Complicated Composting: Persistent Pyridine Carboxylic Acid Herbicides

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Executive Summary

This paper reviews pyridine carboxylic acid herbicide impacts on compost. Pyridine carboxylic acid herbicides are not completely broken down during grass growth, harvest and drying of hay, in the digestive tract of livestock, or during composting. These herbicides are a popular choice for broadleaf weed control because of this persistence: they remain effective for months or years. Pyridine carboxylic acids are also more effective than the common herbicide 2, 4-dichlorophenoxyacetic acid and can be applied to pastures with grazing livestock because they have low mammalian toxicity. The growth-inhibitory action of naturally occurring pyridine compounds has been researched since the discovery of α-picoline-γ-carboxylic acid in the early 1900's. These pyridine carboxylic acid compounds mimic plant growth hormones called auxins, causing plants to grow abnormally and then die. Plants injured by auxinic herbicides have poor seed germination, twisted growth, cupped or enlongated leaves, misshapen fruit, reduced yields, and ultimately die. Picloram (4-amino-3,5,6-trichloropyridine-2-carboxylic acid) was developed by Dow Chemical Company as a systemic herbicide for herbaceous weeds and woody plants in rights-of-way, forestry, rangelands, pastures, and small grain crops. Clopyralid (3,6-dichloropicolinic acid) was also developed by Dow Chemical Company to control annual and perennial broadleaf weeds in crops and turf. Another Dow herbicide, aminopyralid (4-amino-3,6-dichloro-2-pyridine carboxylic acid), is used for broad leaf weed control in pastures. Aminocyclopyrachlor (6-amino-5-chloro-2-cyclopropylpyrimidine-4-carboxylic acid) is the first pyrimidine carboxylic acid herbicide and was developed by DuPont for weed and brush control on uncultivated non-agricultural areas, uncultivated agricultural areas, industrial sites, and natural areas. Clopyralid compost contamination was reported in 2000 at four different facilities including Washington State University. In Vermont, compost samples were tested and found to contain aminopyralid, clopyralid, and picloram in 2012. Across the U.S. since 2000, there have been many reports of apparent plant injury from compost contaminated with auxinic herbicides. Because of the limited testing facilities and expense of chemical testing, the majority of these reports remain anecdotal. If the history of a compost feedstock is unknown, bioassays are recommended to test compost for the presence of auxinic herbicides. Even though pyridine carboxylic acid herbicides are sold with proper labeling and restrictions, compost contamination is continuing. Adjustments should be made for the registered uses of these herbicides, and herbicide applicators need improved education about the implications of contaminating compost feedstock.

Introduction

Pyridine carboxylic acids are popular herbicides used for broadleaf weed control that have recently been implicated in cases of "killer compost" where plants fail to thrive due to residual herbicide in compost. The problem with pyridine carboxylic acid herbicides is persistence: they are not completely broken down during grass growth, harvest and drying of hay, in the digestive tract of livestock, or even during composting. Composting has become popular because it not only reduces the load on landfills but also creates an organic soil amendment to use in gardens. Increased use of pyridine carboxylic acids has the potential to contaminate compost feedstock including hay, manure, grass clippings, and brush. Herbicide persistence involves chemical, biological, and communication issues that are causing concerns for the composting industry as well as backyard composters. The objective of this paper is to review the history of pyridine carboxylic acid herbicide use and compost contamination, specificially discussing picloram, clopyralid, aminopyralid, and aminocyclopyrachlor.

History and Background

Naturally occurring pyridine compounds inhibit plant growth and have been researched since the early 1900's (Hall, 1985). The first reported pyridine derivative having growth-inhibitory action was α-picoline-γ-carboxylic acid isolated from infertile soil in Takoma Park, Maryland (Schreiner and Shorey, 1908). Schreiner and Shorey studied the effects of pyridine (C_5H_5N), picoline ($C_5H_4NCH_3$), and piperidine ($C_5H_{11}N$) on wheat seedlings.They found that pyridine in concentrations as low as 50 mg/kg inhibited plant growth and turned the leaf tips brown. Picoline killed wheat seedlings at a concentration of 1000 mg/kg and caused injury to plant tops at concentrations between 500-1000 mg/kg. Piperidine killed and injured plants at lower concentrations than pyridine and picoline and appeared to injure roots more than plant tops. Piperidine neutralized with acetic acid was more toxic than the alkaline piperidine by itself (Schreiner and Shorey, 1908). Other naturally occurring pyridine compounds include fusaric acid (5-butyl-2-pyridine carboxylic acid), nicotinic acid (3-pyridine carboxylic acid), nicotine (3-(1-methyl-2-pyrrolidinyl)pyridine), NAD (nicotinamide adenine dicnuleotide), and trigonelline (N-methyl nicotinic acid) (Hall, 1985).

Pyridine carboxylic acid herbicides were developed to control a wide variety of broadleaf weeds and to remain effective for several months to years. Persistence of pyridine carboxylic acid herbicides is actually what makes them a favorite choice for broad leaf weed control. They replaced existing herbicides that required three to five applications per season to control problematic broad-leaf weeds including thistle, ragweed, nightshade, and dandelion (Bezdicek et al., 2001). These herbicides are also significantly more effective at killing some broadleaf weed species than the common herbicide 2,4-D (Hall, 1985). Found in a variety of formulations, pyridine carboxylic acids are registered for use on pastures, forage crops, grain crops, some fruits and vegetables, turf, wildlife management areas, forests, industrial sites, and rights-of-way. Toxicity data submitted to the United States Environmental Protection Agency (USEPA) show that pyridine carboxylic acids have very low mammalian toxicity; this makes them a popular choice because livestock can safely graze treated pastures and consume treated hay (Hagood et al., 2012). Fluroxypyr and aminopyralid are registered by DowAgroSciences under the USEPA Conventional Reduced Risk Pesticide Program, which means that they presumably have a low impact on human health, lower toxicity to non-target organisms, low use rates, low pest resistance potential, and are compatible with Integrated Pest Management practices (USEPA, 2012). The most prevalent persistent herbicides in the pyridine carboxylic acid family are picloram, clopyralid, aminopyralid, and aminocyclopyrachlor (USCC, 2013).

Plant Uptake and Symptoms

Pyridine carboxylic acids are growth-regulator herbicides that work by mimicking plant growth hormones called auxins, causing plants to grow abnormally. Synthetic auxin herbicides bind at the receptor sites normally used by the plant's natural growth hormones; this causes abnormal growth and ultimately plant death (DowAgroSciences, 1998). In 1946, F.A. Gilbert made the statement that auxin herbicides cause susceptible plants to "grow themselves to death," and researchers have had a difficult time discovering the details of these herbicides' phytotoxicity (Grossmann, 2000).

Auxinic herbicides mimic the biphasic effects of indole-3-acetic acid (IAA), which is the main natural auxin in higher plants (Grossmann, 2000). At low concentrations, auxins stimulate growth through cell division and elongation. At higher concentrations, many growth abnormalities occur within 24 hours, including leaf epinasty (downward curving of leaves), stem curvature, root and shoot growth inhibition, intensified green leaf pigmentation, and decrease in stomatal aperture. Next, rapid senescence with chloroplast damage and chlorosis occurs along with membrane and vascular system destruction. This causes dessication, necrosis, and ultimately plant death. The effects of auxinic herbicides are sometimes described as an "auxin overdose" (Grossmann, 2000).

Overproduction or abundance of auxins may also induce synthesis of 1-aminocyclopropane-1-carboxylic acid (ACC) synthase which catalyses the production of ethylene (Grossmann, 2000). Ethylene is a gaseous hormone that is involved in plant responses to stress and regulation of plant growth and senescence. The herbicide effects of leaf abscission and epinastic growth could be caused by auxin or ethylene. Additionally, when ACC is oxidized to form ethylene, cyanide is also produced in plant-damaging concentrations. Cyanide accumulation could be another mechanism for plant growth inhibition and tissue necrosis. Auxin compounds also cause accumulations of abscisic acid (ABA) in root and shoot tissue. ABA is an important hormone for promoting leaf senescence and controls plant growth through stomatal movement and cell division. A combination of actions influenced by auxins, ACC, ethylene, cyanide, and ABA may ultimately lead to plant death from pyridine carboxylic acids (Grossmann, 2000).

Pyridine carboxylic acid herbicides cause the following plant injury symptoms: poor seed germination, twisted growth, twisted or elongated leaves, cupped leaves, misshapen fruit, reduced yields, and plant death (Hagood et al., 2012). Figures 1 – 6 below are photos from Washington State University's Whatcom County Extension website (http://whatcom.wsu.edu/ag/aminopyralid/images.html) of plants damaged by aminopyralid.

Figure. 1: Cupped leaves on a tomato plant

Figure 2: Abnormal leaf growth on a pepper plant

Figure 3: Damaged tomato plant

Figure 4: Cupped sunflower leaves

Figure 5: Curled and twisted eggplant

Figure 6: Damaged spinach

Picloram

Picloram (4-amino-3,5,6-trichloropyridine-2-carboxylic acid) was developed by Dow Chemical Company and registered for use in the U.S. in 1964 (USCC, 2013). Picloram is a systemic herbicide used to control deeply rooted herbaceous weeds and woody plants in rights-of-way, forestry, rangelands, pastures, and small grain crops. It is applied in the greatest amounts to pasture and rangeland, followed by forestry. Since 1978, picloram has been a restricted-use pesticide, and may only be applied by (or under the supervision of) a licensed pesticide applicator (USEPA, 1995). Some of the weeds controlled by picloram include chicory (*Cichorumintybus* L.), dandelion (*Taraxacum officinale* L.), goldenrod (*Solidago canadensis* L), horsenettle (*Solanum carolinense* L.*)*, common plantain (*Plantago major* L.*)*, annual ragweed (*Ambrosia artemisiifolia* L.*)*, leafy spurge (*Euphorbia esula L.)*, yellow starthistle (*Centaurea solstitialis* L.*)*, bull thistle (*Cirsium vulgare* L.*)*, and common vetch (*Vicia sativa* L.*)* (Dow AgroSciences, 2009) (NRCS, 2013).

Picloram is very active in soil, and residues as low as 0.25 μg/kg soil can be toxic to sensitive broadleaf crops (Jotcham et al, 1989). The half-life of picloram in soil ranges from 167-513 days (USEPA, 1995). Soybean injury has been observed up to five years after applying 2.75 kg/ha of picloram. A study of picloram persistence in silt-loam soil found that soybeans would not grow 269 days after the soil was treated with 3.8 kg/ha picloram. When picloram was applied at 0.38 kg/ha, soybeans would grow nine months after treatment (Jotcham et al., 1989). Picloram is extremely mobile, has a high potential for leaching, and is resistant to biotic and abiotic degradation (USEPA, 1995). The USEPA Office of Drinking Water's STORET database reported that picloram was found at detectable levels in 420 of 744 surface water samples. Picloram is regulated by the Safe Drinking Water Act (SDWA) and water supply systems are required to sample for it (USEPA, 1995).

In Virginia, the following herbicides registered for use contain picloram: Forestry Tordon™ K, Forestry Tordon™ 101, Forestry Tordon™ 101R, Grazon® P+D, HiredHand™ P+D, Outpost® 22K, Pathway®, Picloram +D, Picloram 22K, Sekor™ P+D, Surmount™, Terva™ 22K, Toram™ P+D, Tordon™ 101 Mixture, Tordon™ 101 Weed and Brush Killer, Tordon™ 101R Forestry Herbicide, Tordon™ RTU, Tordon™ K/22K, Trooper™ 22K, Trooper™ Extra Selective, Trooper™ 101 Mixture, Trooper™ P+D, and Trooper™ Pro (Hagood et al., 2012). Many of these herbicides contain 2,4-D in addition to picloram. Excerpts from Tordon™ 101 Mixture's label are as follows (DowAgroSciences, 2009):

- *Recommended for the control of herbaceous broadleaf weeds, woody plants, and vines on forest planting sites and non-crop areas including industrial, manufacturing, and storage sites; rights-of-way, such as electrical power lines, communication lines, pipelines, highways, railroads, and wildlife openings in forest and non-crop areas.*
- *Herbicidal effects of Tordon 101 Mixture occur primarily from uptake by plant foliage and translocation throughout the plant, however, herbicidal activity may occur from soil uptake of picloram. Very small amounts can kill or damage broadleaf plants. To prevent damage to crops and other desirable plants, carefully follow all directions and precautions.*
- *Do not rotate food or feed crops on treated land if they are not registered for use with picloram until an adequately sensitive bioassay or chemical test shows that no detectable picloram is present in the soil.*
- *Grazing restrictions: there are no grazing restrictions for non-lactating dairy animals or other livestock. Do not allow lactating dairy animals to graze treated areas within 7 days after application. Do not harvest grass cut for hay from treated areas for 30 days after application. Meat animals must be withdrawn from treated forage at least 3 days before slaughter.*
- *Do not transfer livestock from treated grazing areas onto sensitive broadleaf crop areas without first allowing 7 days of grazing grass that has not been treated with picloram. Otherwise, urine may contain enough picloram to cause injury to sensitive broadleaf plants.*
- *Do not use manure from animals grazing treated areas on land used for growing broadleaf crops, ornamentals, orchards, or other susceptible, desirable plants. Manure may contain enough picloram to cause injury to susceptible plants.*
- *Do not use plant material from treated areas for composting or mulching of susceptible broadleaf plants.*
- *Tordon 101 Mixture should not be applied on residential or commercial lawns or near ornamental trees or shrubs.*

Clopyralid

Clopyralid (3,6-dichloropicolinic acid) was discovered in 1961, and was launched for use in the late 1970's in Europe to control annual and perennial broadleaf weeds in certain crops and turf (DowAgroSciences, 1998). Clopyralid was registered for use by DowAgroSciences in the U.S. in 1987 (Cox, 1998).Clopyralid is particularly effective against these broadleaf weed families: *Asteraceae* (sunflower, cocklebur, ragweed, chicory, scent less chamomile, sagebrush, Canada thistle, knapweed, dandelion, and perennial sow-thistle); *Fabaceae* (clover, black medic, vetch, mesquite); *Solanaceae* (nightshade, jimsonweed); and *Polygonaceae* (buckwheat)

(DowAgroSciences, 1998). Clopyralid has a low toxicity to animals including humans (Bezdicek et al., 2001).

Clopyralid is soluble in water, and does not breakdown by hydrolysis or exposure to sunlight. Clopyralid has a half-life of greater than 30 days in water with or without sunlight exposure. Studies using 19 different U.S., Canadian, and European soils were conducted to determine clopyralid half-life under different conditions. For warm (25° Celsius) and moist conditions, the half-life range was 8-250 days and averaged 28 days. For cool (10° Celsius) temperatures, the average half-life increased to 64 days. For hot soil temperatures (30°-35° Celsius), the average half-life was 19 days. A study of 20 field sites found similar results with an average half-life of 25 days and a range of 8-66 days. However, water logged soils and anaerobic conditions slow the degradation of clopyralid; in these conditions the half-lives were greater than 365 days (DowAgroSciences, 1998).

Several standard vegetable plants in the *Leguminosae, Solonaceae, and Compositae* families are sensitive to clopyralid at levels as low as 10 µg/kg (Michel and Doohan, 2003). These plants include tomato (*Solanum lycopersicum* L.), red clover (*Trifolium pratense* L.), lettuce (*Latuca sativa* L.), pea (*Pisum sativum* L.), lentil (*Lens culinaris* L.), sunflower (*Helianthus anuus* L.), pepper (*Capsicum* L.), and beans (*Phaseolus vulgaris* L.) (NRCS, 2013). Taking into consideration the half-lives listed above and that the application rate of clopyralid on grass is 10,000-50,000 µg/kg, there is great potential for clopyralid residues in compost to damage crops. Clopyralid compost contamination was documented in Ohio, Washington, Pennsylvania, New Jersey, and California between 2000 and 2003 (Michel and Doohan, 2003).

Commercial herbicides containing clopyralid may contain 3,6-dichloropicolinic acid or its three salts: triethylamine, triisopropylamine, and monoethanolamine. Some herbicides contain clopyralid in combination with triclopyr, MCPA, and/or 2,4-D (Cox, 1998). There are many products currently registered for use in Virginia that contain clopyralid: Accent Gold®, Anderson's products with Millennium™ Ultra, Brazen™, Broadstrike™ Plus, Broadstrike™ Plus Corn PRE/PPI, Clean Slate®, Clopyr Ag, Clopyralid 3, Confront®, Curtail®, Cody, Hornet®, Howard Johnson's Weed & Feed with Millennium™ Ultra, Kudzu Killer, Lebanon Proscape, Lontrel Turf and Ornamental, Millenium™ Ultra 2, Quali-Pro 2-D, Redeem R & P®, Refute®, Scott's fertilizer plus Confront®, Spur®, Staunch™, and Stinger® (Hagood et al., 2012). Clopyr Ag, manufactured by United Phosphorus Inc., is specified for use on non-residential turf, cemeteries, golf courses, industrial sites, non-cropland, parks, rights-of-way, and roadsides. It has a 48 page product label, and following are some excerpts (United Phosphorus Inc., 2005):

- *Do not send clippings to a compost facility.*
- *Do not collect grass clippings for mulch or compost.*
- *Applicator must give notice to landowners/property managers to not use grass clippings for composting.*
- *Do not use on residential turf.*
- *Do not transfer livestock from treated grazing areas (or feeding of treated hay) to sensitive broadleaf crop areas without first allowing 7 days of grazing on an untreated pasture (or feeding untreated hay). If livestock are transferred within less than 7 days of grazing untreated pasture or eating untreated hay, urine and manure may contain enough clopyralid to cause injury to sensitive broadleaf plants.*
- *Because Clopyr Ag can affect broadleaf plants by either foliar contact or root uptake, it should not be applied either directly or indirectly(via spray drift) to the following plants: grapes, flowers, tomatoes, potatoes, beans, lentils, peas, alfalfa, sunflowers, soybeans, safflower, vegetables not listed on this label, and other susceptible broadleaf crops/ornamentals*
- *Do not use hay or straw from treated areas or manure or bedding straw from animals that have grazed or consumed forage from areas treated with Clopyr Ag to compost or mulch where susceptible plants may be grown the following season.*
- *The decomposition of clopyralid (the active ingredient) in crop residues or manure is optimal in warm, moist soil conditions and may be enhanced by supplemental irrigation. To promote the decomposition of Clopyr Ag in plant residues or manure, the residues should be either burned or evenly mixed with the soil.*

Aminopyralid

Aminopyralid (4-amino-3,6-dichloro-2-pyridine carboxylic acid) was registered for use in the U.S. in 2005 by DowAgroSciences (USEPA, 2005). Aminopyralid is used for broad leaf weed control because it is a long-lasting, systemic herbicide that has pre- and post-emergence activity. One of aminopyralid's biggest selling points is that livestock can safely graze treated pastures. Products registered for use in Virginia that contain aminopyralid are: Capstone™, Chaparral™, Opensight®, ForeFront® HL, ForeFront® R&P, Milestone™, Milestone™ VM, Milestone™ VM Plus, and Radar™ (Hagood et al., 2012).

Aminopyralid half-life in five different soils ranged from 32 days to 533 days (Mikkelson and Lym, 2011). When aminopyralid is applied in the fall, it tends to remain in the soil longer because little degradation occurs when soil temperatures are cool or below freezing. Predicting soil persistence is difficult because it relies on many factors including specific microorganisms, environmental conditions, and soil characteristics (Mikkelson and Lym, 2011).

Dow has created an aminopyralid stewardship website that describes key facts, use precautions, and restrictions. The use precautions and restrictions are clearly outlined in the website and on herbicide labels that contain aminopyralid (DowAgroSciences, 2013):

- *Treated hay may not be exported from the United States.*
- *Aminopyralid does not degrade in plants and takes 3 days to pass through a grazing animal's digestive system. Therefore, manure from animals that have grazed/eaten hay from treated areas within the previous three days may only be used on pasture grass, wheat, or corn.*
- *Hay, straw, and manure from treated areas may not be composted.*
- *Wait one year to rotate from treated pasture to cereal or corn crops. Wait at least two years to rotate to broadleaf crops. Prior to planting broadleaf crops, perform a bioassay test to ensure the remaining aminopyralid will not harm the broadleaf crop.*
- *If treated hay is sold, the buyer must be informed about the aminopyralid use restrictions.*

The website also provides a sample form that can be used to inform hay recipients about restrictions, as well as the graphic in Figure 7 below.

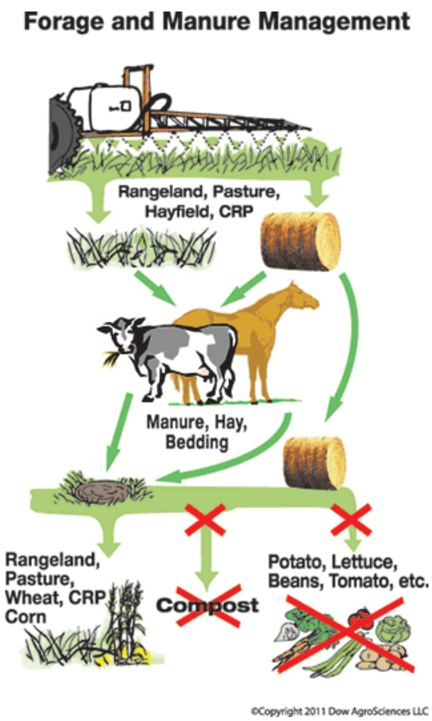

Figure 7. From www.dowagro.com/range/aminopyralid_stewardship.htm

Aminocyclopyrachlor

Aminocyclopyrachlor (6-amino-5-chloro-2-cyclopropylpyrimidine-4-carboxylic acid) is the first pyrimidine carboxylic acid herbicide (Bukun et al., 2010). Aminocyclopyrachlor is structurally similar to aminopyralid, clopyralid, and picloram, but it has one additional nitrogen atom in the heterocyclic ring and a cyclopropyl side chain (Bukun et al., 2010). DuPont developed aminocyclopyrachlor and applied for registration with the EPA on January 27th, 2010. Aminocyclopyrachlor products marketed in Virginia include Method® 50SG, Method® 240SL, Perspective®, Streamline®, and Viewpoint® (Hagood et al., 2012). Method® 50SG is registered for general weed and brush control on uncultivated non-agricultural areas (airports, highway, utility and railroad rights-of-way, etc.), uncultivated agricultural areas (farmyards, fencerows, fuel storage areas, ditchbanks, etc.), industrial sites (lumberyards, pipeline and tank farms, etc.), and natural areas (wildlife management areas and habitats) (DuPont, 2012).

DuPont's aminocyclopyrachlor herbicide Imprelis® was developed and marketed to turf professionals to treat difficult turf weeds, and appeared to be a great choice because of its low toxicity to mammals (Patton et al., 2011). Imprelis® was first sold in October 2010, and by late spring 2011 damage to trees was being reported. The EPA issued a stop-sale order on August 1, 2011 because Imprelis® was damaging trees even when applicators followed label directions. Imprelis® damaged many evergreen trees, especially Norway spruce and white pines. DuPont voluntarily stopped selling Imprelis® and created a return and refund program for landowners with trees harmed by Imprelis® in October 2011. Information about this program can be found at http://imprelis-facts.com (Patton et al., 2011).

After tree damage was caused by Imprelis®, DuPont revised the product labels for other aminocyclopyrachlor products to include cautions about toxicity to trees and runoff issues. Some restrictions and precautions listed on the Method® 50SG label are as follows (DuPont, 2012):

- *Do not apply this product in areas where the roots of desirable trees and/or shrubs may extend unless injury or loss can be tolerated. Root zone areas of desirable trees or vegetation are affected by local conditions and can extend well beyond the tree canopy.*
- *Do not apply this product if site-specific characteristics and conditions exist that could contribute to movement and unintended root zone exposure to desirable trees or vegetation unless injury or loss can be tolerated.*
- *During periods of intense rainfall, applications made to roadsides or other non-crop areas, to soils saturated with water, or soils through which rainfall will not readily penetrate may result in runoff and movement of Method 50SG. Do not apply Method 50SG when these conditions exist.*
- *Do not use on lawns, walks, driveways, tennis courts, or similar areas.*
- *Do not graze or feed forage, hay, or straw from treated areas to livestock.*
- *Do not use plant material treated with this product for mulch or compost.*
- *If non-crop sites treated with Method 50SG are to be converted to a food, feed, or fiber agricultural crop, or to a horticultural crop, do not plant the treated sites for at least one year after the Method 50SG application. A field bioassay must then be completed before planting the desired crop.*

- *Certain species may be sensitive to low levels of Method 50SG including but not limited to, conifers (such as Douglas fir, Norway spruce, ponderosa pine, and white pine), deciduous trees (such as aspen, Chinese tallow, cottonwood, honey locust, magnolia, poplar species, redbud, silver maple, and willow species), and ornamental shrubs (such as arborvitae, burning bush, crape myrtle, forsythia, hydrangea, ice plant, magnolia, purple plum, and yew).*

Case studies involving pyridine carboxylic acid persistence in compost

In Washington state during 2000, clopyralid contamination was reported at Washington State University (WSU), Spokane regional compost facilities, a yard trimmings compost facility in Cheney, and at a horse facility in Whitman County (Bezdicek et al., 2001). The Spokane regional composting facility processed 25,000 tons per year, and the clopyralid problem surfaced in 2000 with injuries to tomato plants. Samples of the affected compost, taken 9 months after the problem was discovered, showed clopyralid levels of 31-75 μg/kg. The contaminated compost had been in open windrows, and was changed to an ag-bag aerated windrow in an attempt to degrade the herbicide more quickly. Samples taken in March 2001 showed clopyralid levels were still at 57-67 μg/kg. In May 2001, the finished compost failed a bioassay test, and subsequent testing showed clopyralid levels at 73-83 μg/kg. In April 2001, the city of Spokane asked Dow to halt distribution of clopyralid in the area. Dow complied for residential use and made a flyer to distribute to landscaping companies telling of the problem. Spokane also educated the public about why grass treated with clopyralid can't be composted. A company named Norcal purchased the Spokane regional compost facility and worked with Dow to research a way to break down clopyralid in compost (Bezdicek et al., 2001).An article in Spokane's *Spokesman Review* updated readers about the Clopyralid contamination at the composting facility run by the city of Spokane (Brunt, 2012). The article states that $4 million worth of damages were incurred by the city, and the city had a confidential settlement with Dow in March of 2011. Tests by WSU showed crops were significantly affected by the contaminated compost the first year, but not the next year. Clopyralid is no longer allowed on residential lawns, but is allowed on golf courses and for agricultural use (Brunt, 2012).

The WSU compost facility produced 25,000 cubic yards of composted animal manure and bedding per year, and sold it to plant nurseries (Bezdicek et al., 2001). WSU compost was contaminated with picloram, which was traced back to a field sprayed with Tordon-101 and harvested for hay off-label. This hay was fed to WSU livestock, composted, sold to nurseries, and re-sold to homeowners who discovered plant-deforming symptoms on vegetable plants and shrubs. The affected vegetable plants included peas, beans, tomatoes, potatoes, and sunflowers. After this incident, WSU began analytical and bioassay tests for herbicides in their compost. In 2001, clopyralid contamination was found. Subsequently, WSU began certifying vendors of grass hay and straw (purchased for WSU livestock) to guarantee the products were free of herbicide contamination. WSU also took a proactive lead in the community by hosting educational meetings, creating a bioassay test program, and suggesting remediation strategies like frequent watering and planting cover crops to facilitate herbicide breakdown. WSU was not able to sell compost for two years following this ordeal, and the cost for loss of revenue, additional testing, and additional labor is estimated to have cost $250,000 (Bezdicek et al., 2001).

In Vermont during June 2012, Green Mountain Compost started receiving calls from customers reporting symptoms of persistent herbicide damage (Green Mountain Compost, 2012). Samples of compost were tested and found to contain aminopyralid, clopyralid, and picloram (USCC, 2013). The Vermont Department of Health analyzed lab test results and determined that the trace amounts of herbicides in the compost didn’t pose a risk to human health, and vegetables from gardens suspected of containing persistent herbicides were safe to eat (Green Mountain Compost, 2012). Green Mountain Compost has stopped all sales of compost products through the 2013 season and has notified customers and issued press releases. Green Mountain Compost is owned and operated by the Chittendon Solid Waste District, and they are working with the Vermont Agency of Agriculture to determine how the compost feedstocks were contaminated. Meanwhile, the Chittendon Solid Waste District has spent an estimated $270,000 settling customer complaints and retrieving unsold products, plus $372,000 has been spent on testing and legal assistance (USCC, 2013). Loss of sales through the 2013 season will likely bring the contamination cost above $792,000 (USCC, 2013).

The curious part of the current contamination is that any contaminated feedstocks would be in violation of herbicide label requirements and Vermont regulations (Green Mountain Compost, 2012). Clopyralid contamination was traced back to two horse farms that used Purina feed. Samples of Purina feed were sent to Anatek Labs in Moscow, Idaho to be tested. Anatek was chosen because it is one of the few labs that can test for clopyralid and picloram at levels below 10 ppb. Four of the samples contained clopyralid in levels ranging from 142 ppb to 465 ppb (Biocycle, 2012). A second lab confirmed the presence of clopyralid in Purina horse feed (Reading, 2012). The second lab was Carbon Dynamics, Dow's contracted lab, and they found low levels of clopyralid in five feed samples (Sullivan, 2013).

The presence of clopyralid in a national brand of horse feed focused the attention of the USEPA. Representatives of the USEPA met with officers and advisors of the U.S. Composting Council (USCC) on August 16, 2012 (USCC, 2012). The USEPA confirmed that they are working with Dow, DuPont, and the Chittendon Solid Waste District to review methods to analyze contamination and determine appropriate action. The USCC and USEPA are forming a task force to develop a method to pre-screen herbicides for fate-in-compost. This is timely because some of the persistent herbicides are up for re-registration with the EPA in 2014 (USCC, 2012).

Reports of persistent herbicides contaminating compost and injuring plants have come from North Carolina, Maryland, Virginia, California, Montana, Washington, Vermont, and Nebraska, but unfortunately most have been anecdotal and were not confirmed through testing. Currently, the majority of state labs do not have the expertise, time, or resources to measure trace levels of pyridine carboxylic acids in complicated matrices like manure and plant tissue (F.C. Michel, personal communication, 2013). In 2009-2010, Montana State University's Schutter Diagnostic Laboratory received over 112 garden samples showing symptoms of exposure to auxinic herbicides (Tharp, 2010). According to Dr. Michel (Ohio State University), there are several immediate research needs that need to be fulfilled before the scope of auxinic herbicide contamination can be determined. These research needs include systematic compost testing from facilities across the U.S., reliable plant bioassays that can be used by compost producers or testing labs to determine herbicide contamination, chemical test methods to be used by state labs to detect herbicides in composts at μg/kg detection limits, and microorganism inoculants that can be used to quickly degrade persistent herbicides in compost (F.C. Michel, personal communication, 2013).

Compost Quality

Improper composting methods could also produce compost that is phytotoxic to plants, and may show symptoms similar to plant damage by pyridine carboxylic acids. Compost quality must be taken into consideration when diagnosing possible pyridine carboxylic acid compost contamination. Composts are considered finished and are marketed and distributed between 30 and 300 days (Blewett et al., 2005). This wide time span may result in varying degrees of compost maturity, an attribute that describes both the degree of decomposition (i.e., stability) and the finished product's potential for phytotoxicity. While the USCC has established methods for assessing compost maturity (USCC, 2010), much of the compost marketed in the U.S. does not undergo any testing. With increasing awareness of "killer compost," problems with composting blamed on herbicide contamination could actually arise from the composting process itself. Salinity and maturity are significant factors in compost quality that may cause phytotoxicity symptoms. High salinity affects water relations of the plant, which may cause yield reduction, leaf burning, leaf deformation, and epinasty. Immature compost may contain volatile organic acids, fatty acids, phenolic acids, ammonia, or ethylene oxide in sufficient quantities to cause stunting and other phytotoxicity symptoms (Blewett et al., 2005). These symptoms may appear very similar to herbicide toxicity symptoms, especially to the untrained eye.

The lack of quality standards for compost means that immature compost is widely available and should be of concern to gardeners. Immature compost could be phytotoxic for several reasons: natural production of phytotoxic degradation intermediates like organic acids and phenolic acids, high salt concentration, ammonia build-up, and inadequate decomposition of phytotoxic xenobiotics like herbicides (Blewett et al., 2005).

The lack of regulated standards for compost as well as the lack of education of consumers is a concern, especially because consumers assume that compost has been sanitized and chemically stabilized. The State of New York has defined compost maturity as "the characteristics of a soil conditioning material that render it harmless to the plant grown when used as a topsoil or soil supplement" (Blewett et al., 2005). While it would be helpful to have national compost maturity regulations, implementation would most likely increase the cost to the composting industry (Blewett et al., 2005).The USCC and the USEPA should take the lack of composting regulations into consideration as they work together to eliminate incidences of persistent herbicide contamination in compost.

Bioassay methodology

Plant bioassays are useful for determining the presence of herbicides in soil and compost, and a variety of methods have been developed (Brinton et al., 2006). While bioassays are not as specific as chemical tests, they are much less expensive and can sometimes detect the activity of phytotoxic metabolites that are not detected in chemical tests. Care should be taken to use a bioassay method that accurately distinguishes plant damage from herbicide residues as compared to damage from other factors like salinity and pH (Brinton et al., 2006). Due to the potentially high salinity of compost, it is not a good idea to use 100% compost as the growing media for bioassays. A survey of 39 green composts in Washington state found a range of salinity (measured by electrical conductivity) between 0.4 and 24.2 dS m^{-1} (Brinton et al., 2006). Commonly accepted salinity levels for seedling growth media range between 1.99 and 3.5 dS m^{-1} (Brinton et al., 2006). Most all bioassay methods use various ratios of compost blended with another growing medium to ensure good seed germination.

Bioassay methods are listed for various crops on herbicide labels containing aminopyralid, aminocyclopyrachlor, clopyralid, and picloram. The WSU Whatcom County Extension has developed a protocol for gardeners that want to perform a bioassay test for the presence of auxinic herbicide residues. The protocol can be accessed at http://whatcom.wsu.edu/ag/aminopyralid/bioassay.html. In this protocol, published in 2011, pea plants are grown for several weeks until they have developed three sets of leaves. This bioassay explains that suspect compost should be mixed in a 2:1compost to topsoil ratio. This should eliminate the concern of plant damage from other compost factors described above. Pea plants are chosen because they are very sensitive to auxinic herbicides. The WSU protocol also includes pictures of mild and severe plant damage for reference.

Horse Ownership and Persistent Herbicides

Horse owners frequently buy hay from other farmers because they don't have the acreage and equipment to make their own hay. Horse owners are also notoriously picky about purchasing top-quality hay that is free from weeds, dust, and mold. Horse manure is often composted and given or sold to gardeners who use it to improve soil structure and nutrient content. All of the links of this common chain of hay supplier, horse owner, and gardener need to be educated about the potential transfer of pyridine carboxylic acids from the hay field or pasture to the garden. When horse owners inquire about hay quality, they need to remember to ask if any herbicides were sprayed on the hay. If herbicides were sprayed, find out if the active ingredients included aminocyclopyrachlor, aminopyralid, clopyralid, or picloram. Additionally, if horse owners hire someone to spray herbicides on their pastures and they plan to compost their manure, they need to communicate this to the herbicide applicator. While the labeling on herbicides is now clearer with respect to warnings about composting, the warnings are useless unless the person doing the composting reads them.

The recent discovery of clopyralid in Purina horse feed may significantly impact the options horse owners have for disposing of their manure (Reading, 2012). After two separate laboratories confirmed the presence of clopyralid in Purina feed purchased in Vermont, Holly Flemister from Land O'Lakes Purina Feed issued this response:

> *We have not independently verified the accuracy of these tests. Land O'Lakes Purina Feed has not confirmed the presence of any harmful pesticide residues in our horse feed, and our customers have not reported any related problems or complaints concerning their horses. The indicated levels of residues reported fall well within the tolerance levels set by the EPA for residues on various types of grains/commodities that are used in the animal feed formulations. Even though*

these residues at these levels are not harmful to the health of the horse, the existence of various chemicals in manure may cause the manure to interact with compost in a way that could be harmful to plants. Consumers having difficulty related to their composting issues should contact their local horticultural expert (Biocycle, 2012).

Horse owners should stay updated on the outcome of these discoveries in Vermont and the impact they may have on disposal of horse manure.

Another interesting issue of persistent herbicides in horse feed and manure involves the U.S. Forest Service's (USFS) requirement for certified noxious weed free hay and straw (NAISMA, 2013). As of 2009, only certified weed free hay and straw are supposed to be used in all USFS Wilderness areas in the U.S. All of the USFS lands in the Rocky Mountain and Intermountain regions require certified weed free hay. Many states in the western U.S. require certified weed free hay on all public lands including Bureau of Land Management lands, state parks, national parks, military reservations, and U.S. Fish and Wildlife areas (NAISMA, 2013). Unfortunately, the certification appears to vary from state to state, and it is difficult to find literature, details, and directions without calling the USFS office of the area being visited. The USFS hopes to prevent additional invasive plant seeds from being imported in horse hay and feed, but ironically might be importing persistent herbicides that could kill native broadleaf plants. The lack of clear details on the USFS weed-free certification program seems to indicate that these regulations are not taking the potential for pyridine herbicide persistence into account.

Media portrayal of "killer compost"

An article from *The Observer* in June 2008 portrays compost contaminated with aminopyralid as prevalent in the U.K. (Davies, 2008). The Royal Horticultural Society received more than 20 phone calls per week about grossly deformed vegetable crops. The most popular aminopyralid herbicide in the U.K. is ForeFront, manufactured by Dow. Colin Bowers, the UK Grassland Marketing Manager for Dow, stated "The chain is horrendously complicated. In the cases we have managed to trace back, we might find that the farmer who supplied the manure didn't spray anything himself, but he might have brought in a couple of bales of silage from one of his neighbours, and that farm might have sprayed." Shirley Murray, who owns an allotment near south-west London, said several of her neighbors had used the same manure bought from a stable and all were affected. She said "I am absolutely incensed at what has happened and find it scandalous that a weed killer sprayed more than one year ago, that has passed through an animal's gut, was kicked around on a stable floor, stored in a muck heap in a field, then on an allotment site and was finally dug into or mulched on to beds last winter is still killing "sensitive" crops and will continue to do so for the next year." Gardeners with injured plants and produce are being advised not to eat the produce by both Dow AgroSciences and by the Royal Horticultural Society (Davies, 2008).

Mother Earth News published short articles about "killer compost" in 2008, 2011, and 2013. The 2008 article begins by warning gardeners that straw, manure, and compost could kill their gardens thanks to the Dow chemical company (Pleasant and Long, 2008). *The Observer* article discussed above is mentioned, as well as a short discussion of clopyralid and aminopyralid. Dow's herbicides Confront, Milestone, and Forefront are singled out. The article also mentions that the half-life of aminopyralid can be up to 533 days, but that Milestone and Forefront were quickly approved by the EPA through the Reduced Risk Pesticide Initiative. The article ends by cautioning readers to be careful when buying compost or manure, and to spread the word about the problem (Pleasant and Long, 2008).

The April/May 2011 issue of Mother Earth News (Pleasant and Long, 2011a) provides details from different episodes of persistent herbicides harming field crops. The article mentions lab tests confirming aminopyralid toxicity from soil samples in 17 counties in Montana, reports of contamination and lost crops to state weed specialists in Pennsylvania and North Carolina, and the losses to gardens and farms following compost contamination in Whatcom County Washington. A North Carolina hay field treated with Milestone in 2006 is still unfit for tomatoes in 2009 according to the article. Pleasant and Long (2011a) also mention the risk to endangered native plants as well as legumes, which would "cripple nature's fertility cycle." The article does mention that contamination warnings now appear on the front of Dow's Milestone herbicide.

The October/November 2011 issue of Mother Earth News held an article about aminocyclopyrachlor, specifically discussing DuPont's Imprelis herbicide. Details of Imprelis registration with the EPA in August 2010, subsequent tree damage, and sales being halted in August 2011 are discussed. The main focus of this article is the failure of the Imprelis label to properly warn applicators of possible problems. Imprelis has a nine page label, and buried on page seven in a bulleted list is the warning to not compost grass clippings. The label does not say what could happen if clippings are composted. Concern about unrealistic, lengthy, and confusing instructions for applicators is discussed, especially the concept of the herbicide label being the law because it gives herbicide companies a liability release (Pleasant and Long, 2011b).

The February/March 2013 issue of Mother Earth News has a letter in the "Dear Mother" section as well as an article about killer compost. The letter is from David Goodman, and he details his personal experience purchasing a load of composted cow manure that caused vegetable plants to have "bizarre growth or no growth at all." Goodman looked up "manure, distorted growth, and leaf curl" on the internet to find information about aminopyralid. When he confronted the farmer who sold him the manure, the farmer confirmed that he had sprayed his cow field with Grazon, which contains aminopyralid. The farmer said he had no idea that

Grazon would go through the cow and ruin the manure. The farmer refunded the $60 Goodman had spent on the manure, but Goodman still lost about $1000 worth of plants (Goodman, 2013). In the same issue, Dan Sullivan details the recent issues of herbicide damage at Green Mountain Compost in Williston, Vermont (Sullivan, 2013). In this situation, contamination was traced by Anatek Laboratories back to Purina horse feeds contaminated with clopyralid at 142-465 ppb. Dow sent additional samples to their contracted lab, Carbon Dynamics, and found low levels of clopyralid in five samples of commercial livestock feed. Sullivan takes a strong position in the article, stating:

> *The solution is simple - stop allowing companies to profit by selling these plant-killers while hiding behind "do not compost" warnings buried in nine-page labels. Our guess is that while the [U.S.] Composting Council agrees with us, it doesn't want the public to know about this issue because then we might stop buying its products. Well, as much as it pains us to say it, we believe the time has come for the public to stop buying compost or manure products unless they come from suppliers that are able to afford testing and can screen feedstocks for herbicide residues. A few companies have told us this is the only way they know of to deliver safe compost to their customers, but the added costs for testing make competing tough.*

Conclusions

Composting is one of the best methods available for reducing the volume of organic material destined for landfills. In fact, at many landfills yard trimmings are banned. Schools, environmental organizations, and local governments have invested time and money to educate citizens about the value of composting. Not only does composting reduce the waste stream headed to landfills, but it also creates a naturally produced and beneficial soil amendment for use on vegetable gardens and landscape plantings. Persistent herbicides have the potential to destroy public confidence in composting at a time when there is increased interest in backyard gardening using organic methods.

The companies that manufacture herbicides containing aminocyclopyrachlor, aminopyralid, picloram, and clopyralid have revised the warning labels on their products to include cautions about the persistence of these chemicals in soil, compost, hay, and manure. As of now, the benefits of these herbicides (excellent broadleaf weed control and low toxicity to mammals) appear to outweigh the risks. However, these risks are still significant. Relying on herbicide label restrictions is not adequate because there is often a disconnect between the herbicide applicator and the person collecting materials for composting.

Extension agents, Soil and Water Conservation District employees, and herbicide company representatives need to teach farmers and homeowners why they should be so careful with pyridine carboxylic acid herbicides, rather than just preaching that "the label is the law." Herbicide labels are filled with "dos and don'ts," and it is human nature to disregard rules that don't seem to have merit. If people understood why those label restrictions were in place, then they would be more likely to follow the label directions. It is also likely that many herbicide applicators do not read the label restrictions, either because they don't want to bother or because the label gets torn, dirty, or misplaced. Some herbicide applicators may not be able to read the labels due to the confusing language and scientific terms.

Additional education about auxinic herbicides should be added to the VDACS pesticide applicator certification course. As of February 2013, the test for a private pesticide applicator certificate has not been revised since 1995. Because the test was created prior to pyridine herbicide persistence issues, there are no questions about this on the test. The best recommendation for pyridine carboxylic acids should be to only use them on pastures that are owned by the herbicide applicator and that will remain pasture for many years after the herbicide is applied. Private pesticide applicators are only required to keep records for two years. Persistence in soil, compost, or stored hay could outlast this recordkeeping requirement.

Compost contamination from pyridine carboxylic acid herbicides may be prevented with education and improved communication. Herbicide applicators must not forget to document the chemicals they use and share this information with everyone who will come in contact with the treated grass for the next two years. Anyone purchasing hay must remember to ask what the hay was treated with, and composters must give extra thought to the chain-of-custody of their feedstock materials. If unknown feedstocks are added to compost, a bioassay should be performed prior to using the compost. For homeowners, the best thing to do with grass clippings is to grass-cycle by letting the clippings fall back onto the lawn. This not only prevents movement of herbicides and pesticides, but also nourishes the lawn as the clippings decompose. It is apparent that current herbicide label restrictions are not working, so revisions of EPA-registered uses may be necessary. Now that the issues of pyridine carboxylic acid herbicide persistence in "killer compost" are known, changes need to implemented along the supply chain in order to prevent further damage to garden plants, composting businesses, and consumer confidence.

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