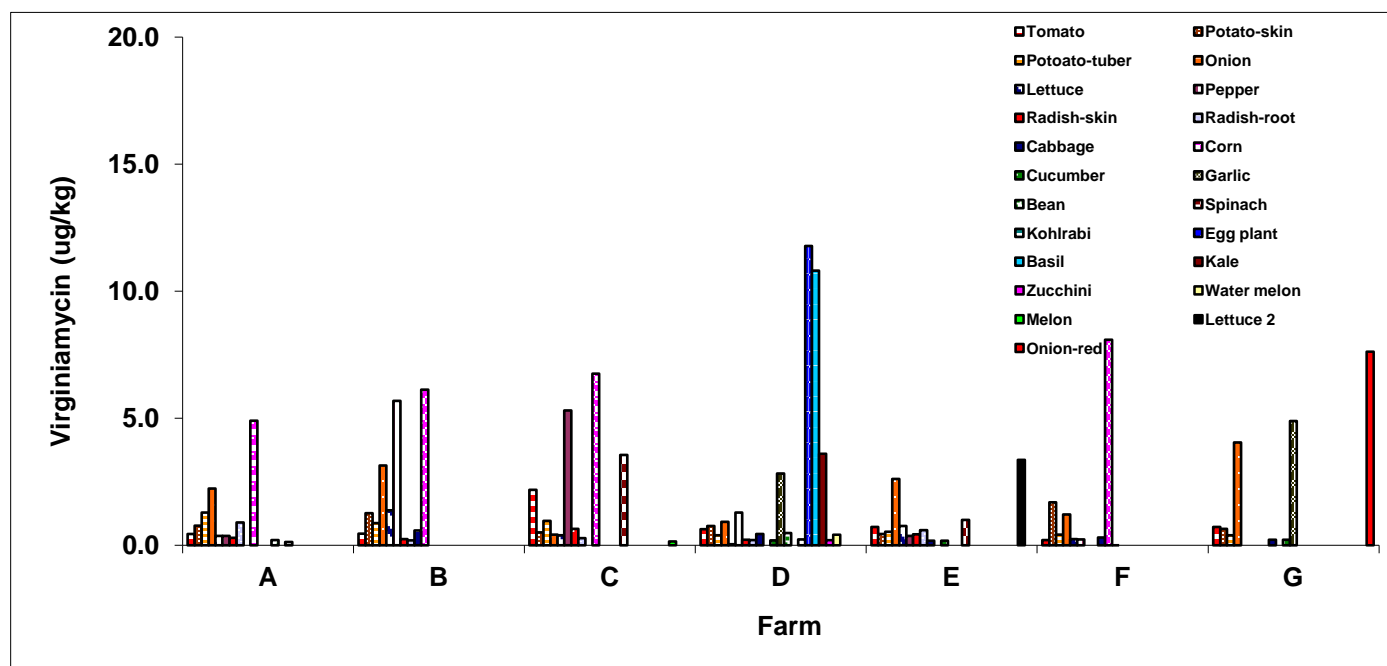


Figure 71: Virginiamycin concentration in various vegetables from certified organic and conventional farms using manures. The LOQ for virginiamycin was 1.58 $\mu\text{g/kg}$. Farms A, B, C, E, and F were certified organic farms whereas farms D and G were conventional farms.

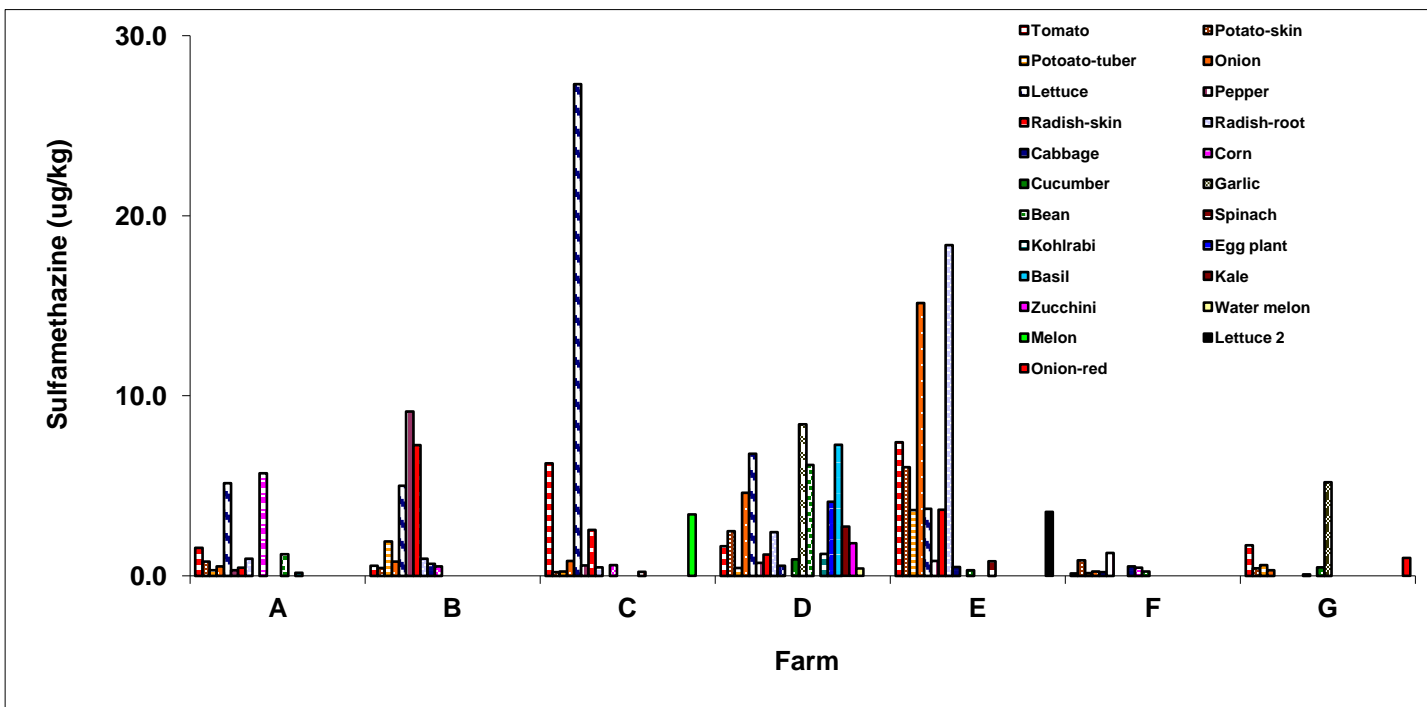
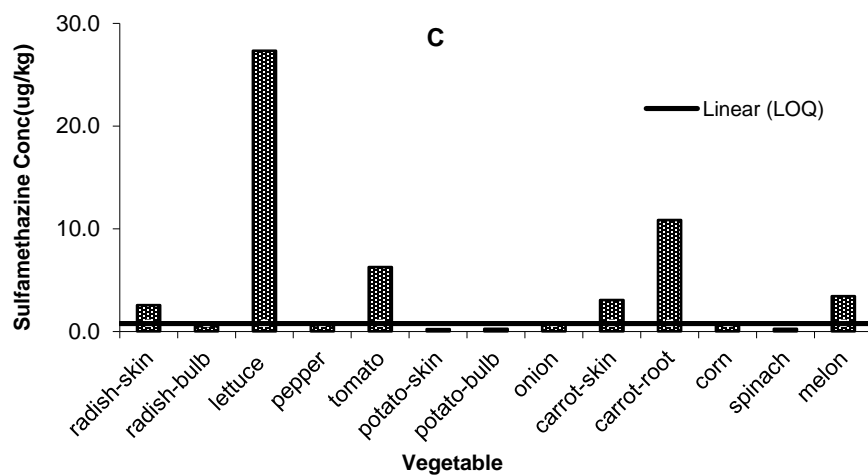
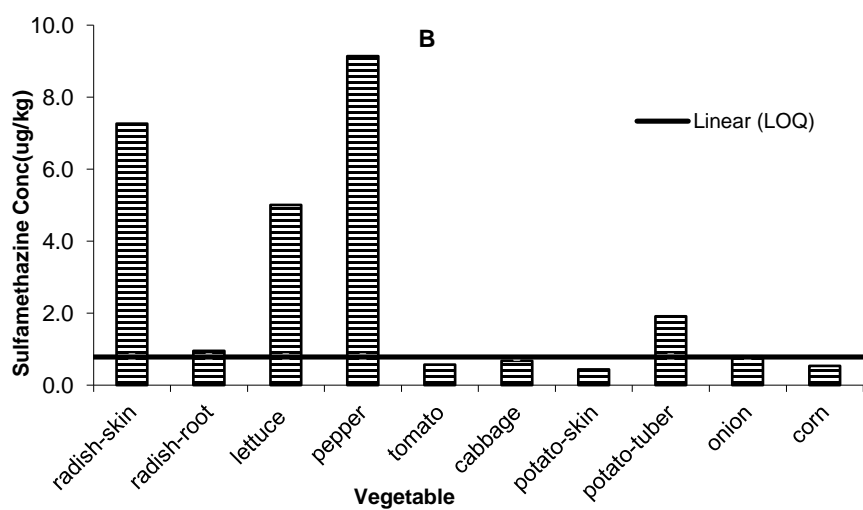
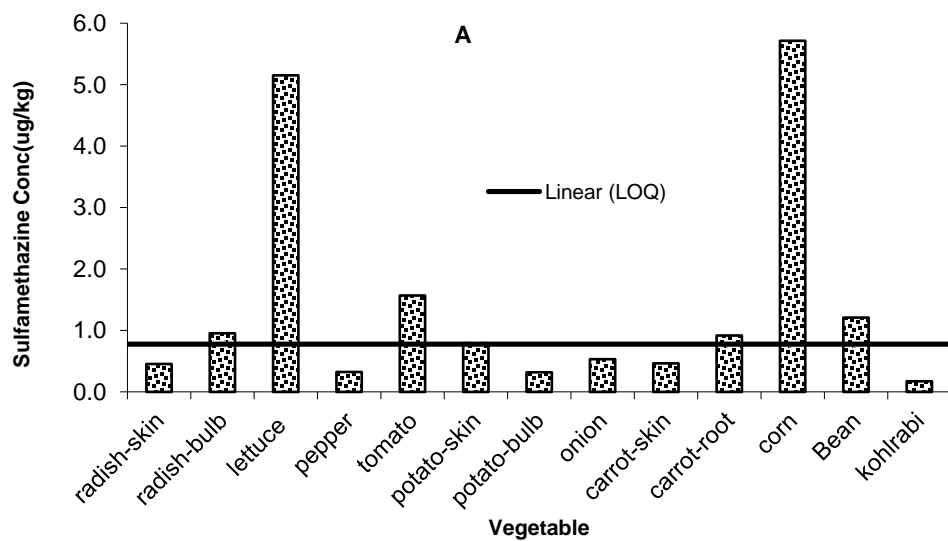
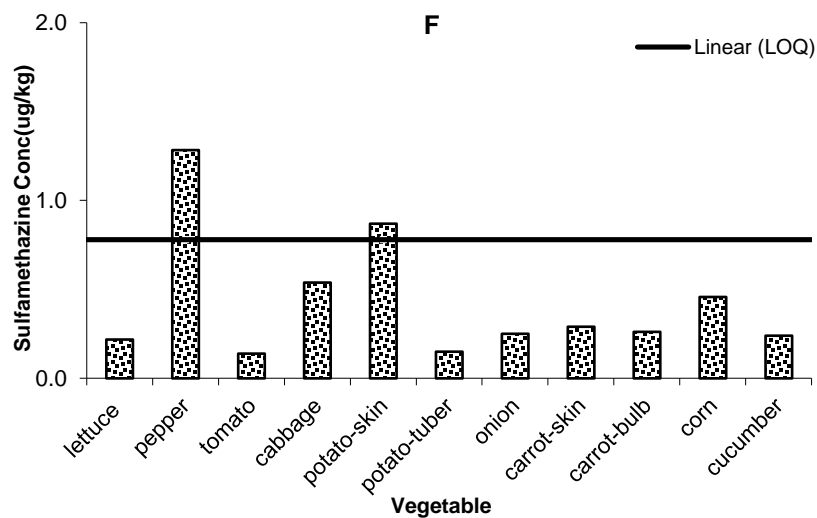
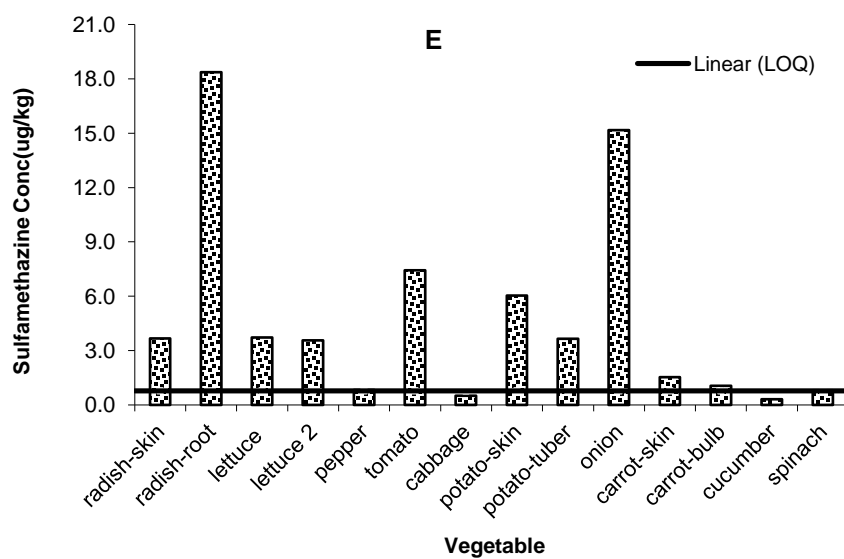
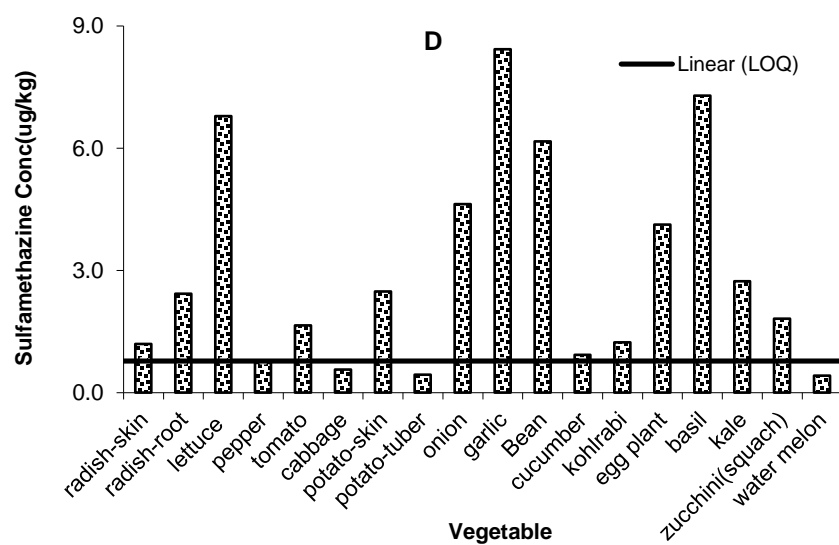


Figure 72: Sulfamethazine concentration in various vegetables from certified organic and conventional farms using manures. The LOQ for sulfamethazine was 0.78 $\mu\text{g/kg}$. Farms A, B, C, E, and F were certified organic farms whereas farms D and G were conventional farms.





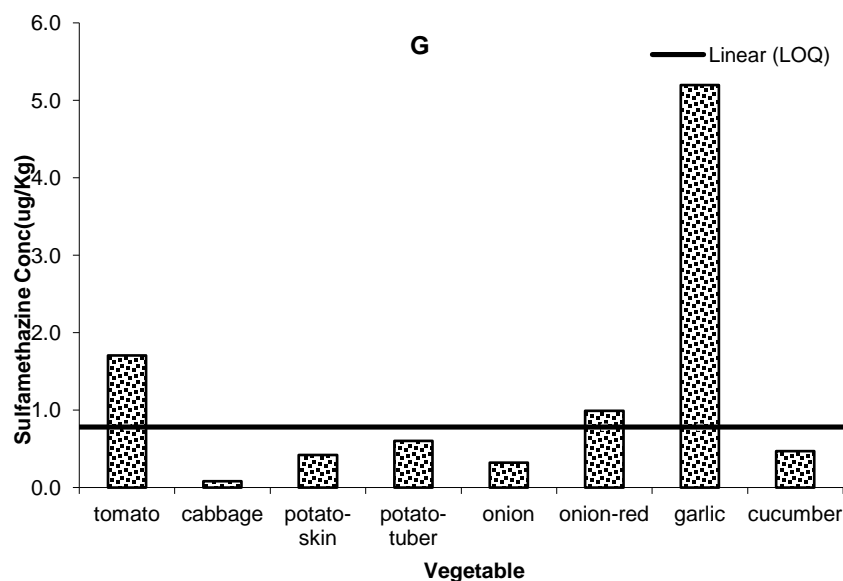


Figure 73. Sulfamethazine concentrations in various vegetables collected from local producers. The LOQ for sulfamethazine was 0.78 $\mu\text{g/kg}$. Farms A, B, C, E, and F were certified organic farms whereas farms D and G were conventional farms. Please note the scale differences between Figure A thru G.

Table 20. Vegetable analyzed for presence of antibiotics and the place of their production from two grocery stores that sell organic produce.

Vegetables	Grocery Store # 1	Grocery Store #2
Radish	Wisconsin	California
Potato	Minnesota	Wisconsin
pepper	Wisconsin	California
Cabbage	Minnesota	Wisconsin
Onion	Wisconsin	California
Carrot	Wisconsin	California
Tomato	N.A. [§]	California
Lettuce		California

[§] N.A.: Not available

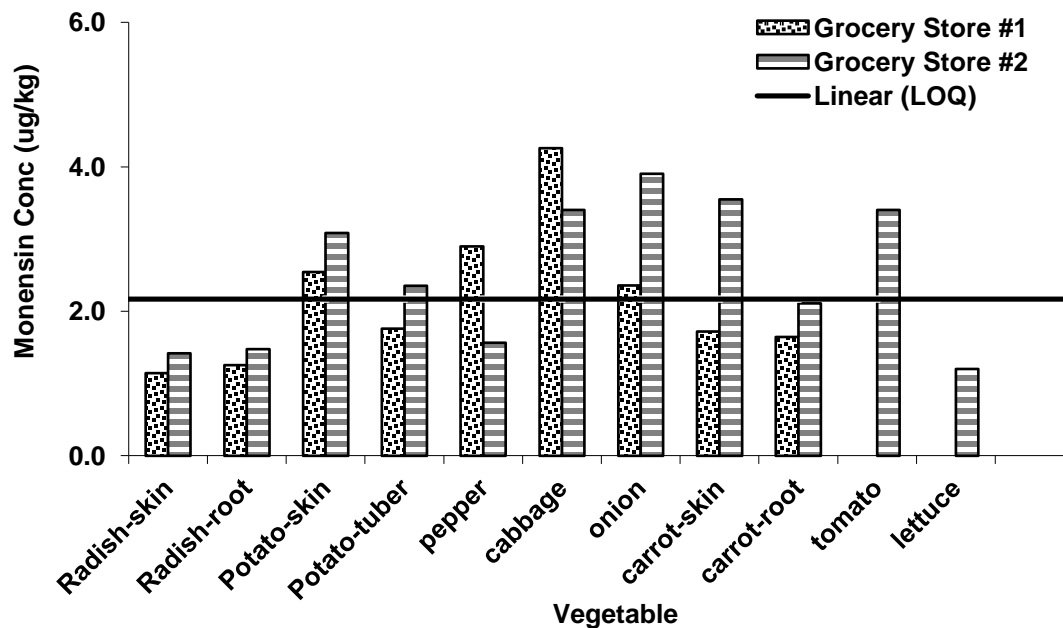


Figure 74. Monensin concentrations in vegetables bought from two grocery stores that sell organic food. The LOQ for monensin was $2.18 \mu\text{g kg}^{-1}$.

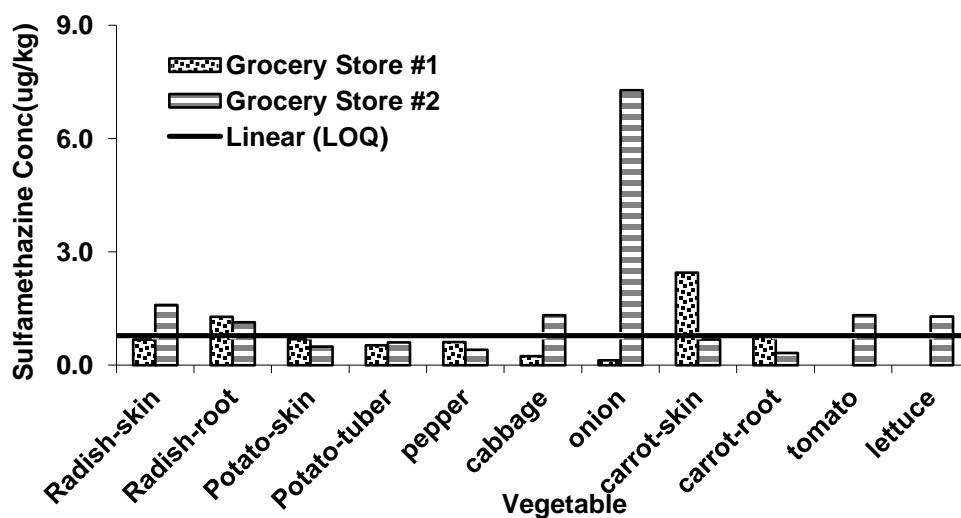


Figure 75. Sulfamethazine concentrations in vegetables bought from two grocery stores that sell organic food. The LOQ for sulfamethazine was $0.78 \mu\text{g kg}^{-1}$.

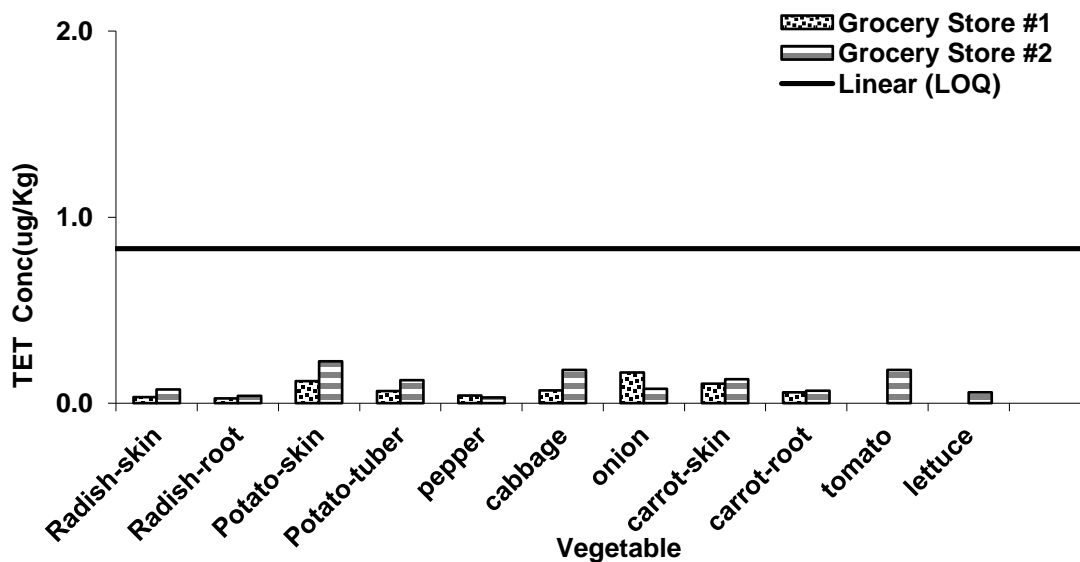


Figure 76. Tetracycline concentration in vegetables bought from two grocery stores that sell organic food. The LOQ for tetracycline was $0.83 \mu\text{g kg}^{-1}$.

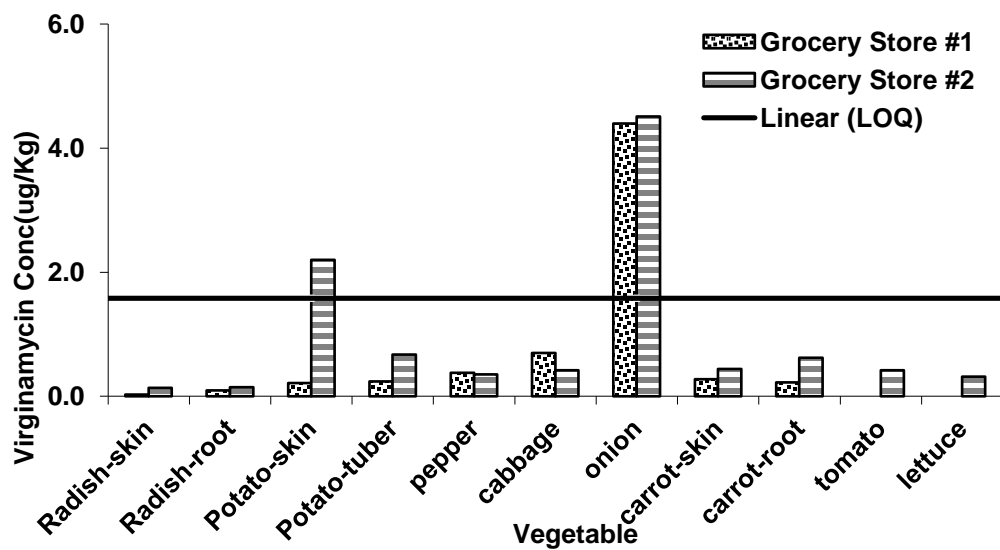


Figure 77. Virginiamycin concentration in vegetables bought from two grocery stores that sell organic food. The LOQ for virginiamycin was $1.58 \mu\text{g kg}^{-1}$.

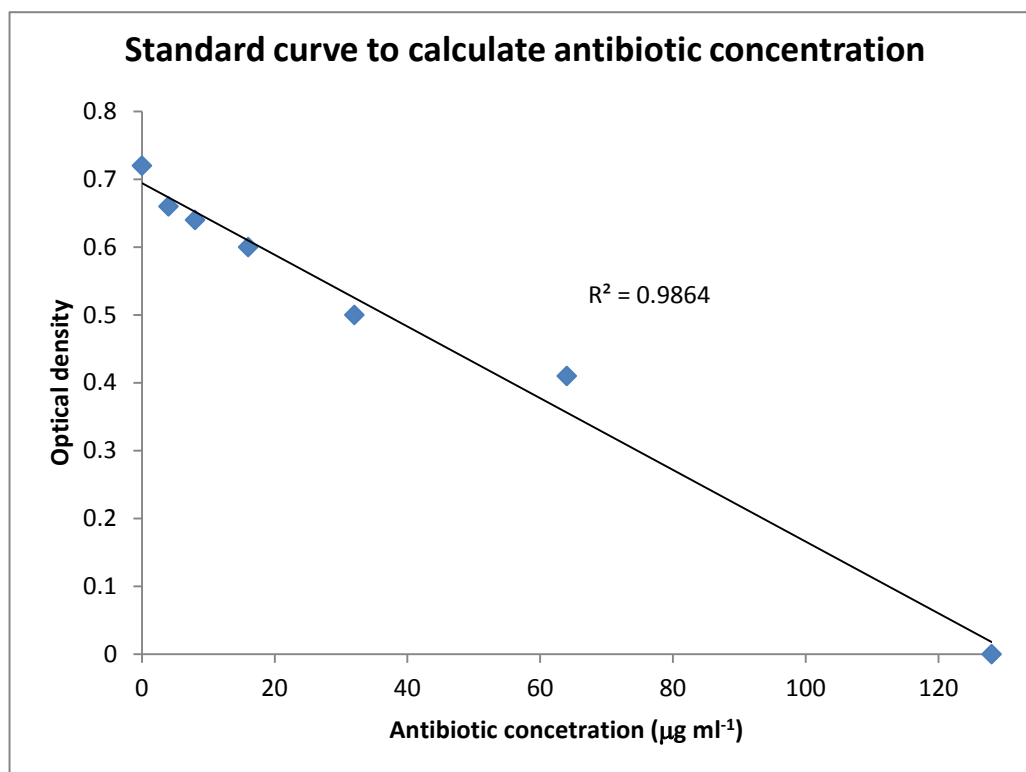


Figure 78: Standard curve for growth of *A. suis* at different sulfamethazine concentrations.

Table 21: Antibiotic daily intake and maximum residue limit of five antibiotics used in this study.

Antibiotic	ADI ($\mu\text{g/kg BW per day}$)	Maximum Residue limit (MRL) in animal tissue, $\mu\text{g/kg}$
Chlortetracycline	30 [§]	100-1200 [§]
Monensin	10 [§]	2-100 [§]
Tylosin	30 [§]	100-300 [§]
Sulfamethazine	-	100 ¹
Virginiamycin	-	100-400 ²

[§] <http://www.fao.org/ag/agn/jecfa-vetdrugs/search.html>

¹ http://www.dsm.com/le/nl_NL/delvotest/downloads/LesRisquesDeResidus_extrait2_En.pdf

² http://www.legislation.gov.hk/blis_ind.nsf/CURALLENGDOC/4D1FA97EA098B39C48256A7F001792E0?OpenDocument

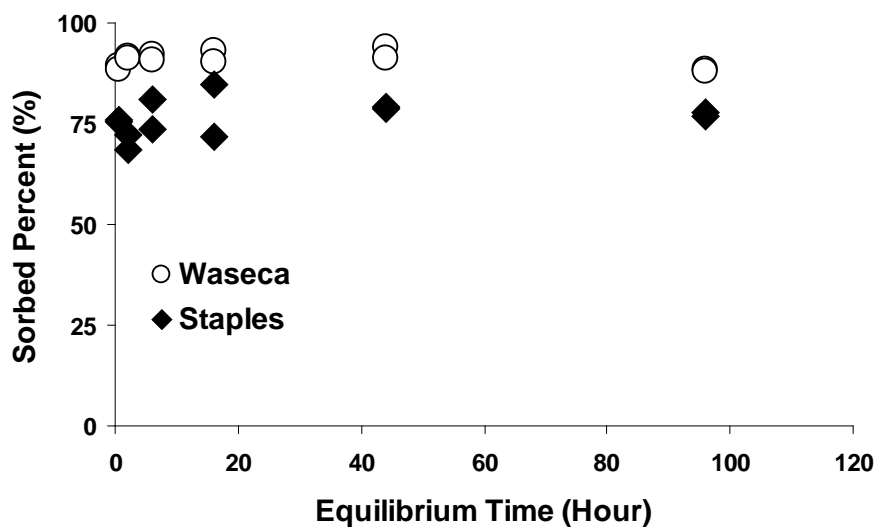


Figure 79. Variation in adsorption of sulfamethazine as a function of equilibration time for Webster clay loam from Waseca, MN and Verndale sandy loam from Staples, MN.

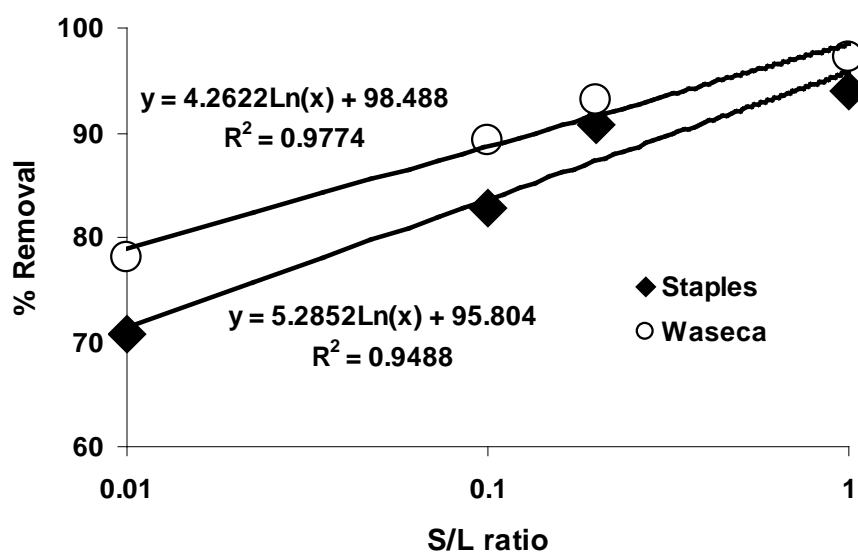


Figure 80. Effect of solid: liquid (S/L) ratio on sulfamethazine removal from solution (adsorption). This test was done with an initial sulfamethazine concentration of 1 μ M, at room temperature and neutral pH, agitation rate of 20 rpm, and agitation time of 18h.

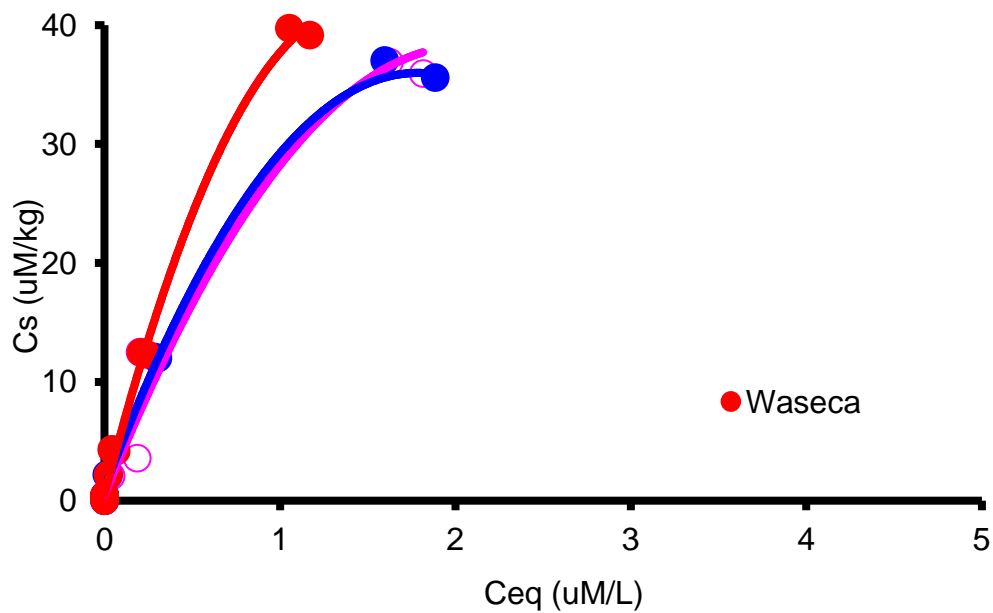


Figure 81. Concentration of soil adsorbed sulfamethazine (C_s) as a function of its concentration in solution (C_{eq}) with and without the presence of phosphate in solution in Webster clay loam soil. Lines are the best fit obtained using the Freundlich adsorption isotherm. \square and \circ signify zero phosphate solution, whereas \blacksquare and \bullet represent 1mM solution, and \blacksquare and \bullet are 10 mM phosphate solution.

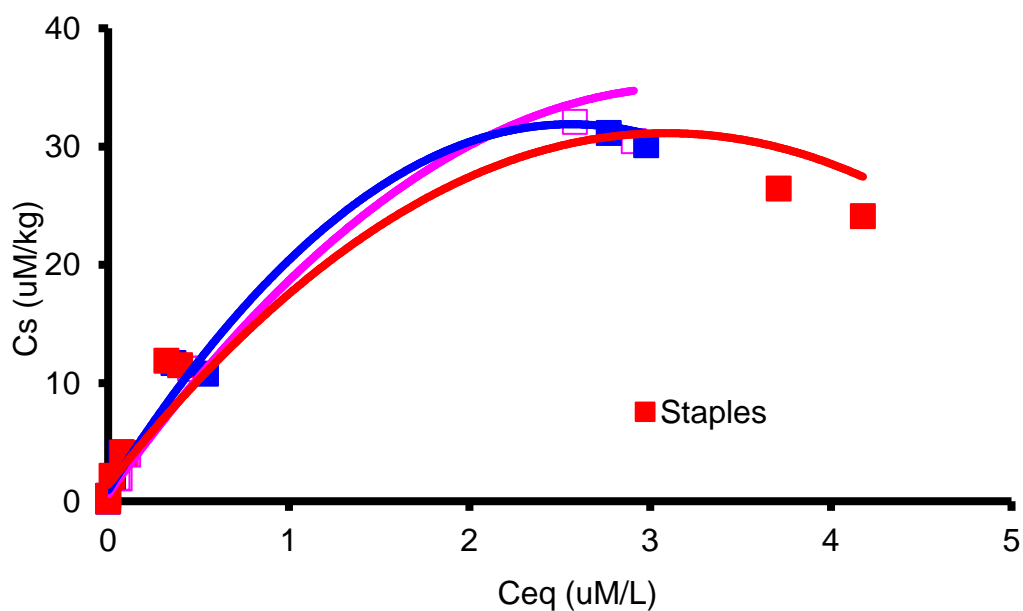


Figure 82. Concentration of soil adsorbed sulfamethazine (C_s) as a function of its concentration in solution (C_{eq}) with and without the presence of phosphate in solution in Verndale sandy loam. Lines are the best fit obtained using the Freundlich adsorption isotherm. \square and \circ signify zero phosphate solution, whereas \blacksquare and \bullet represent 1 mM solution, and \blacksquare and \bullet are 10 mM phosphate solution.

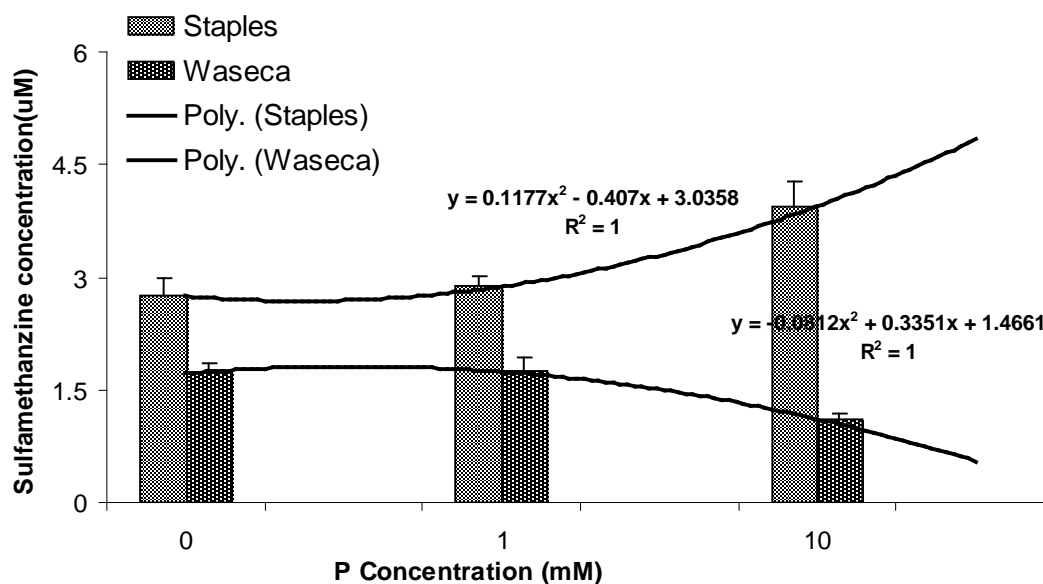


Figure 83. Sulfamethazine concentration remaining in the solution as function of different phosphate concentration. Initial sulfamethazine concentration was 10 μmolar.

Table 22. Freundlich isotherm parameters as a function of phosphate concentration and soil types.

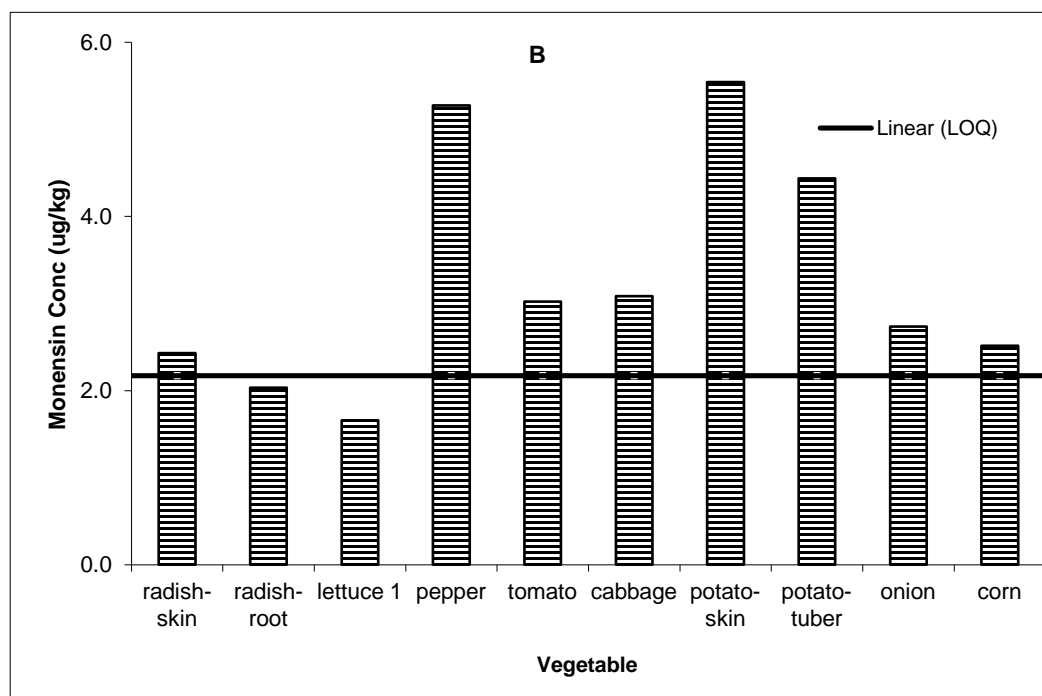
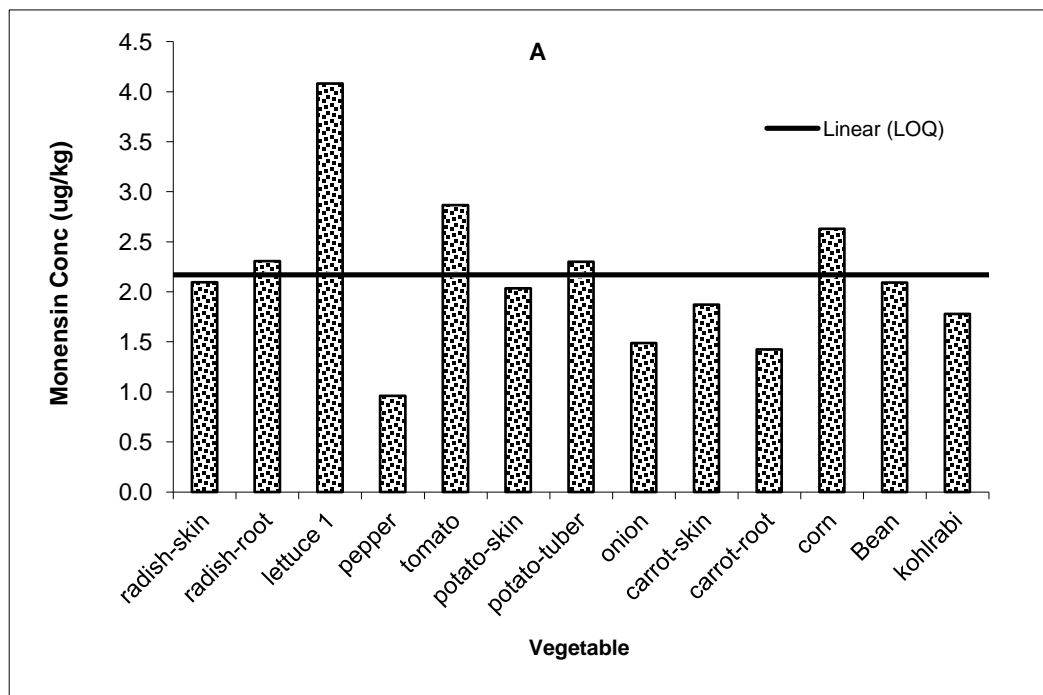
Phosphate Concentration, mM	Webster Clay loam		Verndale Sandy Loam	
	K_f^1	n^2	K_f	n
0	25.02	1.38	16.90	1.44
1	25.52	1.68	17.21	1.76
10	36.34	1.38	14.71	2.08

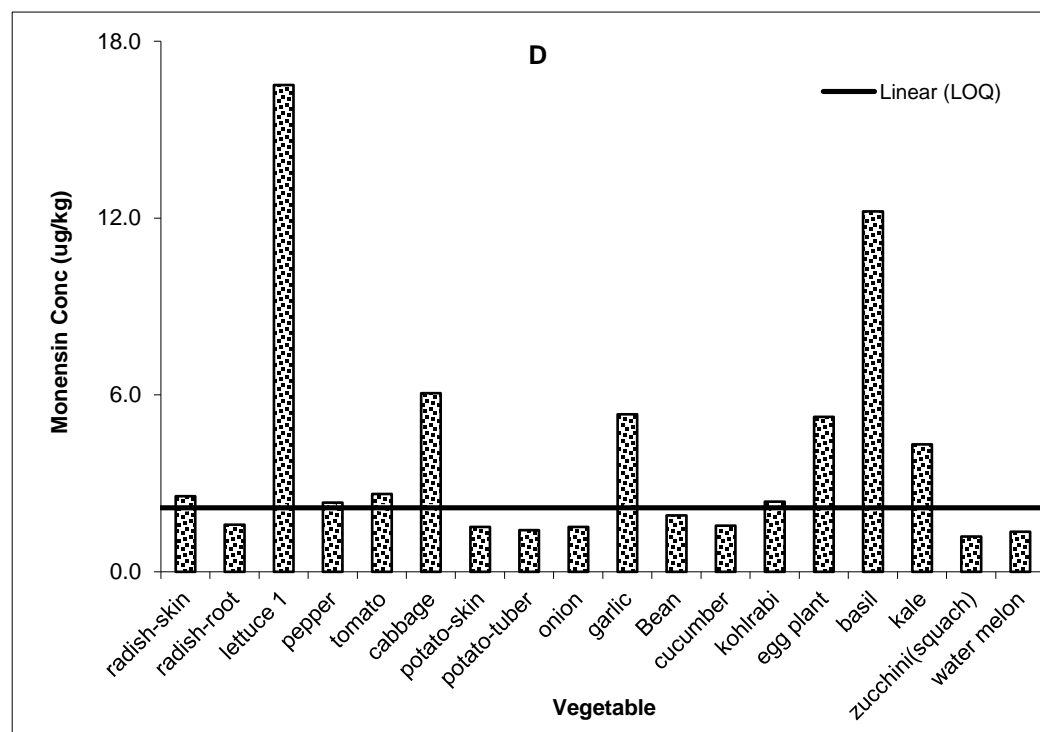
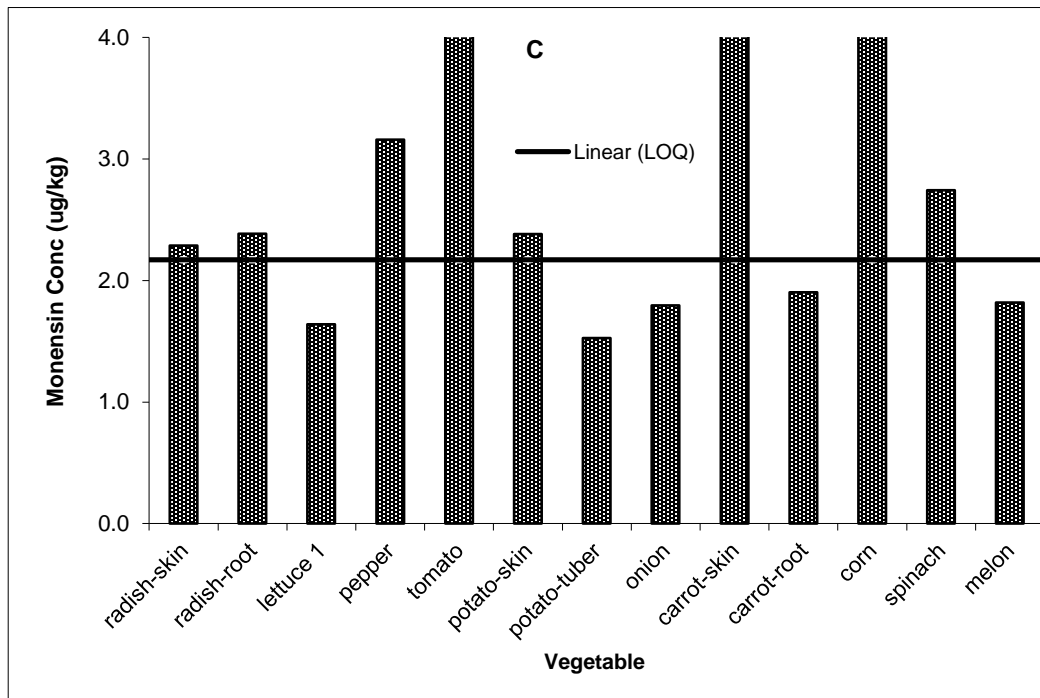
¹ Nonlinear Freundlich isotherm coefficient ($\text{mmol}^{1-n}\text{L}^n\text{kg}^{-1}$)

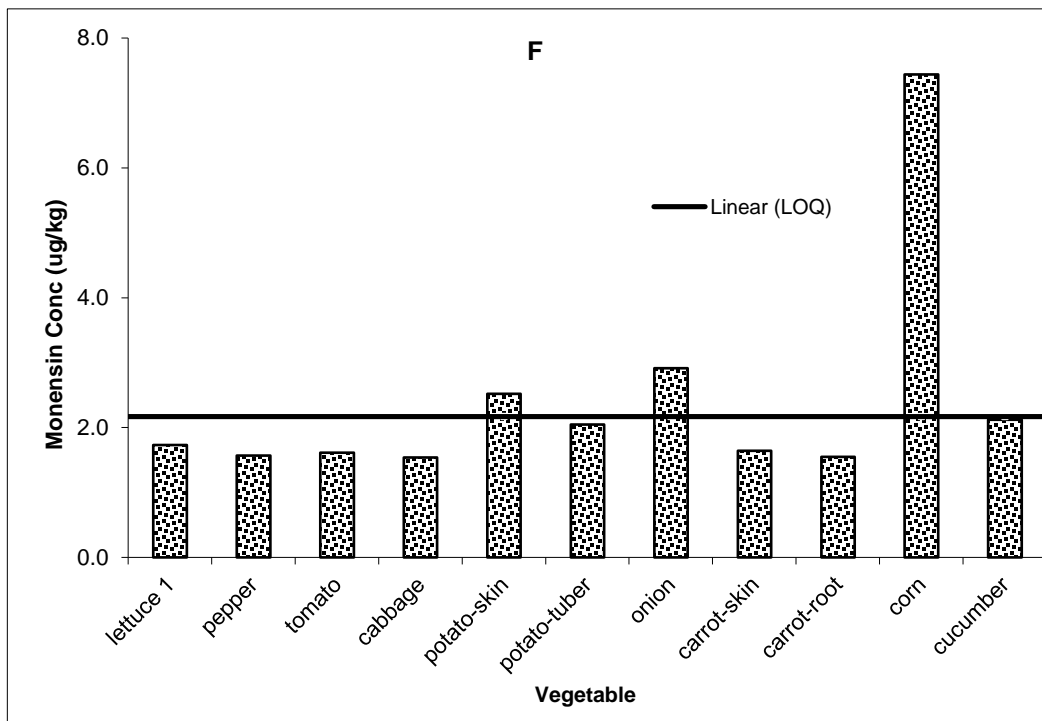
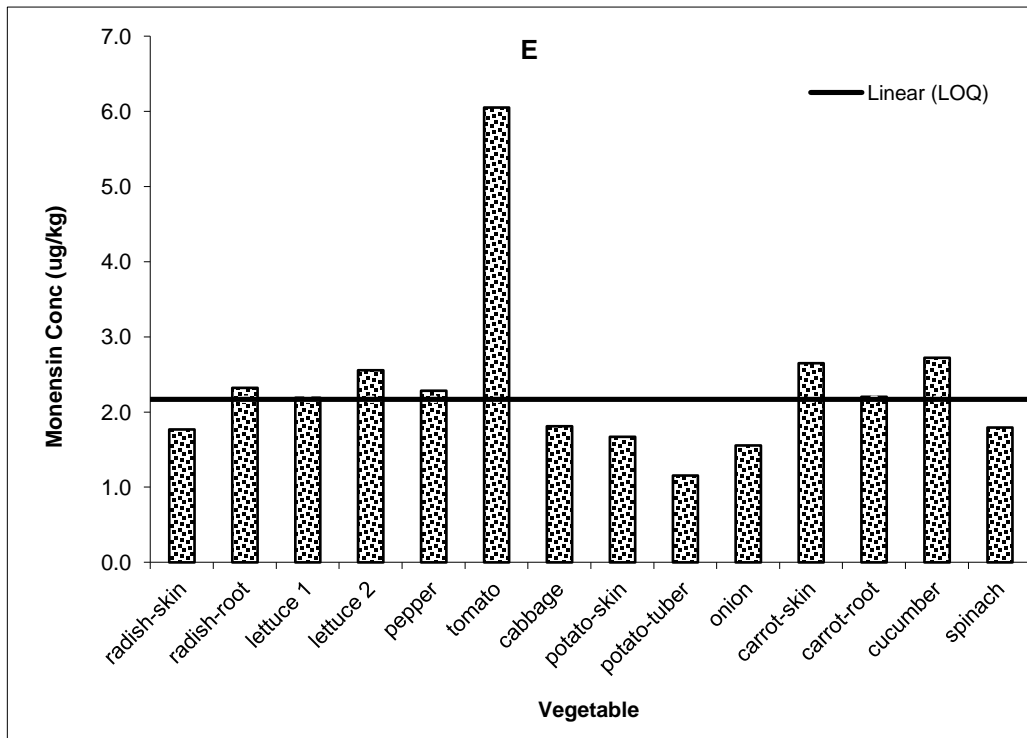
² Adsorption nonlinearity (unitless)

APPENDIX A

Figure 1A: Monensin concentration in various vegetable tissues from organic (A, B, C, E, F) and conventional (D, G) growers farms.







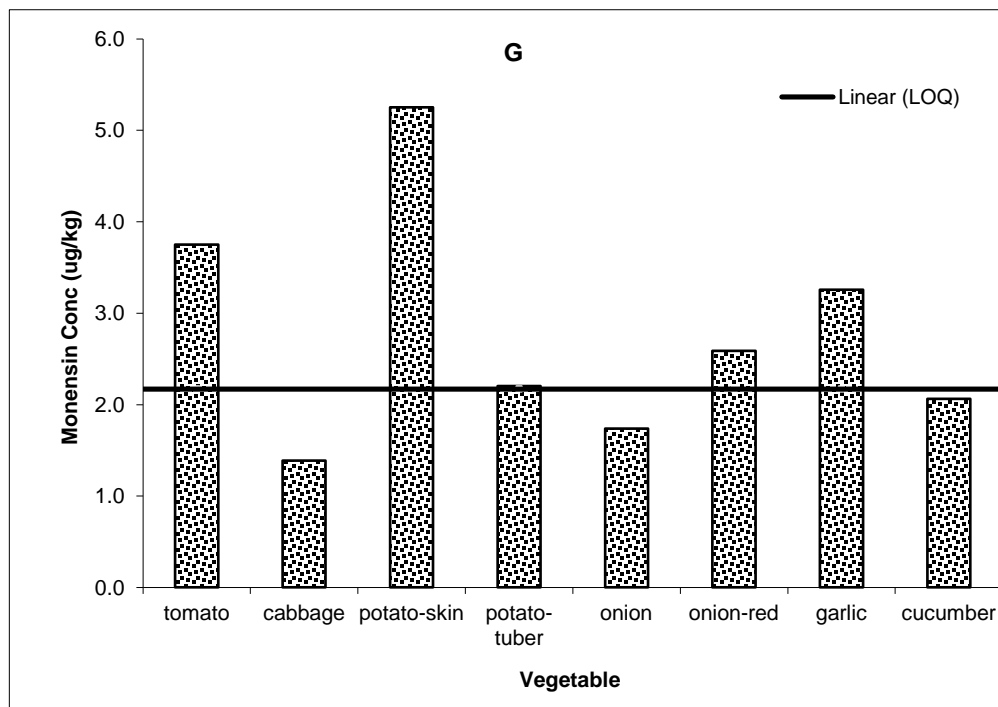
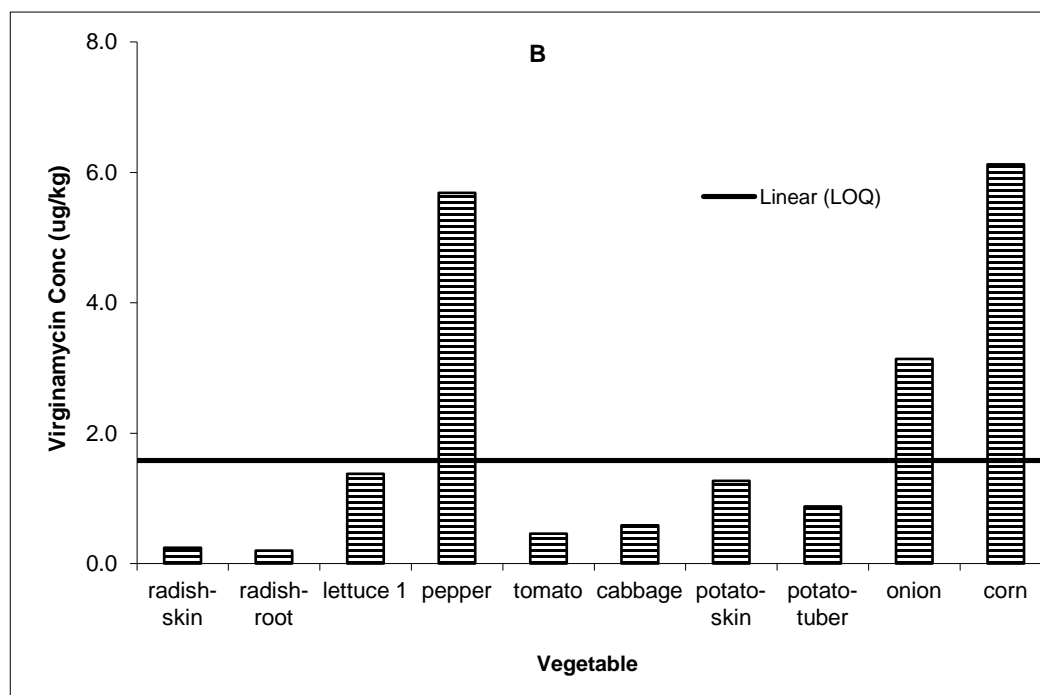
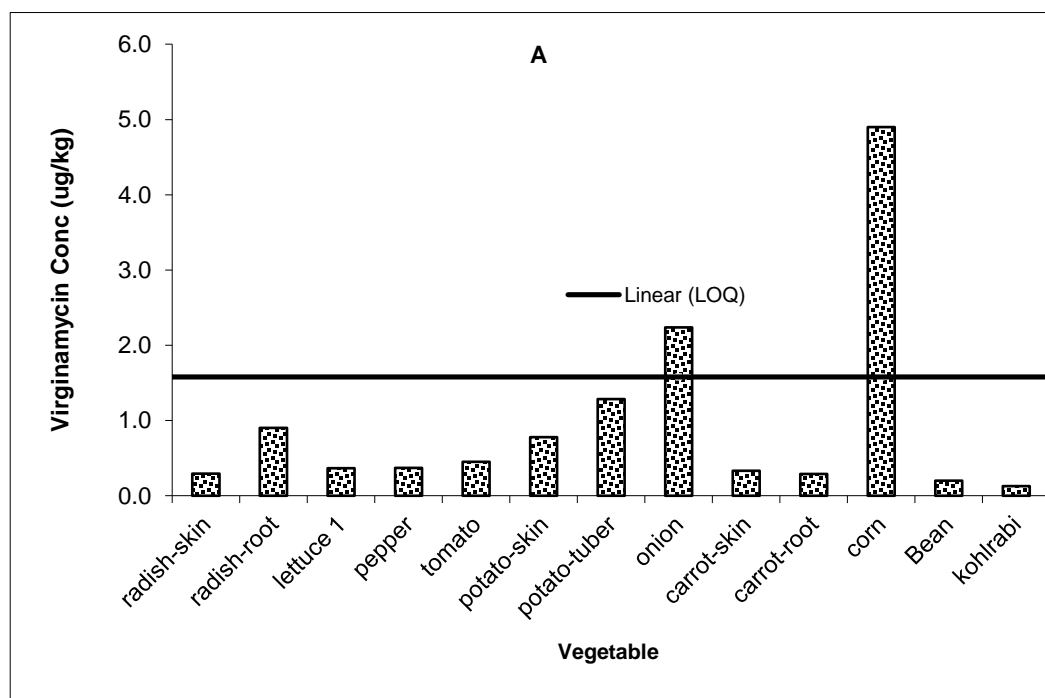
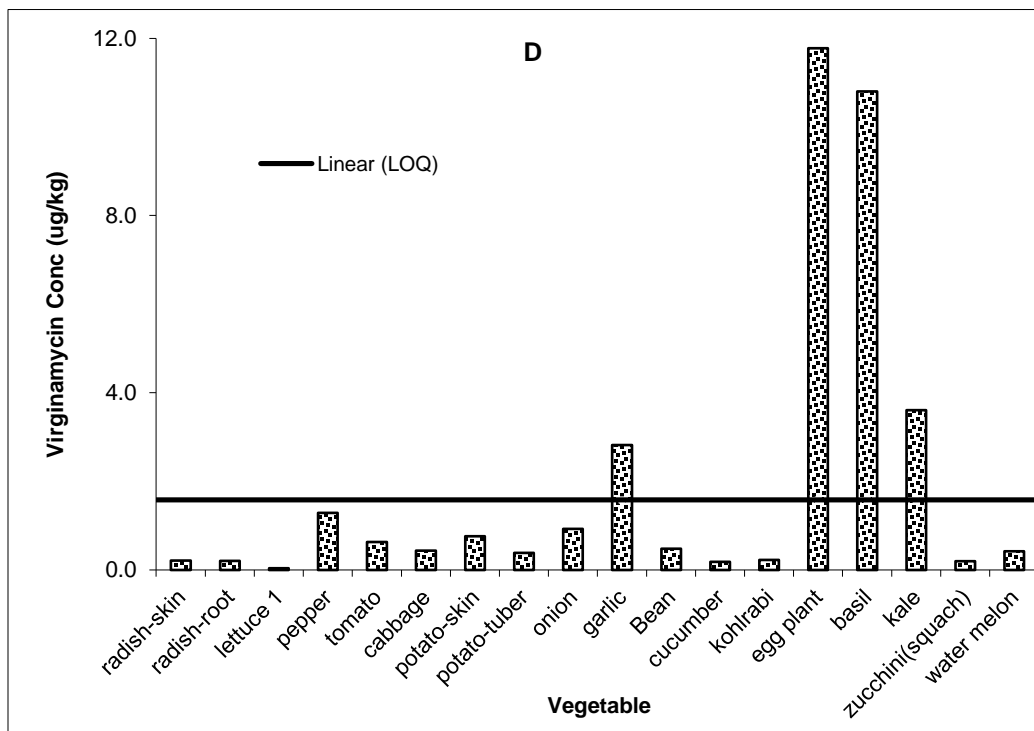
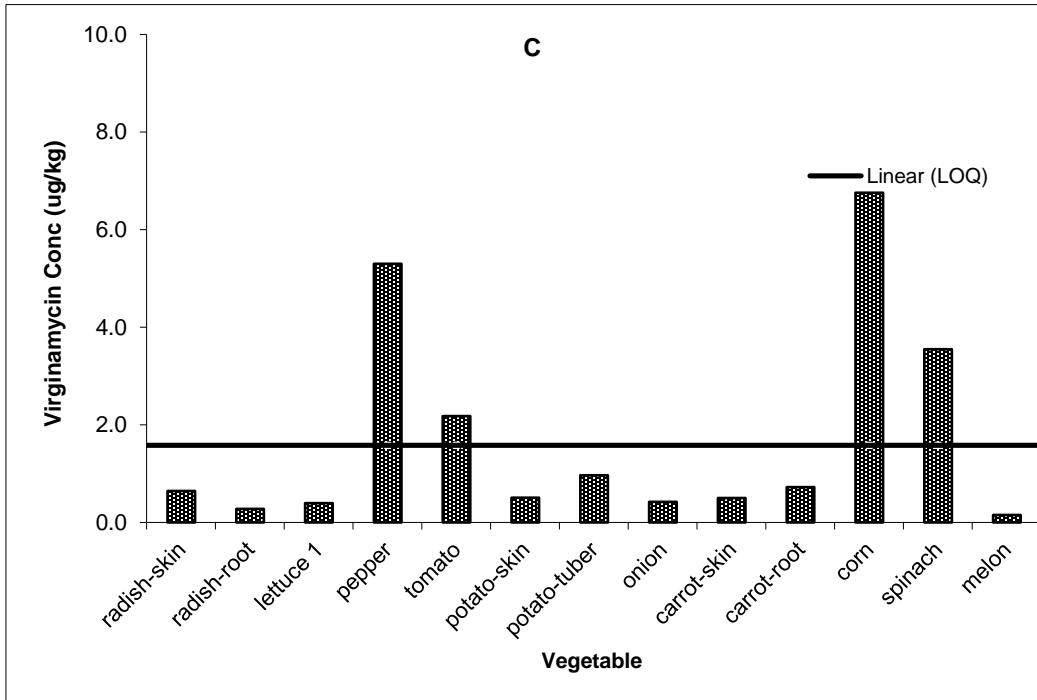
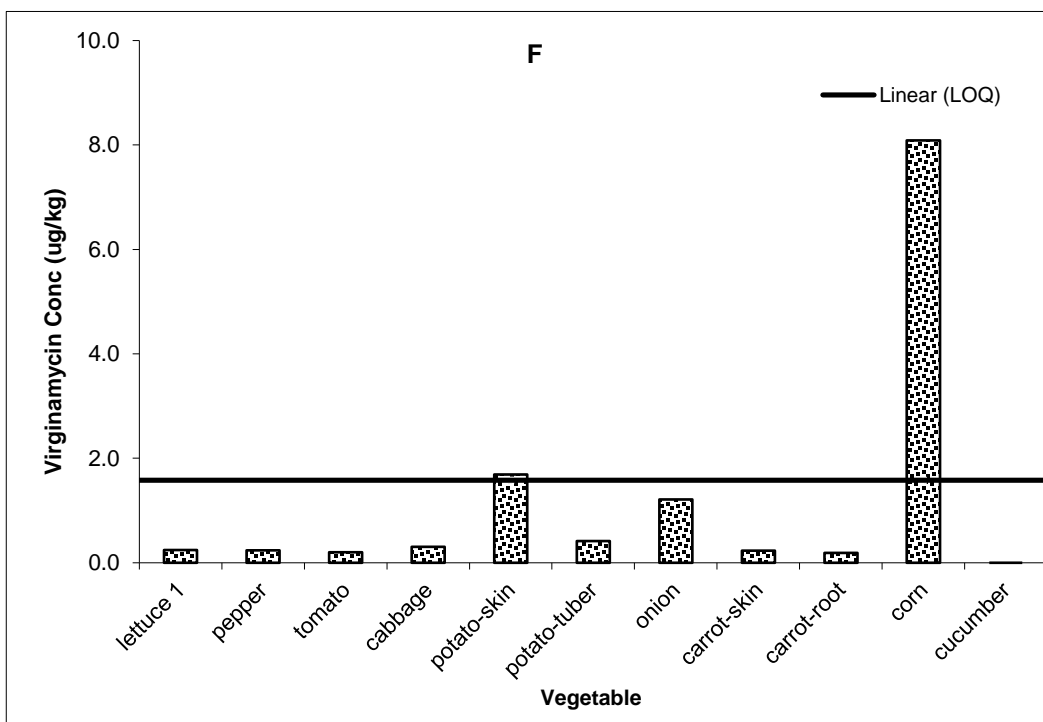
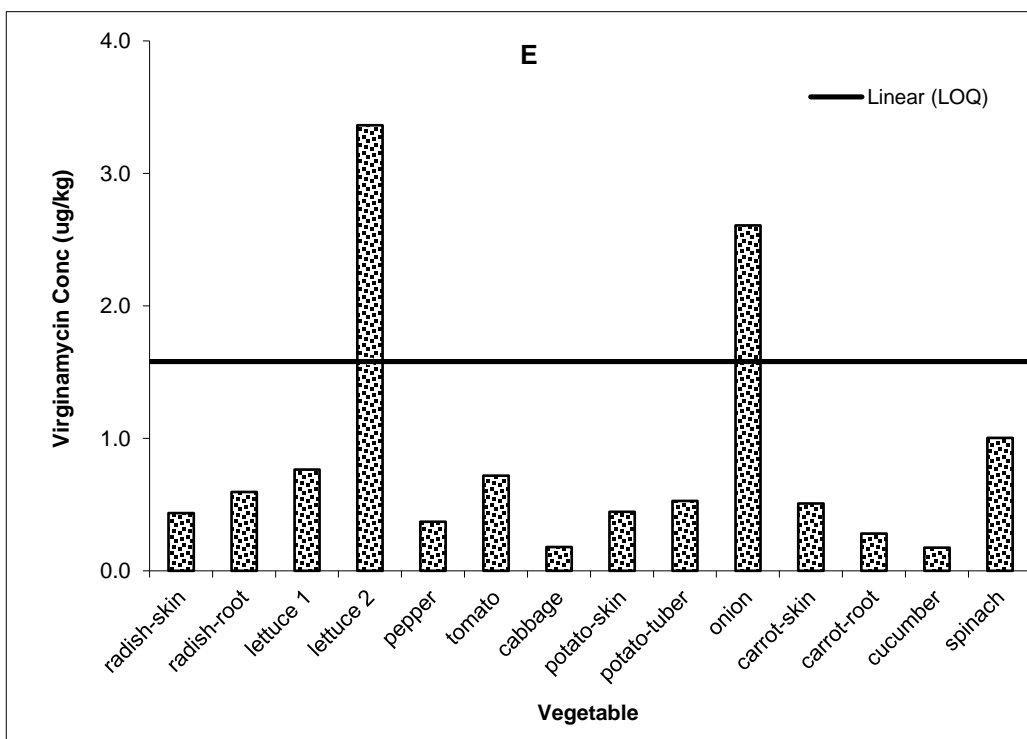


Figure 2A: Virginiamycin concentration in various vegetable tissues from organic (A, B, C, E, F) and conventional (D, G) growers farms.







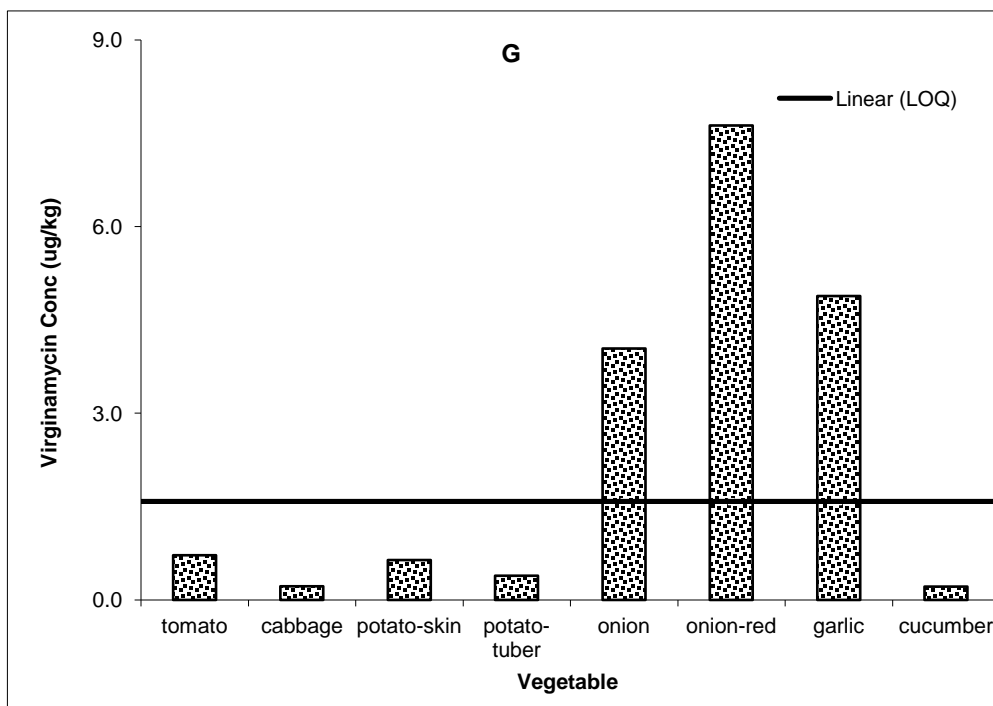


Figure 3A: Tetracycline concentration in various vegetable tissues from organic (A,B,C,E,F) and conventional (D,G) growers farms.

