

# *Control of Drop Size Distribution Through Emulsions*

**Dongyue Wang**

*Experimental High School Attached to Beijing Normal University, Beijing, China  
wangdongyue1710@sina.com*

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**Abstract:** Modern agriculture has been relying on the use of liquid pesticides and fertilizer and these solutions are usually sprayed onto crops. Whether spraying by hand, drones or airplanes, there can be a substantial amount of liquid droplets that are too small and drift by currents in the air away from the targeted crop. Therefore, controlling drop size distribution of sprayed droplets is important for enhancing the efficiency of agrochemical applications. In previous studies, researchers controlled droplet size by altering the nozzle pressure [1], and pressure-and-nozzle combinations [2], as well as the viscoelasticity of the liquid by adding polymers [3]. In this paper, the fluid mechanical response of the liquid to be sprayed is adjusted by formulating oil-and-water emulsions. The presence of such oil-water interfaces introduces a viscoelastic response through the restoring force of surface tension. By analyzing the drop size distribution as a function of the volume fraction of oil in water, it is found that the distributions can be shifted toward maximal size of droplets. In this work, using silicone oil-water combinations, the maximum drop size occurred in the vicinity of 40% by volume of silicone oil.

## **1. Introduction**

In previous studies, researchers investigated controlling drop size distributions by interfacial rheology [4], and Wang et al. [5] studied the effect of the hydrophobicity of emulsions to aid in the attachment of droplets onto plant surfaces. In this paper, we controlled the drop size distribution in a novel way by introducing surface tension [6] restoring forces to create viscoelasticity[7]. Specifically, oil-and-water emulsions were created, and the presence of surface tension between the oil-and-water phases introduced a relaxation time by capillarity. This relaxation time, which can be estimated by

$$\lambda = \frac{\eta R}{\sigma}$$

where  $\eta$  is the viscosity of the droplet,  $R$  is the average radius of the droplet, and  $\sigma$  is the surface tension. This effectively renders the emulsion to be slightly viscoelastic [10]. Since the active ingredients in agricultural sprays are usually contained within the oil phase, the use of such emulsions is particularly convenient for the adjustments of drop size distributions. We also utilized detergents to

independently adjust the surface tension, which further demonstrated the importance of this property to control drop size distribution.



Figure 1 Agriculture spray to apply fertilizer or pesticide on crops [8].



Figure 2 Accuracy of sprayed droplets are vulnerable to wind drift [9].

## 2. Materials

In the experiments, the following materials were used: de-ionized water, silicone oils with viscosities of 0.65cst and 1cst (manufactured by Dow Corning, PMX-200), black non-carbon ink (manufactured by Polit, INK-30-B), black food dye (combination of sunset yellow, food blue, allura red, and tartrazine aluminum lake; manufactured by PCB, 6972957830013), and a household detergent (manufactured by Gold Fish, 100009132441). Photographs of these products can be found in Figures 3, 4 and 5.



Figure 3 Pilot INK-30-B



Figure 4 PCB 6972957830013



Figure 5 Gold Fish 100009132441

## 3. Creation of Emulsions and Emulsion-Detergent Mixtures

The oil and water emulsions were created by first measuring a specific volume of oil and adding it into a known volume of water. Oil volume fractions of 0%, 15%, 20%, 40%, 60%, 80%, and 100% were created for the 1 cst oil and an emulsion of 15% by volume was created for the 0.65 cst oil.

Surfactants were introduced into the emulsions by first placing a small amount of surfactant into water at a concentration of 1.3%. This surfactant solution was then added to 1 cst silicone oil to create a 20% emulsion.

## 4. Experimental Apparatus

### 4.1. Overview of Experimental Apparatus

The experiment was conducted using a special designed spraying platform shown in Figure 6. In the platform, emulsions are sprayed by an air- pressure spray bottle under an inlet air pressure of approximately 0.16 MPa and is held steady during the spray application.



Figure 6 spraying platform. Emulsions are stored in the container attached to the spray gun, the white tube connects the spray gun with an air tank, which pressure is regulated by a pressure valve.

The spray bottle (the gray vessel in Figure 6) contained a magnetic stirrer. The magnetic stirrer agitated the emulsions at the same power for all experiments and this kept the emulsions from phase separating. Figure 7 shows the effect of maintaining the water-and-oil emulsion morphology by rotating the magnetic stirrer.

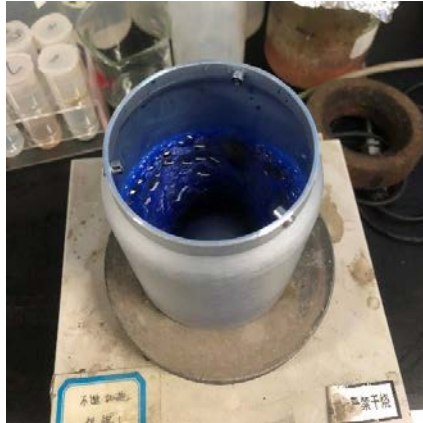


Figure 7 A photo taken of only and magnetic stirrer in the spray platform, demonstrating the effect of the stirrer in preventing phase separating.

#### 4.2. Spray Nozzle Device

The largest nozzle (diameter 1.8 mm) was chosen since the drops produced by the other available nozzles were too small to be seen with naked eye and are outside the range desirable for agricultural sprays. Figure 8 shows the three nozzles that were compared, the first one on the left was used in this work.

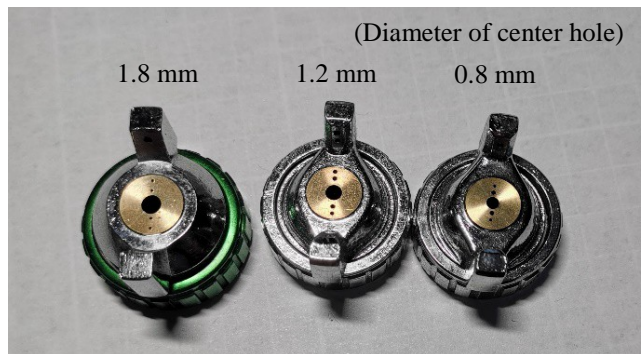


Figure 8 Spray nozzles considered

#### 5. Spraying Process

In conducting the experiments, the trigger of spray bottle was pulled and liquid inside the bottle was sprayed forward, creating small droplets that fell onto a plain A4 paper target that hung at 80 cm in front of the spray nozzle. Figure 9 shows the arrangement of experimental equipment.

While spraying, the magnetic stirrer was constantly agitating the liquid inside the bottle. The force generating the spray was constant since the air pressure in the spray vessel remained unchanged.



Figure 9 A simple sketch of arrangement of spray gun and target paper.

## 6. Visualization Techniques

### 6.1. Dyeing, Scanning, and Processing

To aid in visualization, a black, non-carbon ink was added into the water at a volume fraction of 4% of the mixture; the black food dye (which is an oil-soluble powder) was added into the silicone oil at a concentration of 0.8 g per 100 ml. The colored water and colored silicone oil were then mixed to produce emulsions. Therefore, droplets falling on the paper target left a visible trace.

The paper with colored droplets was then dried and scanned under high resolution imaging (9779\*13889 pixels, approximately 47\*47 pixels in picture in each mm<sup>2</sup> on the paper, data obtained from International Organization for Standardization [11]). The scanned picture was then binarized. Figure 10 shows a sample of raw data and its corresponding binarized data.

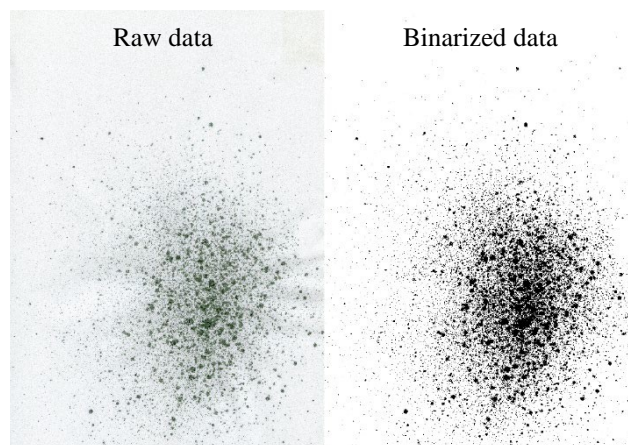


Figure 10 an example of scanned picture and its binarized version. This data is one of the sprayed results of 60% oil (1 cst).

As can be seen in these figures, the density of drops is a maximum at the center of the spray direction. It is also evident that larger droplets having a greater mass, have a relatively higher concentration at the center of the distribution compared to smaller drops. The ellipticity of the spraying patterns, is a result of the presence of small openings in the nozzle that deform the shape of the spray (see Figure 8).

The trajectory of droplets moving through air under the additional influence of gravity is very sensitive to the droplets' mass and it is expected that larger droplets will generally have a ballistic response that will favor their deposition toward the center of the spray whereas smaller droplets are more likely to be deflected away from the center.

The ballistic nature of the droplet trajectories motivated dividing the deposition patterns into ellipses with aspect ratios that resemble the overall shape of the sprays. Figure 11 shows an example of this procedure where a spray pattern has been reduced in size to capture the principal droplet deposition result.

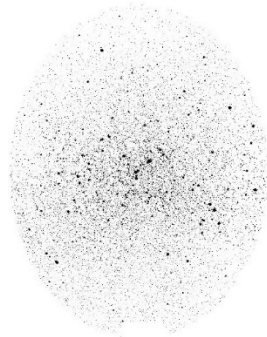


Figure 11

## 6.2. Microscopic Analyzes of Droplets from Spray Emulsions

Figures 12, 13, and 14 show magnified images of the spray droplets that originate from different emulsions. Interestingly, depending on the original emulsion composition, a variety of droplet morphologies were obtained. In general, when water was the major phase (Figure 12), the sprayed droplets consisted of micro drops of oil within a matrix of water. On the other hand, when oil is the major phase (Figure 13 and 14) the droplets consisted of micro drops of water residing an oil matrix.

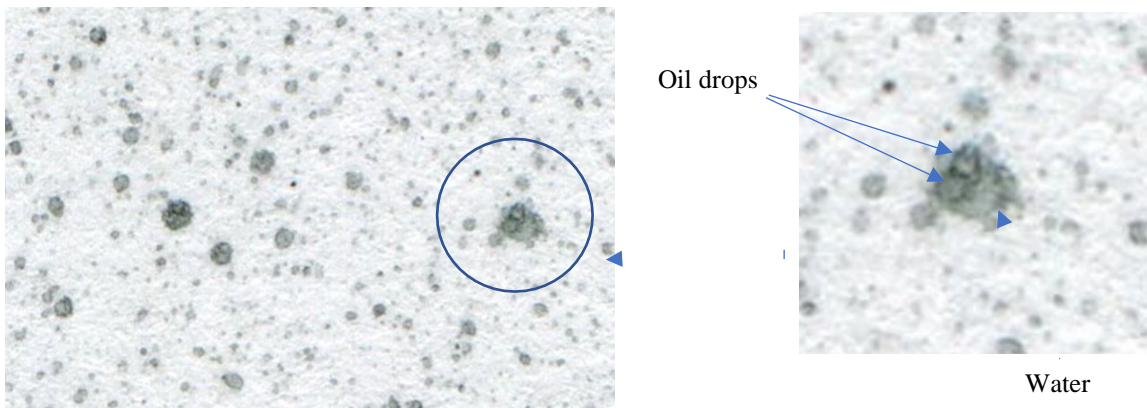


Figure 12 Part of scanned picture of one of the 20% oil (1cst) emulsion spray results.

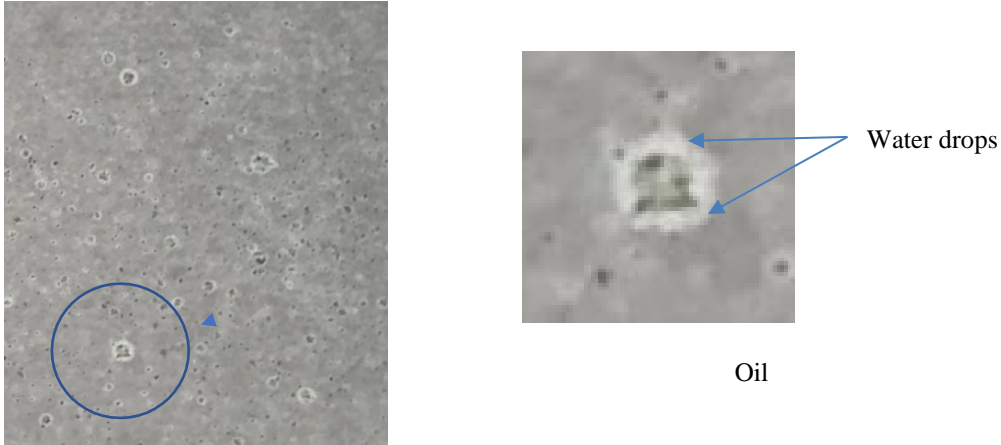


Figure 13 Part of picture of one of the 60% oil (1cst) emulsion spray result taken by camera. Oil appears on the bottom layer and infiltrates into paper after attached (dye stays at the contact area) and make the surrounding area of paper more transparent.

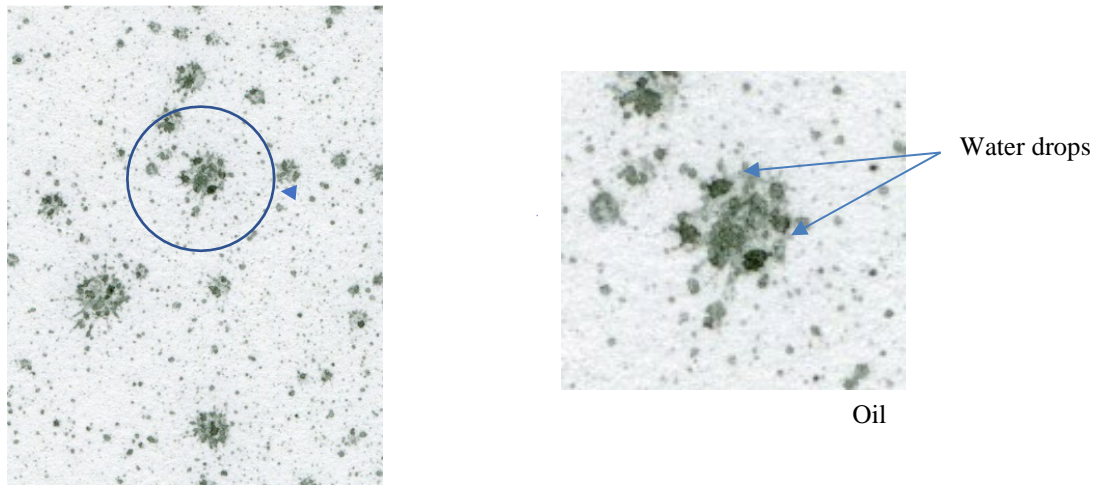


Figure 14 Part of scanned picture of the same result on Figure 14.

## 7. Calculation Method and Discussion of Results

### 7.1. Average Drop Size

A MATLAB program was used to analyze the pictures and calculate the number and size of droplets. An edge computing algorithm called “regionprops” was used to analyze the binarized versions, trace the outline of droplets, where the number and size of drops were computed regardless of whether they are oil-in-water drops or water-in-oil drops. In other words, areas of binarized pictures with black pixels were regarded as the area covered by the liquid drops.

Following this procedure, the area of every drop was calculated. Bins representing ranges of drop sizes were then used to collect the numbers of drops in respected size ranges. Then, in this manner, the drop size distributions were determined.

Results of these calculations are shown in Figures 15, 16, and 17. In Figure 15, the average droplet size is shown as a function of the volume fraction of the silicone oil with viscosity 1 cst, which is close to that of water. Figure 16 displays the average droplet size for emulsions created using the two different silicone oil viscosities, also including pure water. Figure 17 shows the average drop sizes and the effect of adding surfactant to the 1 cst oil emulsions.

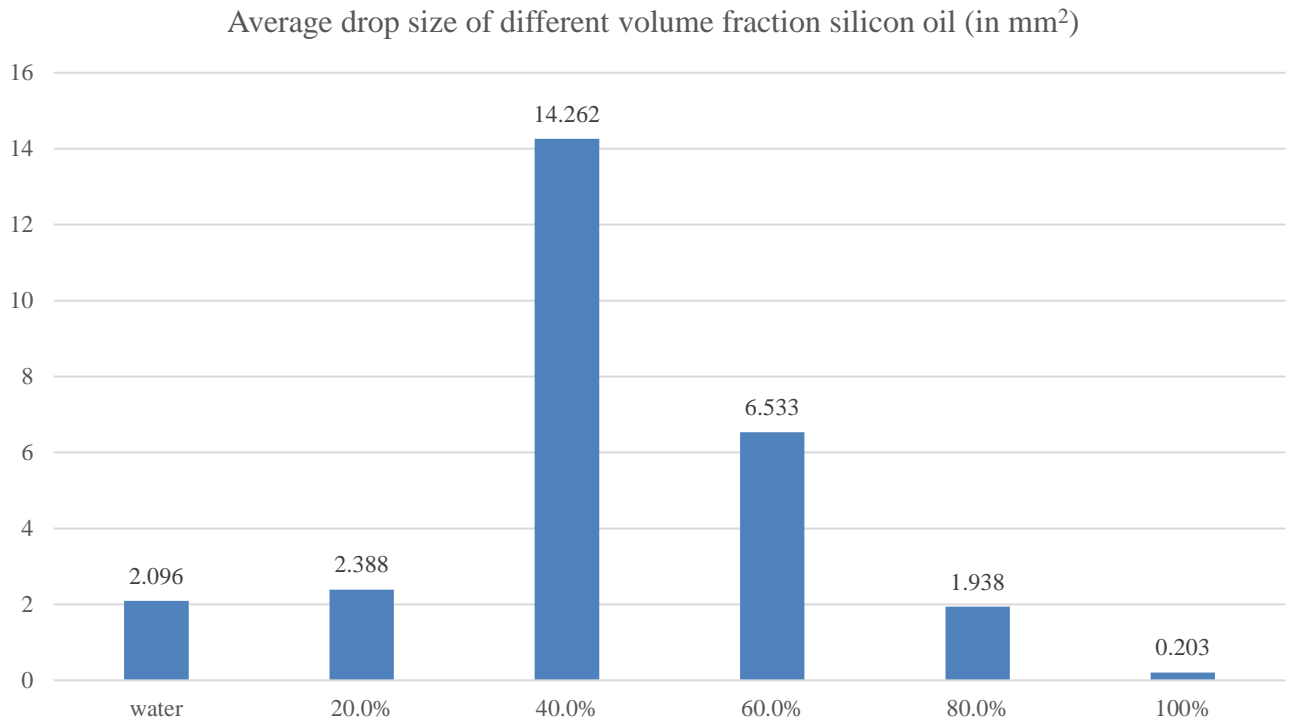


Figure 15 This histogram shows the average drop size of different volume fractions of silicon oil (1 cst). At 40% volume fraction, the average drop size maximized. The column marked “Water” is the control group.

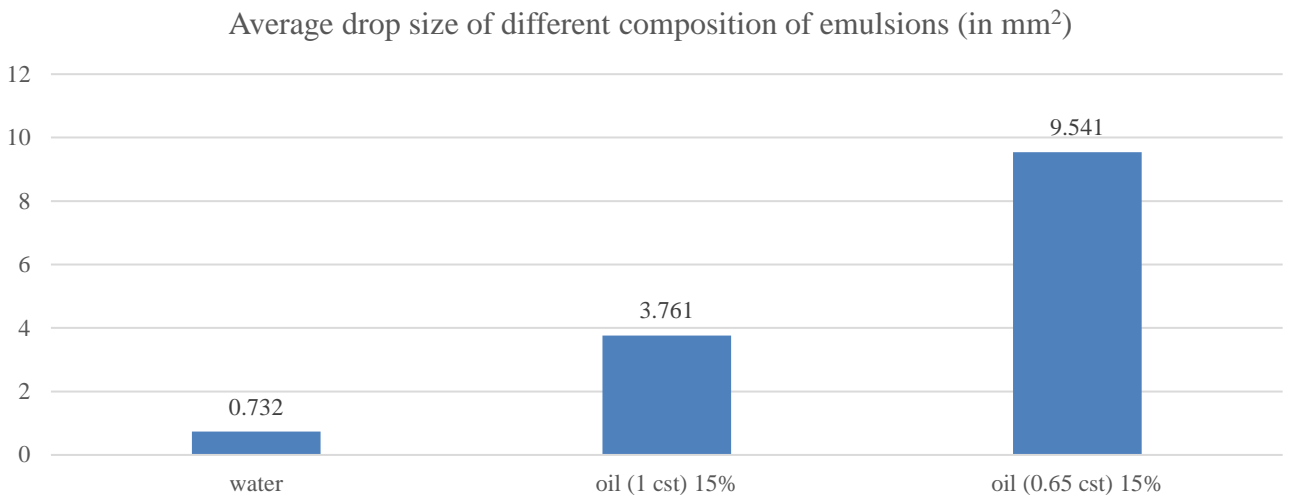




Figure 16 This histogram shows the average drop size of different viscosities of silicon oil (1 cst and 0.65 cst) at 15% volume fraction. The column marked “Water” is the control group.

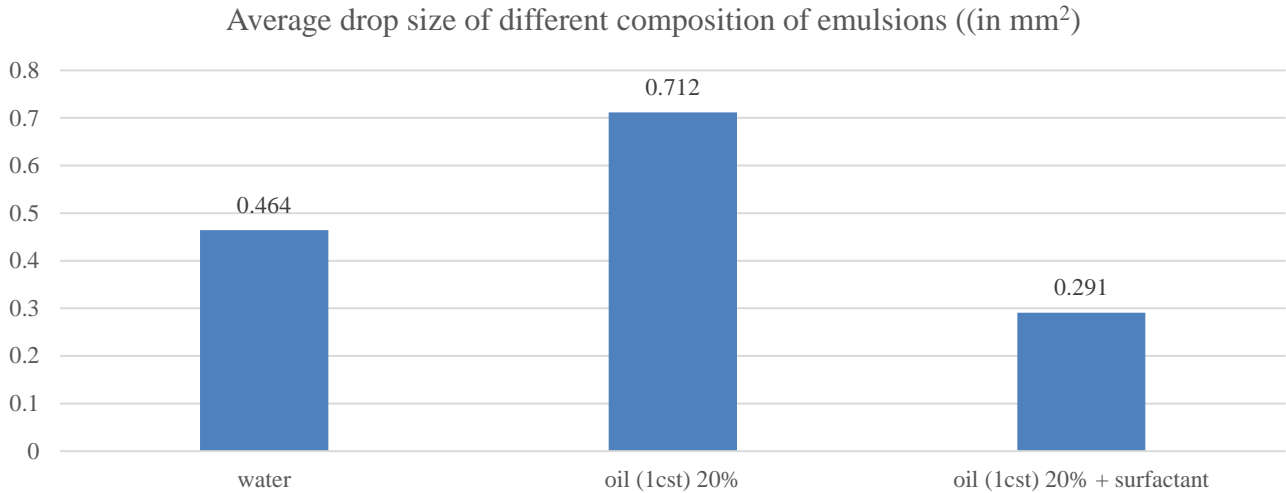


Figure 17 This histogram shows the average drop size of silicone oil (1 cst) at 20% volume fraction and that emulsion added surfactant. The column marked “Water” is the control group. The surfactant lowered the surface tension, demonstrating the importance of surface tension in controlling drop size

## 7.2. Visual Representation of Sprayed Ballistics

It is expected that larger drops, having a greater mass, will be less effective by frictional resistance of the air. For this reason, larger droplets, in general, will have ballistic trajectory, causing them to land closer to the center of the spray. This is a straightforward way to determine the effectiveness of this surface-tension controlled drop size distribution in reducing the amount of small liquid drops that are affected by currents in the air.

To determine the ballistically driven drops, another MATLAB program was designed. In this program, the scanned picture was divided into a number of smaller parts of 500\*500 pixels (approximately 10.6\*10.6 mm<sup>2</sup> on paper). In each of these parts, the number of drops and their sizes were counted, and the number concentration of drops was represented in the graph. The more ballistic the spray is, the more peaked the size distribution will be.

Examples of these calculations are shown in Figures 18 to 23 for different emulsions created using the 1 cst silicone oil but at different volume fractions of the oil.

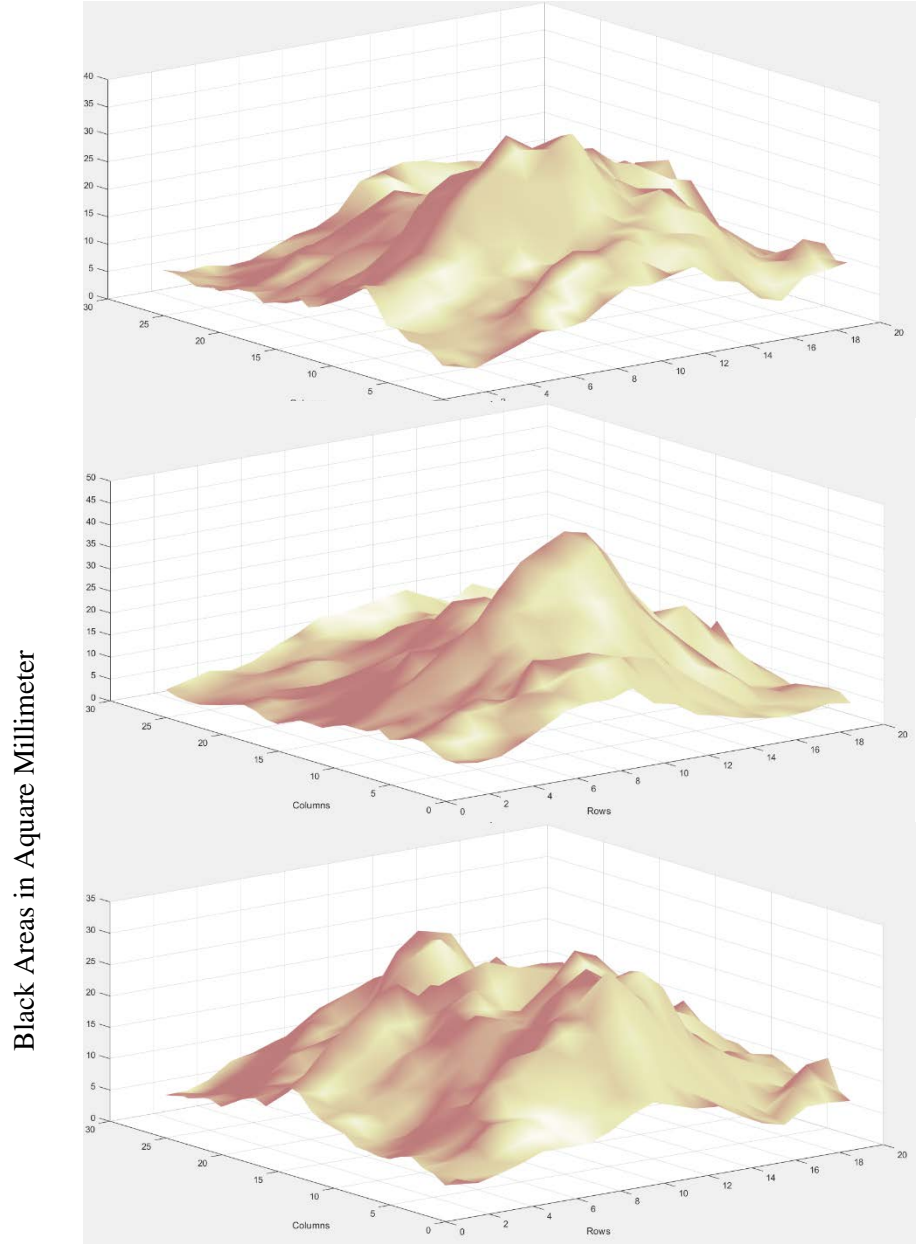


Figure 18 Three examples of ballistics analyze of 0% silicone oil results.

Black Areas in Aquare Milimeter

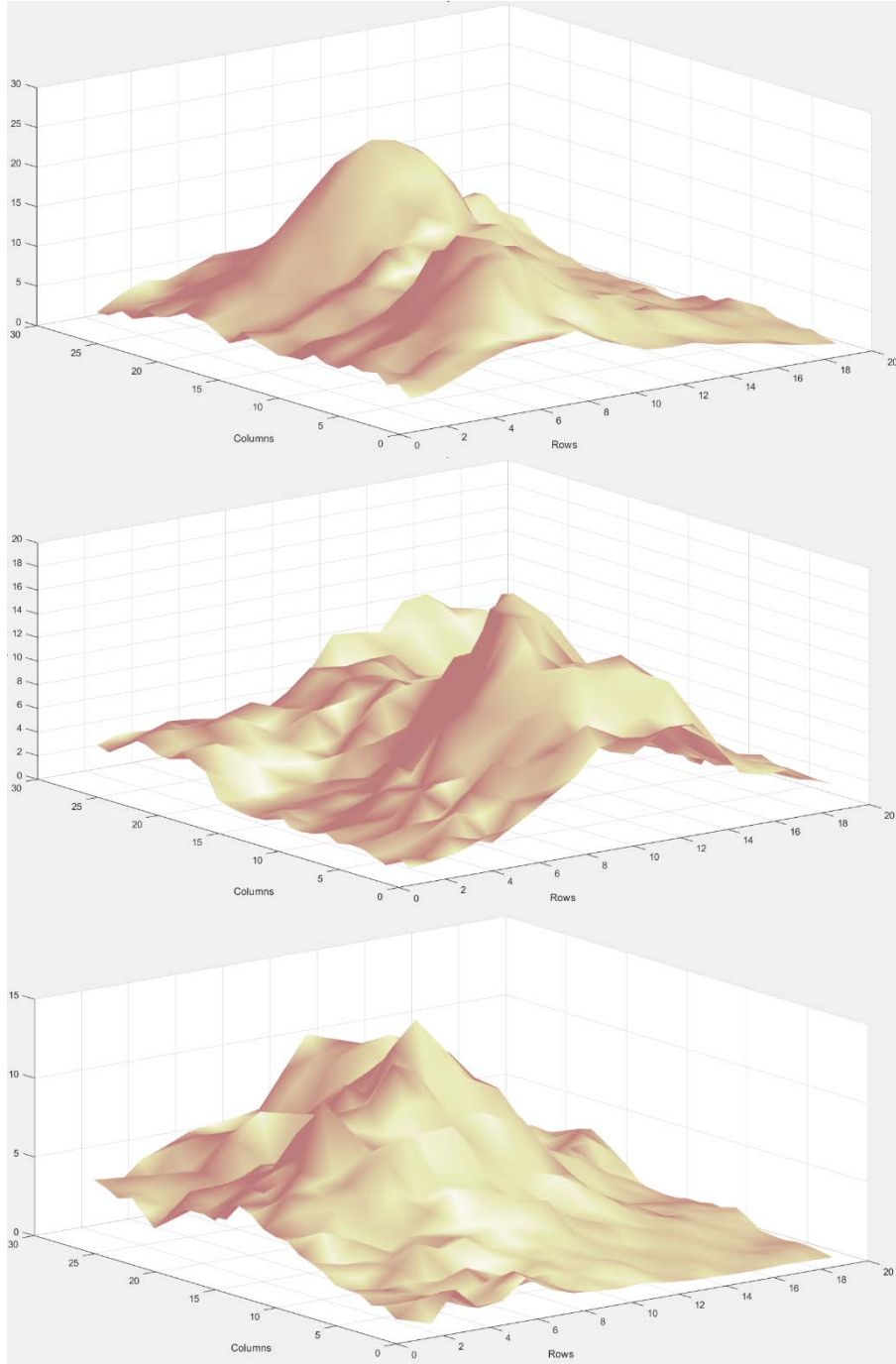


Figure 19 Three examples of ballistics analyze of 20% silicone oil (1 cst) results.

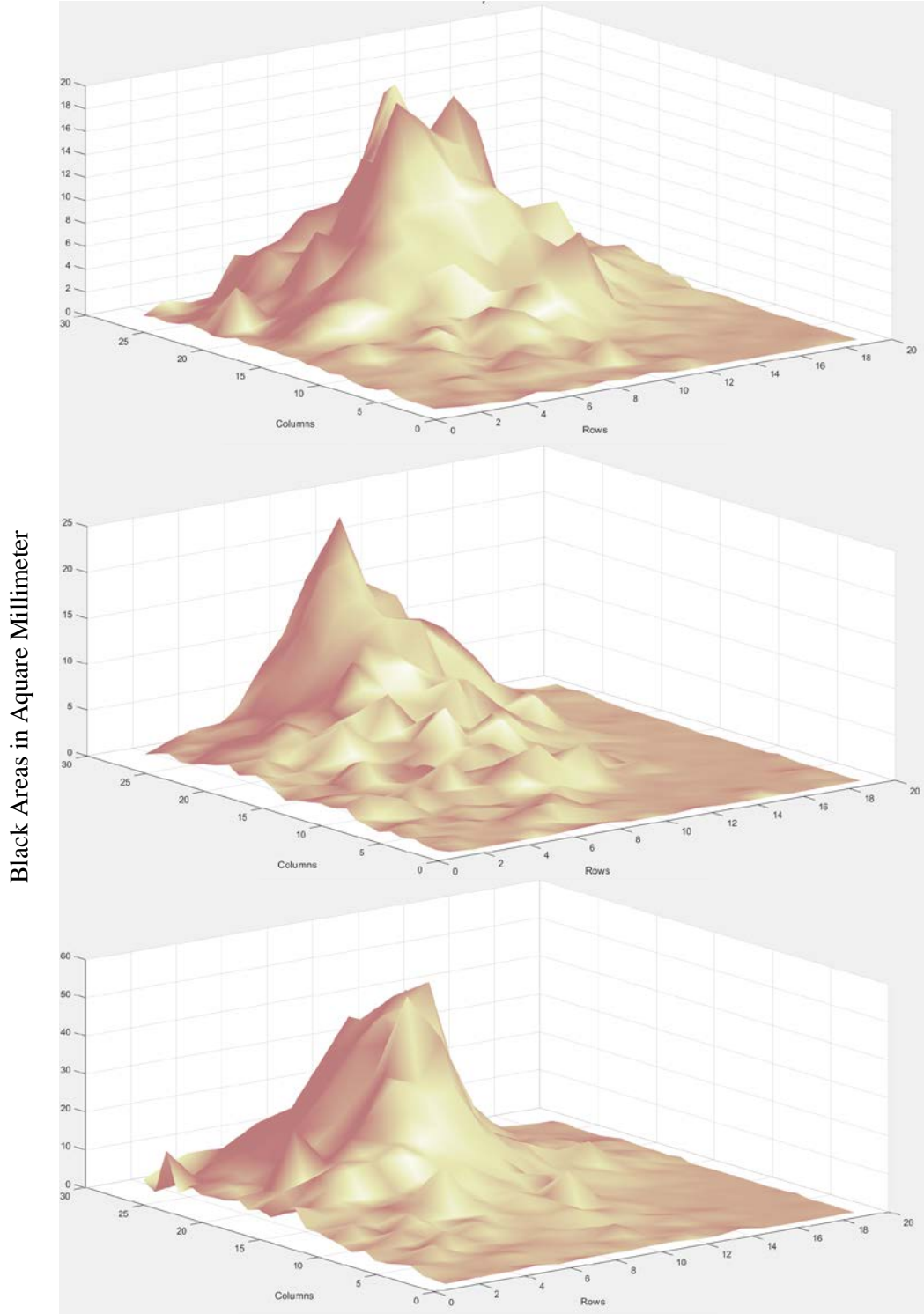


Figure 20 Three examples of ballistics analyze of 40% silicone oil (1 cst) results.

Black Areas in Aquare Millimeter

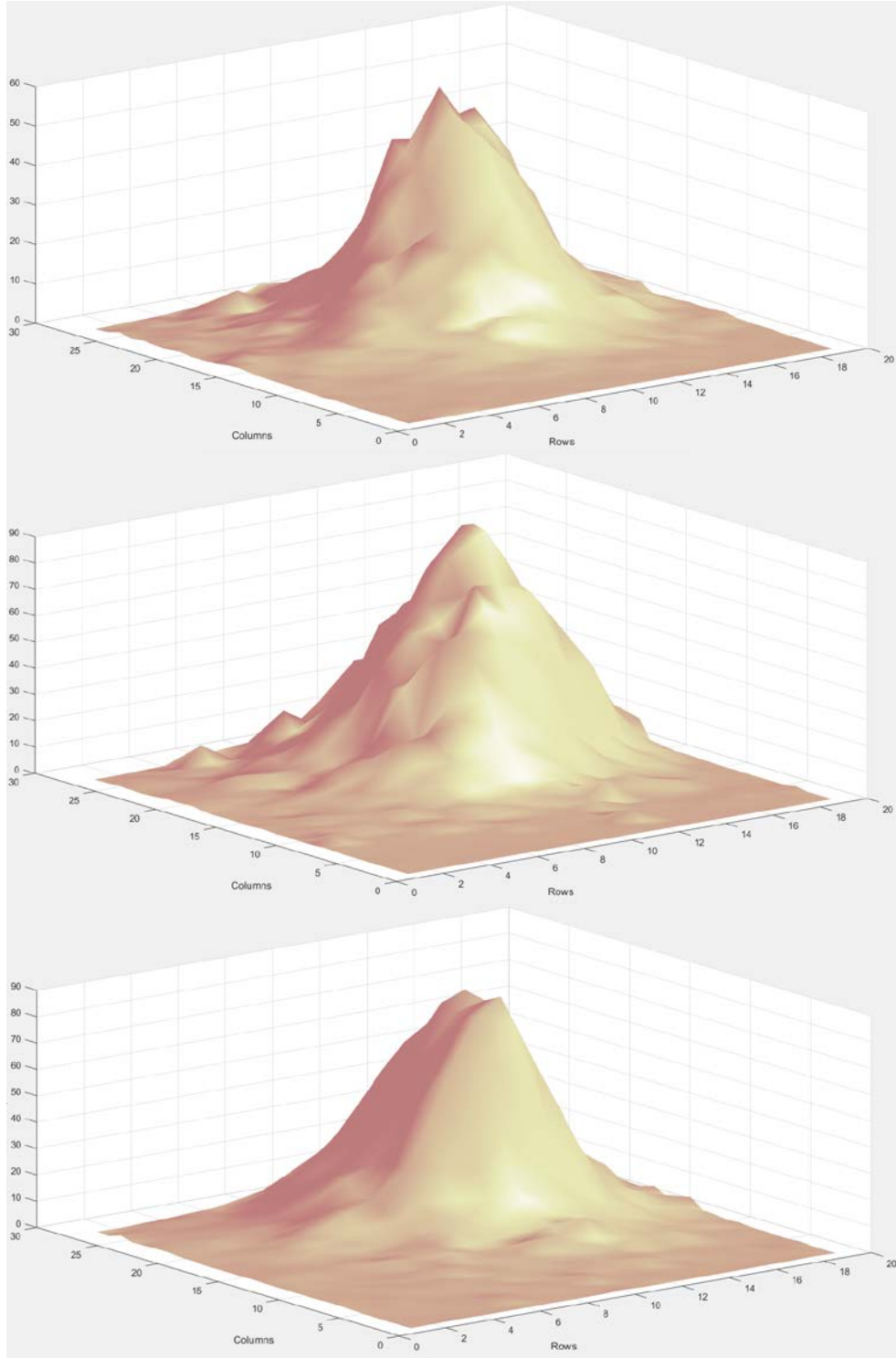


Figure 21 Three examples of ballistics analyze of 60% silicon oil (1 cst) results.

Black Areas in Aquare Millimeter

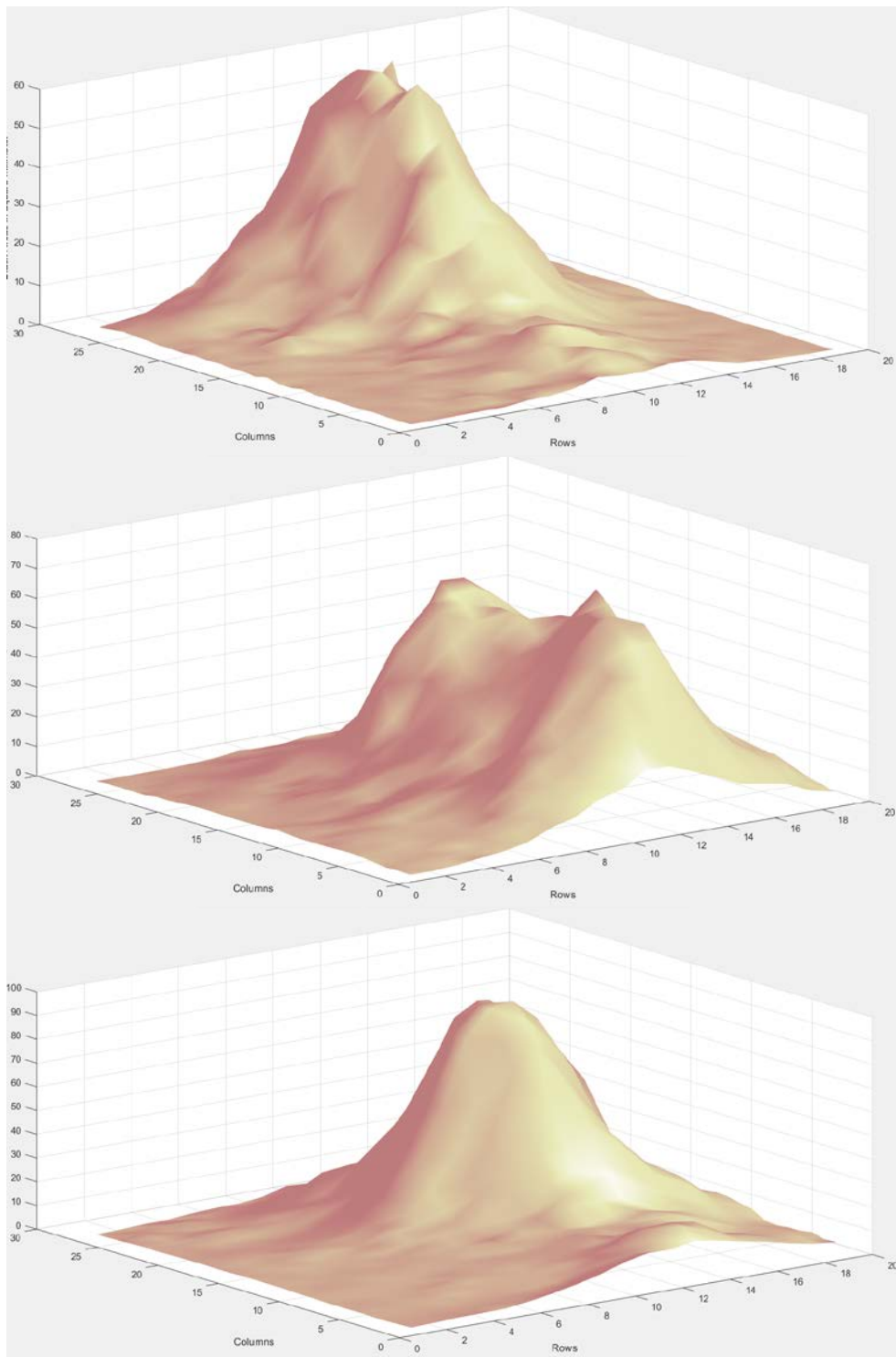


Figure 22 Three examples of ballistics analyze of 80% silicone oil (1 cst) results.

Black Areas in Aquare Millimeter

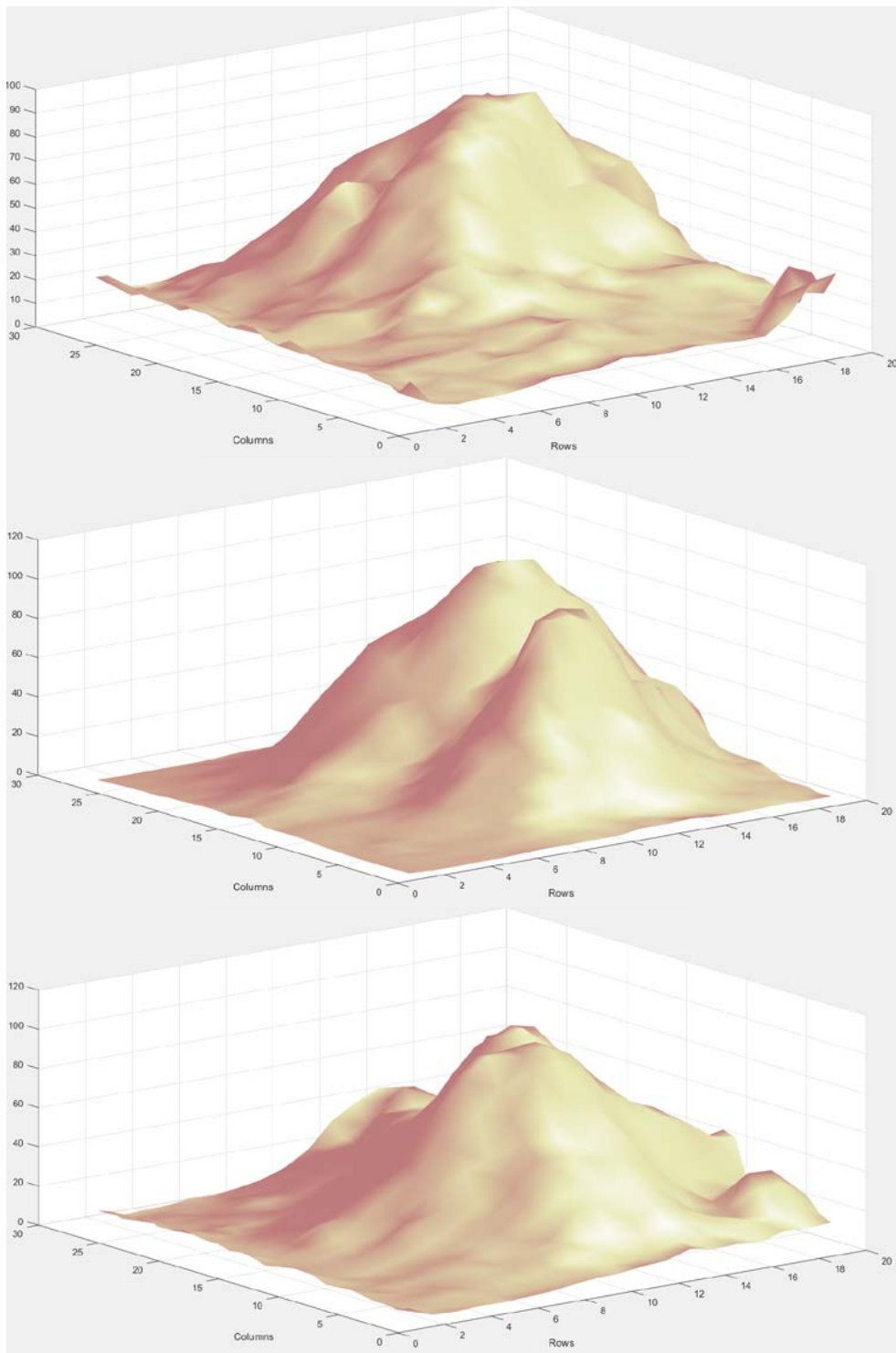


Figure 23 Three examples of ballistics analyze of 100% silicone oil (1 cst) results.

These 3-D figures show the density of drops on target paper in mm<sup>2</sup> per approximately 124mm on target paper. Therefore, its slope can show the ballistic nature of the emulsion. As shown in the figures, the maximum ballistic appears at 40%, 60% and 80%.

## 8. Discussion of Results

We have successfully demonstrated the influence of the restoring force of surface tension in controlling the drop size distribution of oil and water emulsions. Figure 15 clearly demonstrates that the drop size distribution and the control of the average drop size can be systematically controlled by adjusting the volume fraction of oil in such emulsions. Indeed, it appears that the largest average drop size is obtained with an approximately 50-50 water-oil composition for the case when the oil has essentially the same viscosity as water. As shown in Figure 16, it is demonstrated that the viscosity of the oil plays an important role where the lower viscosity oil of 0.65 cst produced, on average, larger drops. Mapping out the full dependence of the water-to-oil viscosity ratio will be an important consideration for future investigations. Figure 17 indicated the influence of the magnitude of surface tension, where the addition of the surfactant, which lowers surface tension, led to a decrease of average drop size.

The influence of the drop size distribution on the patterns of the deposited droplets is shown on Figures 18 to 23. The three-dimensional drop distribution plots reveal the expected result that, on average, sprays containing larger drops are more likely to be highly peaked about the center of gravity of that spray.

## 9. Conclusions

In this paper, we hypothesized and have shown the effectiveness of this novel method of controlling drop size distribution—by utilizing the surface tension of emulsions. This provides an especially helpful and convenient strategy for agricultural sprays of liquid pesticides and fertilizers since it can help create an optimal drop size and thereby reduce waste caused by wind drift. Among the most important findings in this work was the identification of the optimal volume fraction in oil-water mixtures to achieve maximal average drop sizes. Although not explored in great detail, it was found that the addition of surfactants, which reduce surface tension, led to a dramatic decrease in the average drop size. For this reason, the use of surfactant to stabilize emulsions should probably be avoided. Furthermore, it is evident that the viscosity ratio between the different phases in the emulsion is important.

Although our hypothesis has been successfully demonstrated by this research, there are still many aspects to be investigated. For example, the viscosity ratio between the oil phase and the aqueous phase should be explored. Since the surface tension between the phases plays an essential role in this approach, optimizing this variable is also important.

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## Appendix: MATLAB Code

### Average Drop Size

```
clear;
%Mixture1 is the mixture of water and oil with viscosity 1
%Mixture2 is the mixture of water and oil with viscosity 0.65
imageWaterNo1 = imread("C:\Users\wangd\Desktop\E~\oil
60%\img012.jpg");
imageMixture1No1 = imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (1).jpg"); imageMixture2No1 =
imread("C:\Users\wangd\Desktop\E~\oil 100%\oil 100% (1).jpg");

imageWaterNo2 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img013.jpg"); imageMixture1No2 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (2).jpg"); imageMixture2No2 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (2).jpg");

imageWaterNo3 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img014.jpg"); imageMixture1No3 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (3).jpg"); imageMixture2No3 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (3).jpg");

imageWaterNo4 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img016.jpg"); imageMixture1No4 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (4) -
.±±¾.jpg");
imageMixture2No4 = imread("C:\Users\wangd\Desktop\E~\oil 100%\oil 100% (4).jpg");

imageWaterNo5 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img018.jpg"); imageMixture1No5 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (4).jpg"); imageMixture2No5 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (5).jpg");

imageWaterNo6 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img020.jpg"); imageMixture1No6 =
imread("C:\Users\wangd\Desktop\E~\oil 100%\oil 100% (6).jpg");
imageMixture2No6 = imread("C:\Users\wangd\Desktop\E~\oil 40%\oil 40% (8).jpg");

imageWaterNo7 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img023.jpg"); imageMixture1No7 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (6).jpg"); imageMixture2No7 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (7).jpg");

imageWaterNo8 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img024.jpg"); imageMixture1No8 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (7).jpg"); imageMixture2No8 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (8).jpg");

imageWaterNo9 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img025.jpg"); imageMixture1No9 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (8).jpg"); imageMixture2No9 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (9).jpg");

imageWaterNo10 = imread("C:\Users\wangd\Desktop\E~\oil 60%\img026.jpg"); imageMixture1No10 =
imread("C:\Users\wangd\Desktop\E~\oil 80%\oil 80% (9).jpg"); imageMixture2No10 = imread("C:\Users\wangd\Desktop\E~\oil
100%\oil 100% (10).jpg");
%µ±¼Æ→¾«¶Ë!ý,βË±ÐèÒ³½«¼Æ→ÒÖµ»-
bwWaterNo1=imbinarize(rgb2gray(imageWaterNo1));
bwMixture1No1=imbinarize(rgb2gray(imageMixture1No1));
bwMixture2No1=imbinarize(rgb2gray(imageMixture2No1));
bwWaterNo2=imbinarize(rgb2gray(imageWaterNo2));
bwMixture1No2=imbinarize(rgb2gray(imageMixture1No2));
bwMixture2No2=imbinarize(rgb2gray(imageMixture2No2));
bwWaterNo3=imbinarize(rgb2gray(imageWaterNo3));
bwMixture1No3=imbinarize(rgb2gray(imageMixture1No3));
bwMixture2No3=imbinarize(rgb2gray(imageMixture2No3));
bwWaterNo4=imbinarize(rgb2gray(imageWaterNo4));
bwMixture1No4=imbinarize(rgb2gray(imageMixture1No4));
bwMixture2No4=imbinarize(rgb2gray(imageMixture2No4));
bwWaterNo5=imbinarize(rgb2gray(imageWaterNo5));

bwMixture1No5=imbinarize(rgb2gray(imageMixture1No5));
bwMixture2No5=imbinarize(rgb2gray(imageMixture2No5));
bwWaterNo6=imbinarize(rgb2gray(imageWaterNo6));
bwMixture1No6=imbinarize(rgb2gray(imageMixture1No6));
bwMixture2No6=imbinarize(rgb2gray(imageMixture2No6));
bwWaterNo7=imbinarize(rgb2gray(imageWaterNo7));
```

```

bwMixture1No7=imbinarize(rgb2gray(imageMixture1No7));
bwMixture2No7=imbinarize(rgb2gray(imageMixture2No7));
bwWaterNo8=imbinarize(rgb2gray(imageWaterNo8));
bwMixture1No8=imbinarize(rgb2gray(imageMixture1No8));
bwMixture2No8=imbinarize(rgb2gray(imageMixture2No8));
bwWaterNo9=imbinarize(rgb2gray(imageWaterNo9));
bwMixture1No9=imbinarize(rgb2gray(imageMixture1No9));
bwMixture2No9=imbinarize(rgb2gray(imageMixture2No9));
bwWaterNo10=imbinarize(rgb2gray(imageWaterNo10));
bwMixture1No10=imbinarize(rgb2gray(imageMixture1No10));
bwMixture2No10=imbinarize(rgb2gray(imageMixture2No10));
%Ë!ÓÄregionpropsËä·Ë¶±ð³ödroplets£-²çÇÖf¹y²épixelËÿÁ¿¼ÆËädropletsÃæ»y
graindataWaterNo1=regionprops('table',bwWaterNo1,'Area');
graindataOil1No1=regionprops('table',bwMixture1No1,'Area');
graindataOil2No1=regionprops('table',bwMixture2No1,'Area');
graindataWaterNo2=regionprops('table',bwWaterNo2,'Area');
graindataOil1No2=regionprops('table',bwMixture1No2,'Area');
graindataOil2No2=regionprops('table',bwMixture2No2,'Area');
graindataWaterNo3=regionprops('table',bwWaterNo3,'Area');
graindataOil1No3=regionprops('table',bwMixture1No3,'Area');
graindataOil2No3=regionprops('table',bwMixture2No3,'Area');
graindataWaterNo4=regionprops('table',bwWaterNo4,'Area');
graindataOil1No4=regionprops('table',bwMixture1No4,'Area');
graindataOil2No4=regionprops('table',bwMixture2No4,'Area');
graindataWaterNo5=regionprops('table',bwWaterNo5,'Area');
graindataOil1No5=regionprops('table',bwMixture1No5,'Area');
graindataOil2No5=regionprops('table',bwMixture2No5,'Area');
graindataWaterNo6=regionprops('table',bwWaterNo6,'Area');
graindataOil1No6=regionprops('table',bwMixture1No6,'Area');
graindataOil2No6=regionprops('table',bwMixture2No6,'Area');
graindataWaterNo7=regionprops('table',bwWaterNo7,'Area');
graindataOil1No7=regionprops('table',bwMixture1No7,'Area');
graindataOil2No7=regionprops('table',bwMixture2No7,'Area');
graindataWaterNo8=regionprops('table',bwWaterNo8,'Area');
graindataOil1No8=regionprops('table',bwMixture1No8,'Area');
graindataOil2No8=regionprops('table',bwMixture2No8,'Area');
graindataWaterNo9=regionprops('table',bwWaterNo9,'Area');
graindataOil1No9=regionprops('table',bwMixture1No9,'Area');
graindataOil2No9=regionprops('table',bwMixture2No9,'Area');
graindataWaterNo10=regionprops('table',bwWaterNo10,'Area');
graindataOil1No10=regionprops('table',bwMixture1No10,'Area');
graindataOil2No10=regionprops('table',bwMixture2No10,'Area');
%regionprops·µ»ØÖµ,ñË½Ïtable£-½«table,ñË½,ÃÏvector,ñË½½ graindataWater =
[graindataWaterNo1{::};graindataWaterNo2{::};graindataWaterNo3{::};graindataWaterNo4{::};graindataWaterNo5{::};graindataWaterNo6{::};graindataWaterNo7{::};graindataWaterNo8{::};graindataWaterNo9{::};graindataWaterNo10{::}];
graindataOil1 =
[graindataOil1No1{::};graindataOil1No2{::};graindataOil1No3{::};graindataOil1No4{::};graindataOil1No5{::};graindataOil1No6{::};graindataOil1No7{::};graindataOil1No8{::};graindataOil1No9{::};graindataOil1No10{::}];
graindataOil2 =
[graindataOil2No1{::};graindataOil2No2{::};graindataOil2No3{::};graindataOil2No4{::};graindataOil2No5{::};graindataOil2No6{::};graindataOil2No7{::};graindataOil2No8{::};graindataOil2No9{::};graindataOil2No10{::}];
% Total average
avgWater = sum(graindataWater)/length(graindataWater); avgOil1 = sum(graindataOil1)/length(graindataOil1); avgOil2 = sum(graindataOil2)/length(graindataOil2);
disp([avgWater avgOil1 avgOil2]);

```

## Visual Representation of Sprayed Ballistics

```

clear;

imageWaterNo1 = imread("Picture 1");
bwWaterNo1=imbinarize(rgb2gray(imageWaterNo1)); [row,col] = size(bwWaterNo1);
if rem(row,2) == 1
    slice_A = bwWaterNo1((row-13500+1)/2:(row-(row-13500+1)/2),:);
end
if rem(row,2) == 0

```

```

        slice_A = bwWaterNo1((row-13500)/2:((row-(row-13500)/2)-1),:);
    end
    if rem(col,2) == 1
        slice_A = slice_A(:,(col-9500+1)/2:(col-(col-9500+1)/2));
    end
    if rem(col,2) == 0
        slice_A = slice_A(:,(col-9500)/2:((col-(col-9500)/2)-1));
    end
    [row,col] = size(slice_A);

    area = []; area_row = [];
    for i_row = 1:500:row
        for i_col = 1:500:col
            matrix = slice_A(i_row:i_row+499, i_col:i_col+499); total_area = sum(matrix==0,'all');
            area_row = [area_row, total_area];
        end
    end
    area = [area;area_row]; area_row = [];
    area = area/47/47; surf(area);
    colormap(pink); shading interp;
    title('Bullsticity Illustration'); xlabel('Rows')
    ylabel('Columns')
    zlabel('Black Areas in aquare milimeter')

    %mesh(area);

```

end