A review of spray generation, delivery to the target and how adjuvants influence the process

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Summary

The addition of an adjuvant can influence the processes of spray formation, droplet transport, impaction, retention and coverage of spray on the target surface. Methods of producing agricultural sprays are reviewed and methods of measuring and classifying such sprays are identified. These classifications provide useful information relating to spray behaviour but nozzle performance can be substantially influenced by adjuvants. Studies have shown that the mode of spray formation from flat fan hydraulic pressure nozzles can be changed substantially, influencing droplet size, velocities and the volume distribution pattern obtained. Anti-evaporants influence spray behaviour during transport of spray between the generator and targets. Behaviour at the target surface in terms of impaction, retention, coverage and uptake can all be modified. To date there is no clear definition of all of the effects of physical parameters of the spray liquid on behaviour. It is concluded that substantial progress has been made in the development of methods for measuring and predicting spray formation and behaviour, but that there is a need to understand how the addition of adjuvants can influence all of the physical processes involved, as well as the chemical and biological effects. Such information will enable adjuvants to be used to improve spray application in a safe and effective

Introduction

The use of adjuvants to enhance the efficacy of pesticide application is widespread and increasing. These have the potential to influence every aspect of the pesticide application, i.e. spray formation, droplet transport and interaction with the target. Adjuvants are predominantly surfactants which can improve retention, wetting, dispersion, emulsification, solubilization and bio-enhancement (Knowles 1995); mineral or vegetable oils which can reduce drift and wash-off and improve wetting, penetration and translocation (Rogiers 1995) or other substances such as polymers (Akesson et al. 1994). It has been recognised that if adjuvant use is to be fully optimized then the whole process of application, uptake and mode of action of pesticides must be considered - not the constituent parts in isolation (Hall et al. 1993).

This paper considers the methods commonly used to generate agricultural sprays for pesticide application, the mechanisms of spray formation and the techniques for quantifying sprays. Also addressed are the transport processes influencing droplet trajectories between the generator and target surface, the behaviour of droplets impacting on a surface and the role that adjuvants may have on all of the components of this complex process.

Spray generation systems

Hydraulic pressure nozzles

One of the most common ways to form a spray for the application of agricultural pesticides is to use the hydraulic pressure nozzle. In its simplest form, the nozzle consists of a shaped orifice through which liquid is forced under pressure to emerge as a liquid sheet which then breaks up into droplets by three defined modes of action. Flat fan nozzle designs are attractive because, when mounted on a boom structure to give a defined degree of pattern overlap, the resulting distribution of the volume of spray liquid at target level is relatively uniform. Many such nozzles are formed by creating a vee-shaped slot into a blind hemispherical (or approximately so) tube of brass, steel, plastic or ceramic material. Nozzle variables, including the vee angle, depth of vee and tube diameter can influence the flow rate, spray angle and droplet size distribution produced by the nozzle. Many approaches to nozzle design have, until recently, been based onempirical rules and extensive experiments with a range of spray liquids. The development of Computational Fluid Dynamics approaches in which the fluid flow behaviour in a defined geometry is predicted by computer calculations offers some scope for adopting a rational approach to nozzle design (Zhou et al. 1995). It must be recognised that the flow conditions in an agricultural nozzle involve high shear rates and small clearances - conditions which make calculations difficult. However an initial study has already shown that the output from computer modelling studies can provide some useful insight into nozzle design and has the potential to aid substantially the design of nozzles to meet defined criteria (Zhou et al. 1996).

Flat fan nozzles produce a range of droplet sizes with droplets leaving the nozzle at relatively high velocities (Miller et al. 1995c). This design is widely used on boom sprayers and has been adopted as a standard for performance comparison purposes (Doble et al. 1985).

Where a smaller mean droplet size is required at a given flow rate, for example when applying fungicides from a boom sprayer or any treatment with an air-assisted sprayer, then hollow cone nozzles may be more applicable. In such a design, more of the energy in the pressurized liquid is dissipated within the nozzle body to accelerate the flow in swirl producing a finer spray which leaves the nozzle at lower velocities than for an equivalent fan nozzle operating at the same flow rate. Anvil nozzles produce a spray as a result of the impaction of a metered liquid stream against an inclined face which forms part of the nozzle tip. The resulting spray is relatively coarse with a wider range of droplet sizes, travelling at lower velocities, than from flat fan nozzle designs (Miller et al. 1995c).

The need to reduce off-target spray drift has been a major driver in the recent design and development of agricultural spraying systems. Twin orifice flat fan nozzles, venturi nozzle designs and other forms of pressure nozzle creating relatively large droplets and coarse sprays has been a feature of many commercial developments during the past five years. However, although increasing droplet size and decreasing the percentage of spray volume in droplets <100 µm in diameter reduces the risk of drift, spray deposition on target surfaces can also be adversely affected. Droplet bounce and run-off from plant surfaces can give high levels of soil contamination which environmentally may be as important as spray drift.

Other spray generation systems

A number of other operating principles have been used to create sprays for applying agricultural pesticides, and while space does not permit a comprehensive review of these, some important features are worthy of mention. Spinning discs and spinning cages have the potential to produce a spray with good control of the droplet size distribution (Frost 1981). Mean droplet size is a function of physical disc size and design, rotational speed, liquid flow rate and the physical properties of the spray liquid. In many designs, the discs rotate about a vertical axis such that the droplets have an almost horizontal initial trajectory which can limit penetration into dense crop canopies unless operated in conjunction with air assistance techniques. Such units also operate at relatively low flow rates in comparison with those used through hydraulic nozzles and therefore the system is mainly suited to low volume rate applications. Spinning disc application systems have been developed successfully for specific applications spraying relatively high concentrations of herbicide formulations which often include specific adjuvants to match performance to target requirements.

Electrostatic spray generation systems (not systems for charging sprays formed by other means) also require spray liquids that have particular physical properties in terms of electrical conductivity and fluid flow characteristics. This system is a technically elegant method of producing a spray at very low flow rates and with excellent control of the droplet size distribution for a given formulation. The sprays produced are highly electrostatically charged and this has a major influence on trajectories and deposition patterns. However, other than some hand-held applications, this system has not been commercially developed for agricultural pesticide application.

Twin-fluid nozzle designs which use both compressed air and liquid supplied to the nozzle body have important characteristics. They have the ability to operate at a range of flow rates and create sprays with different droplet size distributions from the same nozzle assembly by independently varying air and liquid supply pressures. They are able to operate at relatively low flow rates using an orifice which is large enough not to be easily blocked. The spray formation process occurs more within the nozzle body so that the spray cloud is less influenced by a cross of air flow below the nozzle and can therefore give improved drift control. Research has shown that the larger droplets produced by many twin-fluid nozzle designs have 'air-inclusions' within them, influencing their behaviour in transport and on impaction with target surfaces (Rutherford et al. 1989, Miller et al. 1991).

The Venturi ('bubble jet') nozzle design also uses air within the nozzle to form the spray, similar to the twin-fluid nozzle, but the action of the liquid through a shaped orifice draws air into the nozzle body rather than by using air under pressure. Although Venturi nozzles are less flexible than twin fluid designs, they do create sprays with similar physical characteristics, including the presence of air inclusions in the larger droplets. To increase the flow of air drawn into the venturi nozzle, some systems are designed to operate at relatively high pressures (up to 7 bar on a boom sprayer) which may have implications for both practical and safe operation.

The presence of 'air-inclusions' in the droplets from both Venturi and twin fluid nozzles reduces the velocities of the larger droplets (>100 µm) by, of the order of, 50%, reduces bounce and changes the droplet behaviour on impact with a target surface (Miller et al. 1991). There is, however, relatively little published information on the performance of such spray droplets and this is an area where further research is required.

Spray formation

The formation of a spray is an interaction between the nozzle and the spray liquid and therefore a nozzle's performance is likely to be strongly affected by liquid properties and hence by the addition of adjuvants. Similarly, the way in which any adjuvant may act will be nozzle-dependent and so it is not possible to generalize about the effect of adjuvants on the formation of sprays.

There is a substantial amount of information in the literature concerning the relationship between liquid properties, including surface tension, viscosity and density, spray formation by flat fan nozzles and droplet size distributions. Sprays from flat fan nozzles are produced by the break-up of fan-shaped liquid sheets downstream of the outlet orifice. Dombrowski and Johns (1963) identified that the interaction of the moving liquid sheet with stationary air caused oscillations of the sheet, break-up of the sheet into ligaments and then into droplets. Dombrowski and Fraser (1954) found that viscosity and surface tension influenced liquid sheet break up although the effects may be complex. For example, increasing the surface tension reduced the sheet spray angle, leading to a greater liquid volume in the rims of the sheet and potentially more large droplets. However, the onset of sheet oscillations is moved further from the nozzle, where the sheet is thinner, which would tend to lead to smaller droplets. Theories of droplet formation (reviewed by Lefebvre 1989) suggest that, generally, increasing surface tension increases droplet size and measurements of droplet size in the centre part of the spray support this (Holloway 1994). Thus the effect of changing surface tension may be difficult to predict.

Most agricultural spray liquids consist of relatively small quantities of chemical formulations added to water and hence the density of the spray liquid will be similar to water alone. Viscosity and surface tension effects are therefore likely to be parameters influencing spray formation processes. Other liquid properties may also be important and although some, such as extensional viscosity (Akesson et al. 1994), have been identified the mechanisms through which they influence spray formation are as yet unknown. However, in complex liquids such as emulsions, the terms 'surface tension' and 'viscosity' become inadequate to describe the behaviour of the liquid during spray formation. Theories of spray formation are already mathematically complicated and these have, in the main, been derived assuming spray liquids are pure and Newtonian (Fraser et al. 1962, Ford and Furmidge 1967, Clark and Dombrowski 1972a,b, Rangel Sirignano 1988, 1991)). With more complex liquids, behaviour may be non-Newtonian and surface tension is probably time dependent. Viscosity of many of these liquids is unlikely to be substantially different from water at low shear rates, but at higher shear rates, pseudo plastic or visco-elastic behaviour may result. Recent studies of the spray angle produced by a defined nozzle geometry using computer modelling techniques (Zhou et al. 1996) have found good agreement with practical measurement providing that at low pressures in particular the sheet contraction effects due to surface tension are allowed for using approaches identified by Clark and Dombrowski (1972a,b). Liquid properties are therefore likely to have interactions with nozzle geometry, again making prediction of the effects on droplet formation difficult.

Significant differences in measured droplet sizes with different spray liquids have been reported, changing both mean size distribution (VMD) and the percentage of small and large droplets (e.g. Bouse et al. 1988, Adams et al. 1990, Akesson and Gibbs 1990, Bouse et al. 1990, Akesson et al. 1994, Holloway 1994, Miller, et al. 1995a). Changes in swath pattern have also been reported (Chapple et al. 1993). It has been shown that for sprays generated using flat fan nozzles, the changes in droplet size distribution, droplet velocities and spray cross-sectional area are consistent with observed changes to the method of spray formation (Butler Ellis et al. in press). However, work to relate changes in droplet sizes to liquid properties such as viscosity and surface tension have had limited success (e.g. Chapple et al. 1993, Hall et al. 1993, Hermanski and Krause 1995), partly because of the difficulty in measuring these properties at the short surface ages and high shear rates involved in droplet formation and partly because it is possible that not all the important parameters have yet been identified. Detailed dynamic surface tension measurements, using both the maximum bubble pressure method with commercially available equipment and the experimental oscillating jet method for very short surface ages have been made (Thomas and Hall 1979, Murphy et al. 1993, Brazee et al. 1994). These show large differences in the rate of change of surface tension, within the first few milliseconds of surface age, between spray liquids.

Clearly, nozzles which have a different spray formation mechanism may be

affected in different ways by liquid properties. For example, spraying water plus 0.1% non-ionic surfactant (e.g. Agral, Zeneca Crop Protection) through twin fluid nozzles results in droplets with air inclusions (Miller et al. 1991) which are likely to lead to nominally coarser sprays whereas through a flat fan nozzle, the addition of the same surfactant results in a finer spray (Miller et al. 1995a).

Spray transport

Spray from a generation system mounted on a machine usually travels through the air to arrive at the target surface. If the target is a crop canopy, then losses can occur as spray drift due to the action of the wind and as direct losses by run-off to the ground. The concept of spray accountancy has been considered by a number of authors (Combellack 1982, Parkin et al. 1985, Hislop 1987) with results typically showing that spray drift accounts for a relatively small fraction of the total sprayer output (e.g. 2% at 25 m downwind for a system applying 20 L ha-1, Parkin et al. 1985, and much lower figures for higher volume application rates). The complexity of many target structures and the large imbalance between different and important components of the spray accountancy can make it difficult to draw conclusions from such considerations.

Although only small percentages of sprayer output from boom machines operating over arable crops are lost as airborne drift, this loss from the target area can pose environmental and human safety problems. Considerable technical effort has therefore been directed at developing methods that will minimize drift from both boom and air-assisted machines. The initial droplet velocity and spray structure are key parameters influencing the risk of drift from boom sprayers and have been referred to above (Miller et al. 1995b). Increased downward velocities have a substantial impact on reducing drift and the effect of such variables can now be quantified using computer simulation models (Miller 1988). The forward motion of a boom sprayer causes air to flow through the spray structure and, in the case of the spray from flat fan nozzles, this results in trailing vortices that have been identified as a major factor contributing to spray drift (Young 1991, Miller et al. 1995b). The results from both wind tunnel and field experiments with twin-fluid nozzles (Miller et al. 1991, Young 1991) have indicated that the spray produced by this nozzle design is more porous to the air flow, that air movements around the spray are much less pronounced and as a consequence spray drift is reduced. The air inclusions within the droplets above approximately 100 µm produced by this nozzle design operating with many spray liquids also have implications for spray

drift. The presence of the air in a given size of droplet reduces the density and the terminal velocity such that the droplet is more prone to drift. The air inclusions therefore have no role in reducing spray drift, but do allow relatively coarse sprays to be used without the disadvantages of poor retention characteristics (Rutherford et al. 1989).

Droplets leaving the nozzle entrain an air flow which, in turn, influences the transport behaviour particularly of smaller droplets. A number of studies have aimed at quantifying this air flow (Briffa and Dombrowski 1966, Ghosh and Hunt 1994, Miller et al. in press) and recent studies have also considered how this droplet laden air flow will interact with a cross flow arising, for example, from the forward motion of a boom sprayer (Ghosh et al. 1993, Smith and Miller 1994).

The factors influencing spray drift have been reviewed by a number of authors (e.g. Elliot and Wilson 1982, Miller 1993). For boom sprayers operating in field crops, boom height above the target has been identified as one of the most critical operating variables (Norby and Skuterud 1974, Miller 1988) with the quantities of spray drift more than doubling for an increase in height from 0.5 to 0.7 m (Miller 1988). Environmental factors and particularly wind speed have also been shown to influence drift with a number of authors showing that, for the range of practical field operating conditions, drift increases approximately linearly with increasing wind speed (Miller et al. 1991, Miller 1993).

'Drift-Reduction' adjuvants are available which supposedly modify the spray by increasing the droplet size and thereby reduce the volume of spray contained in droplets which are prone to drift. Miller et al. (1995a) have shown that other adjuvants, not specifically designed or marketed as drift-reducers have a significant effect on the volume of liquid contained in such droplets.

Adjuvants can also influence the rate of evaporation of droplets and the residual droplet size after evaporation. Using a computer simulation model, Hobson et al. (1993) showed that differences in the quantity of spray drift from boom sprayers of more than 100% could be expected from different evaporating conditions and typical mean wind speeds.

Spray generator performance assessments

The main performance parameters relating to spray generation system performance are flow rate, droplet size and velocity distribution and the lateral distribution of spray liquid. For hydraulic pressure nozzles there is now an International Standard relating to the colour coding for identification of flow rate which allows a manufacturing tolerance of ±5% of the nominal flow rate value for a given pres-

Droplet size measurements

Droplet size distributions can be measured by capturing a sample of spray on a surface (water-sensitive paper, magnesium oxide or oil) and sizing using a microscope, purpose-built analyser or image analysis system. However, small droplets tend to follow air streams and therefore the sample that impacts on the collection surface is biased since most of the small droplet fraction will not impact on the surface (May and Clifford 1967). The surface must also be calibrated to account for droplet spreading on contact. Many droplet size distributions are now determined in-flight by using one of a number of designs of laser-based instruments. These instruments use different operating principles and sampling methods and these have been reviewed by Parkin (1993). A number of studies have compared the numerical results obtained with different instrument systems e.g. Arnold (1987) and, although some of the effects of different sampling approaches can be accounted for, numerical results from different systems operating in directly comparable spray conditions still show discrepancies. Because of this, the British Crop Protection Council (BCPC) have defined a classification scheme using reference flat fan nozzles to define five spray categories from very fine to very coarse. This scheme is now well established in the United Kingdom being used on chemical product labels, in codes of practice and in operating instructions for application machinery. Similar schemes have also been adopted in a number of other European countries.

The BCPC classification protocol uses a spray liquid of water plus 0.1% of a nonionic surfactant, Agral (Zeneca Crop Protection). Clearly, other spray liquids, such as those made with a commercial formulation, could result in a droplet spectrum sufficiently different to alter the classification. The addition of adjuvants may further enhance these differences or may mitigate them. The interaction between nozzles and liquid properties are not currently well enough understood to allow reliable prediction of the influence that an adjuvant will have on spray quality.

A further complication caused by the addition of adjuvants is the effect of droplet structure on the measurement technique. Phase Doppler analysis in forward scatter relies upon light refracting through a transparent spherical droplet to measure diameters accurately. Internal interfaces caused by air bubbles or emulsion droplets can disrupt the light path sufficiently to prevent realistic measurements being made. Thus care has to be taken when measuring sprays containing chemicals that the technique employed is not compromised by the liquid.

Wind tunnel measurements

Most existing spray classification systems relate only to the droplet size distribution. While this is a very important parameter determining both the physical and biological behaviour of droplets it does not give the complete picture. Droplet velocities, entrained air flows and spray structures also substantially influence the behaviour of droplets in the transport stage from nozzle to target and in the region of the target. Work in the United Kingdom and in Germany is now being conducted to extend the existing spray classification approaches to include wind tunnel studies of spray behaviour. An initial study defined measurement protocols for a standardized wind tunnel test based on single nozzles, but more recent studies (Miller et al. 1995b) have shown the need to account for adjacent nozzles on a boom, particularly if the spray structure is likely to provide a relatively non-porous barrier to air flow. Wind tunnel studies should provide information on the influence of droplet size, velocity and spray structure on spray transport. These effects are important when assessing the risk of spray drift, the behaviour of droplets on impact with the target and may also enable the classification of those sprays which cannot easily be classified under the existing scheme based solely on droplet size distributions.

Behaviour close to the target

Impaction on target surfaces

The conditions determining whether or not a droplet will impact on a collecting surface has been related to the characteristic dimensions of the surface and the local air velocities in the region of the surface (May and Clifford 1967). Small droplets travelling in slow moving air streams will not impact on surfaces unless the dimensions of the surface are very small. There is little information available concerning the local air velocities around target surfaces, which will clearly depend upon the application method and the crop structure.

Retention and coverage of the target

There are many ways in which adjuvants act (retention, spreading, uptake and translocation will all be affected by liquid properties) and it is difficult to assess the relative contributions of these mechanisms to the biological effect. Retention and spreading of individual droplets are particularly affected by the surfactant-plant interaction, whereas uptake and translocation are more likely to depend upon the more complex surfactant-pesticide-plant interaction (van Toor et al. 1994). In addition, the mode of action of pesticides varies, so it is important to ensure

that, if the adjuvant is to improve the efficacy of the pesticide, the appropriate mechanism is being enhanced.

The mechanisms involved in retention of liquid upon the target leaf and the degree of coverage achieved is as complex (if not more so) than droplet formation and transport. Whether the droplets rebound or adhere, whether they spread, roll off, fragment or coalesce with other droplets, will determine the total amount retained on the target and the uniformity of coverage. These factors are dependent on droplet size and velocity, volume of application, surface and bulk properties of the liquid and plant characteristics. Therefore in addition to altering droplet size and velocity, the use of adjuvants can affect retention and coverage by altering the interaction between the plant and the liquid, with this interaction being dependent on the target species.

Much work has been undertaken to study the effect of liquid properties on aspects of retention and coverage, some of which has been reviewed by Holloway (1995). The sequence of events surrounding the impact of a droplet on a solid substrate has been well documented, and summarized by Holloway (1994) and Tadros (1994). Mathematical descriptions of the processes involved have also been developed (e.g. Hartley and Brunskill 1958, Brazee et al. 1991, Tadros 1994) with varying degrees of complexity.

Experimental investigations into the effect of adjuvants on retention have shown a range of responses. Holloway (1994) investigated the effect of mean molar ethylene oxide (EO) content on retention on a variety of target leaves and determined that EO content was an important factor in retention of spray droplet on difficult-towet targets, with EO contents <10 performing poorly. Because the addition of a surfactant can lead to smaller droplets travelling slower, one would expect retention to be increased as a consequence. However, Hall et al. (1993) reported that an adjuvant which increased the proportion of large droplets in a spray actually reduced the number of droplets which were reflected from the surface of cabbage leaves, despite the fact that larger droplets are likely to be travelling faster and be more susceptible to reflection. Bukovac et al. (1995) also found a number of adjuvants reduced the reflection from cabbage leaves and that there was no correlation between retention and equilibrium surface tension but a high correlation between retention and surface tension at surface ages between 0.5 and 10 ms.

Since the impaction of a droplet on the leaf takes place within milliseconds, initial retention of the droplet on the leaf is more likely to depend upon dynamic properties, such as dynamic surface tension, than on equilibrium properties. Several studies

(e.g. Anderson and Hall 1989, Stevens et al. 1993) have suggested that the adhesion of droplets to the target leaf is correlated with dynamic surface tension. Organosilicone surfactants, for example, have a high rate of change of surface tension and so have the potential to increase adhesion and so improve the retention on the target (Stevens et al. 1993).

However, spraying sufficient volume so that run-off is achieved eliminates many of the dynamic effects and showed that retention increased with increasing equilibrium surface tension and is largely independent of dynamic surface tension (Cooper and Hall 1993). This serves to show that the way in which adjuvants are used in experiments is crucial to the results achieved. In order to ensure that these results are not extrapolated inappropriately it is important to begin to understand the underlying mechanisms.

The theory of spreading of droplets on a solid substrate is outlined by Tadros (1994) and suggests that reducing equilibrium surface tension reduces contact angle and consequently increases spreading. Experimental investigations into the effect of liquids on droplet spreading have shown a wide variety of responses to adjuvants, demonstrating that the processes involved are not straightforward. For example, the addition of surfactants to an acetone-water solution of glyphosate had a significant influence on its spreading properties without changing its equilibrium surface tension (van Toor et al. 1994). Many surfactants have little or no effect on spreading on easy-to-wet targets although the best spreaders currently available, which can have a significant effect, are organosilicone surfactants with a very low (~20 mN m-1) equilibrium surface tension (Holloway 1994). It is commonly assumed that increasing spreading by reducing surface tension leads to improved efficacy (Murphy et al. 1993). However, Brumbaugh et al. (1995) suggest that lowering surface tension and increasing spreading can reduce weed control, which may be due to a reduced concentration gradient (Tadros 1994) or more simply because too much spreading led to run-off and a lower total dose. Organosilicone surfactants were shown to exhibit greater spreading than fluorocarbon surfactants which have a greater effect in reducing surface tension (Murphy et al. 1993). Clearly, factors other than equilibrium surface tension are important and, in particular, interfacial tension (between the leaf and the liquid) and the physical properties of the leaf surface need to be considered.

Conclusions

The addition of adjuvants to pesticide sprays affects all elements of the application process. No one aspect can be considered in isolation when investigating the effect adjuvants have. For example, a surfactant that improves uptake may affect droplet formation and transport and similarly, drift reducers may also influence uptake. Few studies have taken all the different elements into account and there is a need to pull the available information together if the modes of action of adjuvants are to be fully understood. Many authors acknowledge that some adjuvants demonstrate effects on the application of pesticides which are not fully explained by current theories.

Unfortunately, although substantial progress has been made in the development of methods for measuring and predicting spray formation and behaviour, many of the individual processes which adjuvants can influence are not well understood. For example, the mechanisms of spray formation are modified by the addition of adjuvants but all of the relevant physical liquid properties are which are responsible for these changes have not yet been identified. Consequently it is only possible to evaluate the effect of an adjuvant on the efficacy of a pesticide by experimental means and this can rarely be extrapolated to other situations.

Once the underlying modes of action of adjuvants have been identified and understood, it will become feasible to predict their overall effect on the efficacy of a pesticide application. It may even be possible to design adjuvants which will promote the positive aspects (such as target coverage and uptake) and reduce any negative aspects (losses due to spray drift or runoff) of the application process and thereby substantially improve pesticide efficiency.

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