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Manuka Oil, A Natural Herbicide with Preemergence Activity

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Natural herbicides approved in organic agriculture are primarily nonselective burn-down essential oils applied POST. Multiple applications are often required due to their low efficacy. To address this problem, the in vivo herbicidal activity of manuka oil, the essential oil distilled from manuka tree (*Leptospermum scoparium* J.R. and G. Forst), was tested on selected broadleaf and grass weeds. While manuka oil exhibited good POST activity when applied in combination with a commercial lemongrass oil–based herbicide, it ultimately demonstrated interesting PRE activity, providing control of large crabgrass seedlings at a rate of 3 L ha^{-1}. Manuka oil and its main active ingredient, leptospermone, were stable in soil for up to 7 d and had half-lives of 18 and 15 d, respectively. The systemic activity of manuka oil addresses many of the current limitations associated with natural herbicides. Additionally, its soil persistence opens up a multitude of new possibilities for the use of manuka oil as a tool for weed management and may be a potential bridge between traditional and organic agriculture.

Nomenclature: Leptospermone; 2,2,4,4-tetramethyl-6-(3-methyl-1-oxobutyl)-1,3,5-cyclohexanetrione, CAS 567-75-9; large crabgrass; *Digitaria sanguinalis* (L.) Scop. DIGSA.

Key words: Natural product, triketone, phytotoxins, herbicide, mode of action, *p*-hydroxyphenylpyruvate dioxygenase, essential oil, organic agriculture.

Weeds have a greater negative impact on crop yields than any other agricultural pests (Oerke 2006). As a result, conventional agricultural practices rely on highly effective synthetic herbicides for managing weeds, and these compounds account for more than half of the volume of all agricultural pesticides applied in the developed world. Public sentiment toward synthetic herbicides has grown increasingly negative, and new pesticide regulations (i.e., Food Quality Protection Act, 1996) have contributed to a reduction of the number of new products commercialized by the major agrochemical manufacturers (Gerwick 2010). This has had a particularly negative impact on minor crop producers (Gast 2008).

Demand for organic food, on the other hand, has grown tremendously throughout the developed world (Dimitri and Oberholtzer 2009). Organic farming does not permit the use of synthetic pesticides (EPA 2011). Natural alternatives that are currently approved for organic agriculture are mostly nonselective essential oils used as POST, burn-down products. Their low efficacy requires multiple applications of high amounts to achieve good weed control (Young 2004). The process is expensive both in terms of the cost of the material applied and the cost of the applications. There is, therefore, a need for new natural weed management tools.

Two of the major limitations of natural herbicides approved for organic use are their lack of systemic activity and their nonspecific mechanisms of action (Dayan and Duke 2010). In an effort to develop new natural herbicides with superior properties, we recently reported that several natural products including the natural β-triketones present in manuka oil have the same molecular target site as the commercial synthetic herbicides sulcotrione and mesotrione, namely the enzyme *p*-hydroxyphenylpyruvate dioxygenase (HPPD) (Dayan et al. 2007; Meazza et al. 2002; Romagni et al. 2000).

HPPD is a key enzyme in the biosynthesis of tocochromanols (e.g., tocopherols and tocotrienols) and prenyl quinones (e.g., plastoquinone). Since plastoquinone is a cofactor necessary for phytoene desaturase function (Norris et al. 1995), HPPD inhibitors indirectly cause a deleterious reduction in the levels of carotenoids in plants. Subsequently, the reduced pool of carotenoids is not sufficient to quench the excess electrons generated during photosynthesis, causing a rapid degradation of chlorophylls (photodynamic bleaching) (Pallett et al. 1998).

While a structure–activity relationship study demonstrated that the length of the aliphatic side chain of the natural triketones of manuka oil modulated the activity of the compounds (Dayan et al. 2009b), the efficacy of β-triketone–rich manuka oil as a herbicide has not been proven, much less optimized. Hence, it was hypothesized that this essential oil may have in vivo herbicidal activity. Therefore, manuka oil was tested on selected broadleaf and grass weeds, and its effects on chlorophyll, carotenoids, and biomass were measured to determine its PRE and POST activity. The soil stability and bioavailability of manuka oil and its primary component leptospermone (Figure 1) were studied to understand the basis for the PRE activity of this oil.

Materials and Methods

Chemical Supplies. Manuka oil was obtained from Clean & Green Trading Co.[1] The nonionic sticker-spreader Nu-Film P (96% poly-1-*p*-menthene) approved for organic farming was purchased from Miller Chemical & Fertilizer Corporation[2] and the lemongrass oil natural herbicide GreenMatch EX™ (lemongrass oil [50%], CAS 8007-02-1, and a mixture of water, corn oil, glycerol esters, potassium oleate and lecithin) was obtained from Marrone Bio Innovations.[3] All other chemical supplies were purchased from Sigma-Aldrich.[4]

Synthesis of Leptospermone. Dry phloroglucinol (10–12 mM) was added to a solution of phosphorus oxychloride (15 ml) and anhydrous aluminum chloride (4 g) and stirred

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