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## **A Review on the Mode of Action and Current Use of Petroleum Distilled Spray Oils**

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## **1. Introduction**

Petroleum based mineral oils have been used for insect pest control for over a century (Agnello, 2002). However, their use is as current today as it was before the advent of chemical insecticides, given that they are compatible with modern sustainable management practices. These products pose a number of advantages over conventional pesticides and they have very low mammalian toxicity, low residual activity, they have never been associated with development of insect resistance, and are less disruptive to natural enemies than broad spectrum insecticides (Beattie and Smith, 1993). Continuous studies on the efficacy and chemistry of petroleum based oils over the last sixty years, led to identification of the main factors related to their insecticidal activity as well as their phytotoxicity and allowed for the development of more refined and effective spray oils (Agnello, 2002). The efficacy of isoparaffinic petroleum distilled spray oils (PDSOs) typically increases as the molecular weight of their constituent oil molecules increases, but so does the risk of PDSO induced phytotoxicity (Riehl, 1969), which has been one of the main hindrances to the use of these products. Modern PDOs are highly refined, linear molecules with a range between 21 and 24 carbons, to combine good insecticidal efficacy with low phytoxicity. The use of UV additives (e.g. sunscreens) to reduce the detrimental effect of the ultraviolet light on the breakdown of oil molecules has reduced the potential of some PDSOs to damage plants (Hodgkinson, 1999; Hodgkinson *et al.*, 2002). Thus, once limited to early season or dormant sprays to avoid oil injury to green plant tissue, newer narrow-range PDSOs are being reconsidered and assessed for incorporation into integrated pest management programs. PDSOs have been found to be effective against numerous orchard pests including scales and

mites (Beattie *et al.,* 1995; Beattie, 1990; Beattie and Smith, 1993), whiteflies (Larew and Locke, 1990; Liang and Liu, 2002), aphids (Najar-Rodríguez *et al.,* 2007), psylla (Zwick and Westigard, 1978; Weissling *et al.,* 1997), and fruit-feeding Lepidoptera (Davidson *et al.,* 1991; Al Dabel *et al.,* 2008). In apple orchards, the interest in PDSOs as part of integrated pest management programs has increased in the past years, particularly for the control of secondary pests (Fernandez *et al.,* 2005). This is in part due to better PDSOs formulations, but it also arises from a decline in the use of broad spectrum insecticides due to stricter regulations and to the widespread use of mating disruption (Fernandez *et al.,* 2005). Recent studies have also

suggested new uses of PDSOs against a wider range of pests, such as the European corn borer, *Ostrinia nubilalis* Hubner (Lepidoptera: Pyralidae) in maize (Mensah *et al.,* 2005a, 2005b; Al Dabel *et al.,* 2008), *Helicoverpa spp*. in cotton (Mensah *et al.,* 2002, 2001), and the obliquebanded leafroller *Choristoneura rosaceana* (Harris) (Lepidoptera: Tortricidae) in orchards (Wins-Purdy *et al.,* 2009).

When overdosed with PDSOs, small insects die rapidly (Najar-Rodríguez *et al.,* 2007) while large insects are more tolerant and toxicity is often unpredictable (Zerba *et al.,* 2002; Mensah *et al.,* 2005b). As opposed to other synthetic insecticides, oils address multiple targets and do not bind to specific receptors since their toxic effects depend on the interaction between their physical and chemical properties and the anatomical, developmental, physiological and behavioural traits of the target insect. Smith (1952) summarized most of the theories regarding the mode of action of oils, which he categorized according to whether they were applied to eggs or motile forms. He proposed that when used as ovicides, they acted by preventing the normal exchange of gases through the outer coating, prevented hatching by hardening the outer covering, interfering with water balance of the egg, softened or dissolved the outer covering of the egg, penetrated the egg causing coagulation of the protoplasm, or interfered with enzyme or hormone activity. When used as insecticides, oils were thought to cause suffocation by blocking spiracles, to penetrate the tissue in the liquid phase and to 'corrode' it by breaking down tissue structure, and to contain toxic volatile components that act as fumigants. Although some of these hypotheses were tested, most of them were speculative and were not verified. More than fifty years have passed since Smith's (1952) review on action mechanisms of petroleum oils. During this time there have been remarkable advances on petroleum technology, as well as on scientific studies testing their efficacy and potential uses on a wide array of insect species, which have broadened the boundaries for the use of PDSOs. Therefore, we review the literature on the use and effect of PDSOs on various insect taxa and crops, and discuss the action mechanisms identified.

## **2. Mode of action and target sites of PDSOs**

The modes of action of toxic chemicals are traditionally divided into two main categories: baseline toxicity or narcosis, and specific mode of action (Rand *et al.,* 1995). Narcosis can be broadly defined as a state of arrested activity of protoplasmic structures caused by a wide variety of organic chemicals with non-specific modes of toxic action (Veith *et al.,* 1983; Veith and Broderius, 1990), due to a physical action of the molecule and not to a chemical reaction (Ferguson, 1939). Narcosis is believed to be the result of reversible and non-specific disturbance of membrane integrity and function resulting from the partitioning of a given chemical into biological membranes (Escher and Hermens, 2002). Thus, the potency of a baseline toxicant is expected to correlate with its affinity to the cell membrane (Gunatilleka and Poole, 1999). On the other hand, specific toxicity refers to chemicals that interact with or disrupt the function of a defined receptor site (e.g. compounds acting as oxidative phosphorylation uncouplers, respiratory inhibitors, electrophiles, acetylcholinesterase inhibitors, and central nervous system seizure agents). Due to the non specific nature of narcosis, chemicals that meet structural requirements of specific modes of action (e.g. synthetic pesticides) should be excluded from narcosis.The non-specific nature of narcosis and chemicals that meet structural requirements of specific modes of action (e.g. synthetic pesticides) should be excluded from narcosis. However, most substances that are narcotics can be shown to be capable of producing both narcosis and toxicity, depending upon the

concentration used in the case of chemical agents and upon some measure of intensity in the case of physical agents (Mullins, 1954). Additionally, there are numerous mechanisms of narcosis as shown by the great variety of symptoms caused by (Veith and Broderius, 1990). For example, baseline toxicity can be categorized into polar and nonpolar narcosis based on the chemical structure and degree of toxicity of the xenobiotic (Russom *et al.,* 1997). Nonpolar narcosis results from hydrophobic bonding of the chemical to enzymes and/or membranes and polar narcosis may result from the presence of strong hydrogen binding group on the molecule (Veith and Broderius, 1990). However, more recent research proposes that there is no difference in membrane concentrations of the polar or non-polar chemicals, and thus equal intrinsic toxic potency is encountered for these two types (Escher and Schwarzenbach, 2002). As opposed to traditional synthetic insecticides, oils target multiple sites and do not interact with specific receptors since their toxic effects or narcosis, depend on the interaction

between their physical and chemical properties and those of the insect. Oils show affinity to the insect body surface and penetrate the insect cuticle (Stadler and Buteler, 2009), dissolve internal lipids (Taverner *et al.,* 1999) and eventually penetrate internal cell structures (Taverner *et al.,* 2001; Taverner, 2002) (Table 1).

#### **2.1 Effects on insect eggs**

Smith and Pearce (1948) studied the respiratory effects of oils on eggs of the oriental fruit moth, *Grapholita molesta* (Busck) (Lepidoptera: Tortricidae) and found them to be responsible for decreased respiration rate, presumably through mechanical interference with normal gaseous exchange. They further concluded that the less reactive paraffinic oils showed greater ovicidal efficacy than did the more reactive unsaturated oils. A recent study by Al Dabel *et al.* (2008) showed a strong ovicidal effect of nC24 and nC27 PDSOs on *O. nubilalis* egg masses when applied at  $3 - 10\%$  (v/v). The PDSO treatments seemed to stop the embryonic development and killed the embryo in the eggs but the mechanism involved in the ovicidal action remains to be studied. Topical application of 2% Purespray Green Horticultural mineral oil (Petro-Canada) also led to almost complete egg mortality in the obliquebanded leafroller, *Choristoneura rosaceana* (Lepidoptera: Tortricidae) (Harris) through both contact toxicity and suffocation (Wins-Purdy *et al.,* 2009).

#### **2.2 Effects on insect larvae and adults 2.2.1 Spiracle or tracheal blockage**

Insect suffocation by spiracle blockage is usually held as the most accepted theory on the mode of action of mineral oils (Johnson, 1994). The tracheal inflow of oil was reported for the first time by Moore and Graham (1918), and addressed after that by several authors (Roy et al., 1943; Stadler et al., 1996; Taverner et al., 2001). Stadler et al. (1996) found evidence of the inflow of PDSOs to the trachea of Lepidoptera larvae (*Anticarsia gemmatalis* Hub. (Lepidoptera: Noctuidae) by looking at the air-liquid interface inside the tracheae and tracheolar tubes. In *Blatella germanica* L. (Blattodea: Blattellidae), oils appeared to induce mortality due to asphyxia by occlusion of tracheae and tracheoles (Stadler et al., 1996). No chronic damage was observed on *B. germanica* treated with sub-lethal doses of mineral or vegetable oils. Tracheal blockage by PDSOs was observed in living as well as dead insects, showing that the phenomenon is independent of insect metabolism and that it can be described by the Poiseuille equation (Tschapeck, 1961). By this equation, capillary pressure depends mainly on the viscosity of the oil, as well as on the radius of the cylinder (i.e. trachea). Taverner et al. (2001), observed that Ampol Citrus Postharvest Dip (CDP), an



Table 1. Summary of effects of spray oils observed in insects, and probable causes.

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Table 1 (Continued). Summary of effects of spray oils observed in insects, and probable causes.

emulsified C15 alkane used by Australian citrus packers to control surface pests, penetrates the tracheoles of lightbrown apple moth, *Epiphyas postvittana* Walker, (Lepidoptera: Tortricidae). Confocal microscopy showed that if larvae were dipped in oil and then exposed to the air, the oil penetrated the tracheal system, but the extent of penetration varied with the type of oil. An oil with a carbon number of 15 (CPD) was observed to penetrate deeper into the tracheal system than a narrow-range oil with a carbon number of 23 (Ampol D-C-Tron NR), presumably due to a lower interfacial tension between CPD and the tracheal lining. Figure 1 illustrates the tracheal blockage occurring when crickets (Orthoptera:Grillidae) are dipped in oil. Spiracle blockage has also been observed and documented when using other products with similar physical characteristics as insecticide oils. Richling and Böckeler (2008) observed a similar phenomenon when treating *Pediculus humanus* Haeckel, (Phthiraptera: Pediculidae) and *Acheta domestica* (Orthoptera: Gryllidae) with low viscosity silicone. When the insect was immersed in or coated with silicone, the fluid entered all spiracles equally and systematically flowed through the tracheal system and completely filled the trachea in the insect's head in less than one minute. Results from Burgess (2009) bioassays using low viscosity silicones on head lice *P. humanus* show that the most likely mode of action of these substances, when applied in great amounts, is the physical blockage of the outermost sections of the insect respiratory system. Contrary to the widespread opinion that physically acting pediculicides work by suffocation, Burgess (2009) concluded that spiracle blockage causes inhibition of water excretion and further physiological stress, which leads to death either through disruption of internal organs or due to prolonged immobilization.



Fig. 1. Micrograph of tracheal branches from a cricket (Orthoptera:Grillidae) (untreated) [black background - left] and dipped in PDSO nC24 stained with aniline blue (lipophilic) [white background - right].

On the other hand, Najar-Rodriguez *et al.,* (2008) found that oil did not accumulate around the spiracular openings, inside the main trachea associated with the spiracles or in the small tracheoles associated with the gut and nerve ganglia in adult cotton aphids (*Aphis gossypii*  Glover, (Hemiptera: Aphididae)) and cluster caterpillars (*Spodoptera litura* Fabricius (Lepidoptera: Noctuidae)). Unlike some of the other studies in which the insects were dipped in the oil, these authors applied topically only a small amount of a PDSO nC24, at a maximum concentration of 20μl.

Based on these results it appears that the extent of spiracle and tracheal blockage and toxicity depend on the relationship between physical properties of the oil such as its n-paraffin carbon and its viscosity, and the dimensions of insect tracheae. Regardless, this phenomenon is not as significant for insect pest control due the low affinity (wettability) of oil and the internal wall of insect trachea, which leads to a low penetration rate of the oil. Oil will pour through the spiracular valves into the atrium and flow into the trachea as long as spiracles are open and the insects are overdosed (i.e. dipped in oil) (Stadler and Buteler, 2009). In contrast, when small volumes of PDSO contact the insect body (i.e. when the insect moves through a treated substrate), the amount of oil reaching the spiracular valve will not flow into the trachea because of the low surface energy between oil and inner tracheal surface (Stadler and Buteler, 2009).

#### **2.2.2 Effect on the integument**

Symptoms observed on the integument after topical treatment with sub-lethal doses of oils include cell membrane disruption and darkening (Van Overbeek and Blondeau, 1954; Stadler et al., 1996; Najar-Rodríguez et al., 2007). Given their lipophilic nature, PDSOs accumulate in cell membranes and thus affect their structural and functional properties (Mazella et al. 2005). As shown by Najar-Rodríguez et al. (2007) in vitro, oils are able to penetrate the cell membranes, accumulate inside the cytoplasm and cause cell dehydration and DNA condensation inside the nucleus. Furthermore, findings of teratogenic effects to the insect epidermis and aberrant molts have been observed after topical application of oils at the site

where the oil had been applied (Stadler et al. 1996). However, the authors tested unrefined spray oils, containing residues of naftalenic and sulfonable compounds which could have been responsible for the "burn effects" on the insect integument.

Insects may lose water through respiration, excretion, secretion and transpiration through the cuticle. Data from inter-specific comparative studies reviewed by Chown (2002) showed that in 16 insect species belonging to diverse taxa (Blattodea, Orthoptera, Coleoptera and Hymenoptera), transpiration through the cuticle averaged 91.5% and constitutes the main cause of total water loss in insects. Thus, insect resistance to desiccation depends mainly on their epicuticular waterproof wax layer (Hadley, 1994). The insect cuticle is a complex passive barrier to evaporative water loss composed of a mixture of long-chain compounds which include hydrocarbons (saturated, unsaturated, branched), wax esters, free fatty acids, alcohols, ketones, aldehydes, and cyclic compounds, which may be present in very complex mixtures (Dekker *et al.,* 2000). The quali- and quantitative arrangements of waxes on the cuticular surface are specific to each species in adaptation to its environment (Hadley, 1981) and its waterproof effectiveness is greater when they are in a solid rather than fluid state. Damage to the structural integrity of cuticular waxes or its removal by submersion in organic solvents (ether, chloroform, etc.) in insects (Hurst, 1940; Wigglesworth, 1941, 1942) and in millipedes (Cloudsley-Thompson, 1950), may lead to dehydration. Likewise, non-polar substances of low dielectric constant such as PDSOs may come into a competing equilibrium with some of the components of the cuticle wax layer, disrupting its continuity by interacting with the overall lipid mixture (Wigglesworth, 1945). This interaction leads to a decrease in the cuticle's wax layer melting point, with either a broad melting point range or a narrow melting point at the eutectic temperature (Hägg, 1969). This in turn leads to changes in cuticle permeability and dehydration.

Whereas some non-polar hydrocarbons (e.g. ether, chloroform) cause a complete removal of the cuticle wax layer, as well as hardening and stiffening of the cuticle (Hayes and Smith, 1994; Barbakadze, 2005), mineral and vegetable oils can attain a competing equilibrium with some of the components of the insect cuticle wax layer and soften the cuticle. This was proposed by Stadler *et al.* (2002), who found that mineral oils caused a softening of the cuticle in adult cotton boll weevils *Anthonomus grandis* Boh. (Coleoptera: Curculionidae). The variation in cuticle hardness observed, suggests that oils induce structural changes in the cuticle. The authors also found a correlation between cuticle softening and oil toxicity in laboratory bioassays, where a greater softening was associated with increased mortality.

#### **2.2.3 Effect on insect behaviour**

Oils have a repellent effect that discourage egg deposition and feeding. The residual film may inhibit insects from attaching to plant surfaces (Trammel, 1965). Also, it should be noted that "arrested activity" in insects is one recurrent symptom caused by PDSOs that has been reported directly or indirectly by many authors in laboratory toxicity tests (Xie and Isman, 1995; Stadler et al., 1996; Taverner, 2001; Najar-Rodríguez et al., 2007; Najar-Rodríguez et al., 2008).

Antifeedant properties of oils and starvation through deterrence have also been documented (Baxendale and Johnson, 1988; Najar-Rodríguez *et al.,* 2007; Beattie *et al.,* 1995). The deterrent effect of oil residues on oviposition has been observed in the citrus leafminer *Phyllocnistis citrella* Stainton (Lepidoptera: Gracillariidae) (Beattie *et al.,* 1995; Rae *et al.,* 1996), codling moth (L.) (Lepidoptera: Tortricidae) (Riedl *et al.,* 1995), white apple leafhopper (Hemiptera: Cicadellidae) (Fernandez *et al.,* 2001), the pear psylla *Cacopsylla pyricola* (Homoptera: Psyllidae) Foerster (Zwick and Westigard, 1978; Weissling *et al.,* 1997), and whiteflies (Larew and Locke, 1990; Larew, 1988; Liang and Liu, 2002). Studies by Mensah *et al.* (2001, 2002) have also shown that application of 2% (v/v) rate of Canopy® oil (nC27) to cotton and 4-5% (v/v) Texaco® oil (nC24) to maize plants can reduce oviposition of *Helicoverpa spp*. and *O. nubilalis* on cotton and maize plants, respectively. Liu *et al*. (2001) demonstrated that efficacy of PDSOs as deterrents is also related to molecular weight increase, as reflected by *n*C*y* values, and, therefore, to the persistence of oil molecules on sprayed surfaces.

#### **2.2.4 Effect on the nervous system**

Najar-Rodríguez *et al.* (2008) proposed that once the PDSOs penetrate the cuticle, they diffuse and accumulate within lipid-containing tissues, primarily the fat bodies. Results obtained by Schal *et al.* (2001) with *Musca domestica* (L.) (Diptera: Muscidae) suggest that lipophorin is involved in an active mechanism that selectively transports hydrocarbons from the haemolymph to individual tissues. These specific lipoproteic complexes are capable of sequestering hydrophobic core lipids from the hydrophilic environment of the haemolymph (Blacklock and Ryan, 1993) and of transporting lipids and hydrocarbons within the insect body (Schal *et al.,* 2001), which finally accumulate in lipid-containing tissues. Taverner *et al.* (2001) showed that treatment with Citrus Postharvest Dip (Ampol Research and Development Laboratories, Brisbane, Queensland), a formulated C15 alkane, affected neuron lipid membranes in *Epiphyas postvittana* Walker, (Lepidoptera: Tortricidae). Electrophisiology recordings showed that the alkane induced a rapid onset of multiple nerve firing in peripheral nerves of *E. postvittana* larvae. These authors suggested that nerve disruption was due to the displacement of protective neural lipids by solvent action of the alkane, affecting nerve activity by increasing membrane permeability to ion exchange. In another study Najar-Rodríguez *et al.* (2008) reported disruption of the synaptic transmission of nerve ganglia in *S. litura* treated with PDOs at concentrations equivalent to 0.1% v/v.

The effect of the absorption of hydrocarbons into phospholipid membranes is not clear, but is probably not site-specific, given the lack of any apparent structural complexity or stereoisometry of PDSOs (Najar-Rodríguez *et al.,* 2007; Taverner *et al.,* 2001). Thus, nervous disruption by PDSOs would not involve specific chemical binding to receptors or the active sites of enzymes, as is the case with traditional insecticides.

## **3. Discussion**

Almost sixty years have gone by since the last review on the use of mineral oils for pest management. Since then over 50 scientific manuscripts and a book were published (Beattie *et al.,* 2002), as well as an international conference (Spray Oils Beyond 2000, Sydney Australia) on the subject of petroleum derived mineral oils in pest management. Oils have a long history of effective use on fruit trees, particularly in dormant sprays on fruit crops, with a good performance in those cases where the pest is small in size and restricted to a small area during its lifecycle (e.g adults and crawlers of scale insects, aphids, phytophagous mites and their eggs, and nymphs of pear psylla, nymphs of grape leafhopper and eggs of codling moth) (Davidson *et al.,* 1991; Northover and Timmer, 2002). Currently, spray oils are recommended to manage scales, Psylla sp., and leaf miners in some systems (Table 2), as well as mites (Agnello *et al.,* 1994; Girantet *et al.,* 1997; Nicetic *et al,.* 2001). Moreover, the recent studies reporting the effectiveness of PDSOs against other insect pests show the relevance of petroleum derived oils as a current topic in pest management (Table 2). These studies demonstrate that the mode of action of mineral oils is more complicated than it was originally thought, and that suffocation, which was the most accepted theory, may occur

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Table 2. Recent and most relevant attempts to use spray oils in insect pest management.



Table 2 (continued). Recent and most relevant attempts to use spray oils in insect pest management.

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Table 2 (continued). Recent and most relevant attempts to use spray oils in insect pest management.

only in particular cases of overdosing (Table 1). The paradigms describing the mechanism of action of PDSOs have been developed from the works of Smith and Pearce (1948), Van Overbeek and Blondeau (1954), Stadler *et al.* (2002), Taverner *et al.* (2001), Najar-Rodríguez *et al.* (2008) and Stadler and Buteler (2009), who provided evidence and description of the multiple target sites involved in PDSOs toxicity. These include the integument, nervous system, respiratory system and insect behavior.

As opposed to other synthetic insecticides, oils address multiple targets and their effects on the cuticle waxes, cuticle softening, epidermal teratogenicity, tracheal blockage, receptor coating, deterrence and neurotoxicity of spray-oils, are all concurrent phenomena once the oil contacts the insect body surface. The sum of these phenomena plays a leading role in the lethal effect of the oil. Hence, all of these factors can influence the kinetics of a compound and each one occurs with a different intensity on the action sites, depending on the insect species, its developmental stage, the oil type and dose.

It can be concluded that isoparaffinic spray-oils do not interact with specific receptors, showing a non polar narcosis in insects (non-specific mode of action), characterized by progressive lethargy and death without any specific sustained symptoms. Other insecticide oils, containing aromatic residues, would shift the syndrome to polar narcosis. PDSOs are

capable of coating the exterior as well as reaching the interior of the insect, targeting different structures and organs, depending on the oils physical and chemical properties and the insect species and physiology. The effects of PDSOs are variable as well, depending on the site reached, the dose and the affinity between the oil and the target site. Therefore, it is extremely difficult to determine the exact cause of insect death. Future research should explore structure-toxicity relationships for each oil type, and standardize assessment methodology and experimental design. Ideally future studies will provide comparable results across insect taxa and oil types providing a guide of recommended practices for PDSOs use against different pests that would also direct further observations and experimentation.

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This book brings together issues on pesticides and biopesticides use with the related subjects of pesticides management and sustainable development. It contains 24 chapters organized in three sections. The first book section supplies an overview on the current use of pesticides, on the regulatory status, on the levels of contamination, on the pesticides management options, and on some techniques of pesticides application, reporting data collected from all over the world. Second section is devoted to the advances in the evolving field of biopesticides, providing actual information on the regulation of the plant protection products from natural origin in the European Union. It reports data associated with the application of neem pesticides, wood pyrolysis liquids and bacillus-based products. The third book section covers various aspects of pesticides management practices in concert with pesticides degradation and contaminated sites remediation technologies, supporting the environmental sustainability.

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