A COMPARATIVE EVALUATION OF STORAGE STABILITY, MIXING CAPABILITY, PHYSICOCHEMICAL PROPERTIES, SPRAY BEHAVIORAL PATTERN AND DROPLET ADHESION CHARACTERISTICS OF NEW SUMITHION® 20% FLOWABLE AND FOUR FENITROTHION SPRAY FORMULATIONS CONTAINING ATLOX® 3409F AND TRITON® X-114 EMULSIFIERS

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ABSTRACT

A laboratory study was undertaken to investigate the suitability of the newly introduced fenitrothion formulation concentrate " New Sumithion® 20% Flowable " for field use as compared to four currently registered formulations containing Atlox® 3409F and Triton® X-114 emulsifiers, by studying their physical properties, stability, mixing capabilities, storage stability, spray atomization and behavioral pattern, evaporation characteristics and droplet adhesion on conifer needles. At the dosage rate of 210 g AI/1.5 L/ha, the spray mixture provided a thixotropic medium with optimum viscosity, evaporation characteristics and spray deposits, but the droplet size spectra were slightly towards the larger droplet sizes. As a result, the droplet densities were somewhat lower, although this is only a minor drawback which can easily be corrected by optimizing application systems and delivery conditions. The non-Newtonian behaviour of this spray mixture provided some noticeable advantages: exceptionally high stability, optimum viscosity, low evaporation rates, a narrow droplet size spectrum and a high deposit level on glass plates. On a comparative level with the other registered fenitrothion formulations containing Atlox 3409F and Triton X-114, the Sumithion spray mixture is easy to handle in the field conditions because of the readiness with which it flows under an external pressure and can be pumped easily.

The droplet adhesion study, conducted in order to understand the adhesion properties of spray droplets on conifer needles, indicated that under the humidity conditions of $<70\%$ there may be a need for the addition of about 4% oil which contributed to enhancement of droplet adhesion on spruce needles. This aspect, however, requires detailed investigation to optimize the type of oil and the concentration in the final spray mixture.

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INTRODUCTION

New Sumithion® 20% Flowable (Sumithion® 20F) is a formulation concentrate of fenitrothion, developed recently by Sumitomo Chemical Co., Ltd., Osaka, Japan. It is an emulsion containing 216 g AI (active ingredient) per litre of the formulation, and was prepared without adding any traditional emulsifier or cosolvent, as opposed to the currently used emulsion containing the Atlox® 3409F emulsifier and Dowanol® TPM cosolvent. The ingredients used in the formulation have imparted thixotropic properties to the medium, and as a result, the formulation exhibits high viscosity under storage conditions, a phenomenon which provides excellent storage stability of the formulation under a wide temperature range of -20°C to + 35°C. However, under an external shearing force, the viscosity is reduced drastically and therefore, during spray application the mixture flows very readily, providing a thin liquid medium which can atomize most efficiently resulting in a fine droplet spectrum.

The present study describes an investigation carried out to explore the physicochemical properties, stability considerations, mixing capabilities, spray behavioral pattern and droplet adhesion characteristics of the spray mixture of New Sumithion 20% flowable, prepared to provide a concentration of 140 g per litre. In addition four more spray mixtures were investigated. These are: fenitrothion with 1.5% Atlox 3409F and 1.5% Dowanol TPM; with 1.5% Atlox 3409F and 4.0% Dowanol TPM; with 5% Triton X-114 and 7% Triton X-114. These are chosen because they are already registered and listed on the label; and a comparative study with these registered formulations would provide information on the suitability of the New Sumithion 20F for field use, since the registered formulations have been shown to be acceptable for use under field conditions.

MATERIALS AND METHODS

1. Spray Formulations

The formulation concentrate and the other ingredients used in the study are listed in Table 1, along with the names of companies who supplied them. The percentage compositions of ingredients used in preparing the spray formulations are given in Table 2.

2. Mixing Capabilities of Spray Formulations

The four registered formulations FDA-3409-1.5, FDA-3409-4.0, FT-114-5.0 and FT-114-7.0 (Table 2), are aqueous emulsions. To ensure through mixing, the non-aqueous ingredients must be mixed first before adding to water. It is important to bear in mind that the emulsifier Triton X-114 must not be allowed to come into direct contact with water, otherwise gel formation will occur and cause serious mixing problems.

However, the New Sumithion 20F does not need any specific mixing sequence while mixing with water. The concentrate is so easy to mix with water in any order of mixing. Since no additional emulsifier is required, this formulation provides a great advantage for the ease of handling under field conditions.

3. Stability Considerations

The term stability refers to the tendency of the formulation to resist separation into its ingredients. Actual separation of the component phases can occur if stability is low. This phenomenon was studied when the spray formulations were left standing with no stirring or agitation and the findings are listed in Table 3. With gentle stirring however, phase separation may not be observed visually, but a reduction in viscosity can result due to changes in micelle formation and stability. These aspects were also studied at different temperatures and the findings are presented in Table 3.

TABLE 1

Pesticide, pesticide formulation, cosolvent, emulsifiers, and tracer dye used in the study, and the sources from which they were obtained

Name	Abbreviation used	Source
Sumithion® (fenitrothion technical)	Fe-tech	Sumitomo Chemical Co., Ltd. (Osaka, Japan)
Sumithion® 20F	Sumi-20F	Sumitomo Chemical Co., Ltd. (Osaka, Japan)
Atlox® 3409F	Atlo-3409	Atkemix (Brantford, Ont., Canada)
Triton® X-114	Trit-114	Rohm and Haas (Westhill, Ont., Canada)
Erio Acid Red	Erio-Red	Ciba Geigy (Dorval, Quebec, Canada)
Dowanol® TPM	Dow-TPM	Dow Chemical Canada (Sarnia, Ont., Canada)

4. Re-emulsification Capabilities

The term re-emulsifiability refers to the tendency of the separated phases to revert to the emulsion state having the same stability as that of the freshly prepared one. This aspect was studied after gentle and vigorous agitation. Findings are listed in Table 4.

5. Storage Stability of The Formulation Concentrate ' New Sumithion 20F '

The storage stability of the formulation concentrate was tested at -20°C to +5°C, and it was found to be excellent even at -20°C since no phase separation occurred. The concentrate is quite suitable to store under the cold winter temperatures of the Canadian climate. The storage stability of the active ingredient in the formulation concentrate was however not tested in

TABLE 2

Percentage composition of ingredients in spray formulations

Formulation abbreviation	Percentage composition (v/v)
Sumi-20F	New Sumithion® 20% Flowable undiluted 99.8/ Erio-Red 0.2*
Sumi-66.7	Sumi-20F 66.7 / water 33.1/ Erio-Red 0.2
FDA-3409-1.5	Fe-tech 11/ Atlo-3409 1.5/ Dow-TPM 1.5/ water 85.8/ Erio-Red 0.2
FDA-3409-4.0	Fe-tech 11/ Atlo-3409 1.5/ Dow-TPM 4.0/ water 83.3/ Erio-Red 0.2
FT-114-5.0	Fe-tech 11/ Trit-114 5.0/ water 83.8/ Erio-Red 0.2
FT-114-7.0	Fe-tech 11/ Trit-114 7.0/ water 81.8/ Erio-Red 0.2

* Concentration of the tracer dye added was 0.2 g/100 ml. At this level, the final volume of the formulation reached 100 ml.

the present study. It was hoped that the company would provide data in this respect. The freezing point was found to be -15° ± 3°C, and at -10°C it was a thick paste. However, even at -20°C, in spite of the frozen condition, the texture of the frozen material was just like "ice cream", and because of this, the concentrate can still be pumped easily without much difficulty. Upon thawing of the frozen material, no phase separation had occurred and the emulsion was still in tact and had excellent stability. These findings indicate that the formulation would provide excellent advantage for handling under field conditions.

6. Physical Properties of Ingredients

Viscosities of ingredients would provide an insight to their pumping and mixing capabilities. Therefore, viscosities were measured for the ingredients and listed in Table 5, along with their densities which are needed for calcu-

TABLE 3

Stability of spray formulations

Formulation abbreviation	Time (h) required for			
	Phase separation with no agitation		Reduction in viscosity by approx. 20% with agitation	
	5°- 15°C	20°- 25°C	5°- 15°C	20°- 25°C
Sumi-20F	Exceptionally stable at all temperatures			
Sumi-66.7	28 - 36	18 - 24	32 - 48	26 - 36
FDA-3409-1.5	2 - 3	1.5	10 - 15	6
FDA-3409-4.0	3 - 4	2.0	24 - 36	10
FT-114-5.0	3 - 4	2.0	24 - 36	15
FT-114-7.0	28 - 36	18 - 24	32 - 48	26 - 36

TABLE 4

Re-emulsification upon storage at 5°C to 15°C for up to four days

Formulation abbreviation	With gentle mixing	With good agitation	Resettling time (h) after vigorous shaking
Sumi-20F	No need to shake	No need to shake	Exceptionally stable
Sumi-66.7	Very good	Excellent	26 to 36
FDA-3409-1.5	Good	Very good	2.5 to 4.5
FDA-3409-4.0	Very good	Excellent	26 to 36
FT-114-5.0	Very good	Excellent	18 to 28
FT-114-7.0	Excellent	Excellent	48 to 72

lating the proportions of ingredients for preparing the spray formulations.

7. Physical Properties of Spray Formulations

Physical properties of spray formulations are known to influence the atomization charactetistics and consequently the spray deposition on targets. Therefore, viscosities, densities, surface tension values and evaporation characteristics of the spray formulations were studied and the data are listed in Tables 6, 8, 11, 12 and 13. The evaporation characteristics were expressed in terms of the residual weight percentages at time 't' and are presented in Figure 1.

8. Appearance, Solubility in Water, Pour Point, Freezing Point, Flash Point Data for Ingredients

These properties were also gathered for the ingredients used in the study, and listed in Table 7.

9. Spread Factor Data for Spray Formulations on Kromekote® Cards

Spread factor data are required for estimating spray droplet spectra obtained on Kromekote® cards following atomization. These were measured and the data are given in Tables 9 and 10.

10. Spray Atomization and Droplet Spectra on Kromekote Cards

Spray atomization was carried out in a spray chamber of dimensions of 430 cm x 90 cm x 305 cm. A spinning disc nozzle, mounted on a central rail to facilitate movement from end to end of the chamber, was calibrated to deliver the exact amount of 210 g AI/1.5 litre/ha, according to the floor area of the chamber. Spray was applied about 2.90 m above the Kromekote card/glass plate units which were placed about 15 cm above the floor level of the chamber. Four of such units were used for each formulation, and droplets were allowed to settle on the sampling units for 15 min before they were removed from the chamber. Spray

TABLE 5

Viscosity and density values of ingredients used in the spray formulations

Ingredient abbreviation	Physical property at temperatures of				
	5°C	10°C	15°C	20°C	25°C
			Density (g/ml)*		
Fe-tech	1.336	1.328	1.322	1.318	1.315
Dow-TPM	0.996	0.992	0.988	0.983	0.979
Atlo-3409	1.042	1.037	1.031	1.026	1.022
Trit-114	1.065	1.059	1.054	1.050	1.047
Water	1.000	0.9997	0.9991	0.9982	0.9971
			Viscosity (cp)**		
Fe-tech	126	82.5	53.4	40.0	27.7
Dow-TPM	20.2	16.3	13.1	10.8	9.27
Atlo-3409	Paste	5660	443	329	217
Trit-114	1470	974	600	380	204
Water	1.52	1.31	1.14	1.05	0.894

* Measured using density bottle

** Measured using Ostwald viscometer

TABLE 6

Viscosity and density values of spray formulations

Formulation abbreviation	Physical property at temperatures of				
	5°C	10°C	15°C	20°C	25°C
			Density (g/ml)*		
Sumi-20F	1.088	1.087	1.085	1.082	1.080
Sumi-66.7	1.049	1.048	1.047	1.045	1.044
FDA-3409-1.5	1.012	1.011	1.010	1.009	1.008
FDA-3409-4.0	1.041	1.040	1.038	1.037	1.035
FT-114-5.0	1.038	1.037	1.036	1.035	1.034
FT-114-7.0	1.044	1.043	1.042	1.040	1.038
			Viscosity (cp)**		
Sumi-20F	570	533	419	333	256
Sumi-66.7	95.3	76.4	58.3	50.9	38.9
FDA-3409-1.5	2.89	2.53	2.13	1.80	1.49
FDA-3409-4.0	18.7	12.2	11.1	7.57	6.14
FT-114-5.0	6.50	5.33	4.53	8.50	5.30
FT-114-7.0	25.6	15.7	24.6	47.8	22.2

* Measured using density bottle.

** Measured using Ostwald viscometer.

TABLE 7

Properties of ingredients and Pesticide formulation

Ingredient abbreviation	Appearance & colour	Solubility in water	Product nature	Pour point/ freezing point	Flash point
Fe-tech	Clear brownish yellow liquid	Insoluble	Single product	Below 0°C	----
Dow-TPM	Clear thin colorless liquid	Soluble	Single product	Below 0°C	110°C
Atlo-3409	Cloudy amber-colored liquid	Soluble	Formulated product	Pour point 4°C Freezes at 3°C	12.2°C
Trit-114	Clear colorless liquid	Soluble	Single product	Pour point -9°C	>150°C
Water	Clear colorless liquid	------	Single product	Freezes at 0°C	-----
Sumi-20F	Opaque pale-yellow emulsion	Miscible	Formulated product	Pour point -5°C Freezes at -8°C	-----

TABLE 8

Surface tension values of spray formulations

Formulation abbreviation	Surface tension (dyne/cm) at °C of				
	5	10	15	20	25
Sumi-20F	44.2	43.8	43.3	42.9	42.5
Sumi-66.7	47.9	47.5	47.0	46.3	45.8
FDA-3409-1.5	28.9	28.4	28.0	27.6	27.2
FDA-3409-4.0	33.6	33.0	32.3	31.6	31.0
FT-114-5.0	35.6	34.6	33.8	33.0	32.2
FT-114-7.0	33.6	32.9	31.9	30.6	29.5

TABLE 9

Spread factor data for Sumithion flowable formulations

Sumi-20F			Sumi-66.7		
Stain diam. (µm)	Droplet diam. (µm)	Spread factor	Stain diam. (µm)	Droplet diam. (µm)	Spread factor
120	75	1.60	165	86	1.92
160	98	1.63	190	95	2.00
210	122	1.72	240	120	2.00
260	145	1.79	345	148	2.33
389	210	1.85	450	210	2.14
523	275	1.90	540	271	1.99
588	302	1.95	630	300	2.10
740	370	2.00	810	360	2.25
Linear regression equation:			Linear regression equation:		
$d = 21.4 + 0.477\ D\ (R^2 = 99.9\%)$			$d = 11.0 + 0.446\ D\ (R^2 = 98.8\%)$		

TABLE 10

Spread factor data for fenitrothion spray formulations

Stain diam. (µm)	Droplet diam. (µm)	Spread factor	Stain diam. (µm)	Droplet diam. (µm)	Spread factor
FDA-3409-1.5			FDA-3409-4.0		
175	55	3.18	220	65	3.38
230	73	3.15	370	105	3.52
300	95	3.16	565	158	3.58
455	145	3.14	735	205	3.59
672	210	3.20	995	275	3.62
870	270	3.22	1145	315	3.63
1070	330	3.24	1230	340	3.62
Linear regression equation:			Linear regression equation:		
$d = 2.92 + 0.307\ D\ (R^2 = 100\%)$			$d = 4.80 + 0.272\ D\ (R^2 = 100\%)$		
FT-114-5.0			FT-114-7.0		
180	75	2.40	165	65	2.54
240	98	2.45	200	78	2.56
315	125	2.52	255	93	2.74
400	148	2.70	335	115	2.91
490	175	2.80	440	145	3.03
615	215	2.86	650	210	3.10
780	265	2.94	815	255	3.20
880	293	3.00	Linear regression equation:		
Linear regression equation:			$d = 18.0 + 0.292\ D\ (R^2 = 100\%)$		
$d = 23.9 + 0.308\ D\ (R^2 = 99.9\%)$					

Table 11. Evaporation characteristics of liquid film. Temp. °C = 22 ± 1.5. Relative humidity = 45 ± 2%.

Time (min)	Wt. of Liquid Film 1st	2nd	3rd	Residual Wt. % 1st	2nd	3rd	Mean residual weight %	S.D.
				Sumi-20F				
0	0.3800	0.3755	0.3990	100	100	100	100	0.00
2	0.3671	0.3639	0.3878	96.6	96.9	97.2	96.9	0.30
4	0.3535	0.3537	0.3759	93.0	94.2	94.2	93.8	0.69
6	0.3400	0.3323	0.3511	89.5	88.5	88.0	88.7	0.76
10	0.3203	0.3132	0.3372	84.3	83.4	84.5	84.1	0.59
20	0.2633	0.2576	0.2785	69.3	68.6	69.8	69.2	0.60
40	0.2151	0.2148	0.2202	56.6	57.2	55.2	56.3	1.03
60	0.1558	0.1585	0.1648	41.0	42.2	41.3	41.5	0.62
120	0.1292	0.1314	0.1369	34.0	35.0	34.3	34.4	0.51
180	0.1170	0.1167	0.1269	30.8	31.1	31.8	31.2	0.52
				Sumi-66.7				
0	0.3887	0.4374	0.4856	100	100	100	100	0.00
2	0.3758	0.4241	0.4723	96.7	97.0	97.3	97.0	0.29
4	0.3630	0.4117	0.4595	93.4	94.1	94.6	94.1	0.62
6	0.3499	0.3986	0.4474	90.0	91.1	92.1	91.1	1.06
10	0.3267	0.3735	0.4208	84.1	85.4	86.7	85.4	1.31
20	0.2294	0.2784	0.3160	59.0	63.7	65.1	62.6	3.16
40	0.1797	0.2233	0.2568	46.2	51.1	52.9	50.1	3.44
60	0.1205	0.1516	0.1761	31.0	34.7	36.3	34.0	2.69
120	0.0937	0.1056	0.1127	24.1	24.2	23.2	23.8	0.53
180	0.0848	0.0987	0.1106	21.8	22.6	22.8	22.4	0.50

Table 12. Evaporation characteristics of liquid film. Temp. °C = 22 ± 1.5. Relative humidity = 45 ± 2%.

Time (min)	Wt. of Liquid Film 1st	2nd	3rd	Residual Wt. % 1st	2nd	3rd	Mean residual weight %	S.D.
				FDA-3409-1.5				
0	0.2150	0.2205	0.2195	100	100	100	100	0.00
2	0.2002	0.2062	0.2060	93.1	93.6	93.9	93.5	0.37
4	0.1871	0.1917	0.1904	87.0	87.0	86.8	86.9	0.14
6	0.1742	0.1781	0.1775	81.0	80.8	80.9	80.9	0.14
10	0.1488	0.1516	0.1515	69.2	68.8	69.0	69.0	0.23
20	0.0866	0.0894	0.0894	40.3	40.6	40.8	40.5	0.24
40	0.0286	0.0304	0.0301	13.3	13.8	13.7	13.6	0.26
60	0.0283	0.0304	0.0301	13.3	13.8	13.7	13.6	0.26
120	0.0283	0.0304	0.0301	13.3	13.8	13.7	13.6	0.26
180	0.0283	0.0304	0.0301	13.3	13.8	13.7	13.6	0.26
				FDA-3409-4.0				
0	0.2092	0.2505	0.2604	100	100	100	100	0.00
2	0.1968	0.2380	0.2490	94.1	95.0	95.6	94.9	0.78
4	0.1842	0.2252	0.2370	88.1	89.9	91.0	89.7	1.50
6	0.1728	0.2128	0.2237	82.6	85.0	85.9	84.5	1.70
10	0.1471	0.1857	0.1964	70.3	74.1	75.4	73.3	2.65
20	0.0900	0.1238	0.1362	43.0	49.4	52.3	48.3	4.75
40	0.0307	0.0388	0.0401	14.7	15.5	15.4	15.2	0.44
60	0.0304	0.0332	0.0341	14.5	13.3	13.1	13.6	0.79
120	0.0294	0.0319	0.0328	14.1	12.7	12.6	13.1	0.80
180	0.0285	0.0310	0.0321	13.6	12.4	12.3	12.8	0.73

Table 13. Evaporation characteristics of liquid film. Temp. °C = 22 ± 1.5. Relative humidity = 45 ± 2%.

Time (min)	Wt. of Liquid Film 1st	2nd	3rd	Residual Wt. % 1st	2nd	3rd	Mean residual weight %	S.D.
				FT-114-5.0				
0	0.3027	0.3125	0.3296	100	100	100	100	0.00
2	0.2888	0.2978	0.3152	95.4	95.3	95.6	95.5	0.17
4	0.2750	0.2840	0.3016	90.9	90.9	91.5	91.1	0.37
6	0.2614	0.2703	0.2879	86.4	86.5	87.4	86.7	0.54
10	0.2348	0.2442	0.2630	77.6	78.1	79.8	78.5	1.15
20	0.1716	0.1812	0.2008	56.7	58.0	60.9	58.5	2.17
40	0.0800	0.0820	0.0993	26.4	26.2	30.1	27.6	2.19
60	0.0558	0.0564	0.0584	18.4	18.1	17.7	18.1	0.36
120	0.0554	0.0562	0.0581	18.3	18.0	17.6	18.0	0.34
180	0.0554	0.0562	0.0580	18.3	18.0	17.6	18.0	0.35
				FT-114-7.0				
0	0.3335	0.3410	0.3326	100	100	100	100	0.00
2	0.3172	0.3250	0.3166	95.1	95.3	95.2	95.2	0.10
4	0.2990	0.3097	0.3016	89.7	90.8	90.7	90.4	0.63
6	0.2827	0.2962	0.2872	84.8	86.9	86.4	86.0	1.09
10	0.2539	0.2676	0.2592	76.1	78.5	77.9	77.5	1.23
20	0.1851	0.2018	0.1921	55.5	59.2	57.8	57.5	1.86
40	0.0910	0.1002	0.0909	27.3	29.4	27.3	28.0	1.20
60	0.0724	0.0751	0.0710	21.7	22.0	21.4	21.7	0.34
120	0.0723	0.0734	0.0710	21.7	21.5	21.4	21.5	0.17
180	0.0718	0.0734	0.0710	21.5	21.5	21.4	21.5	0.10

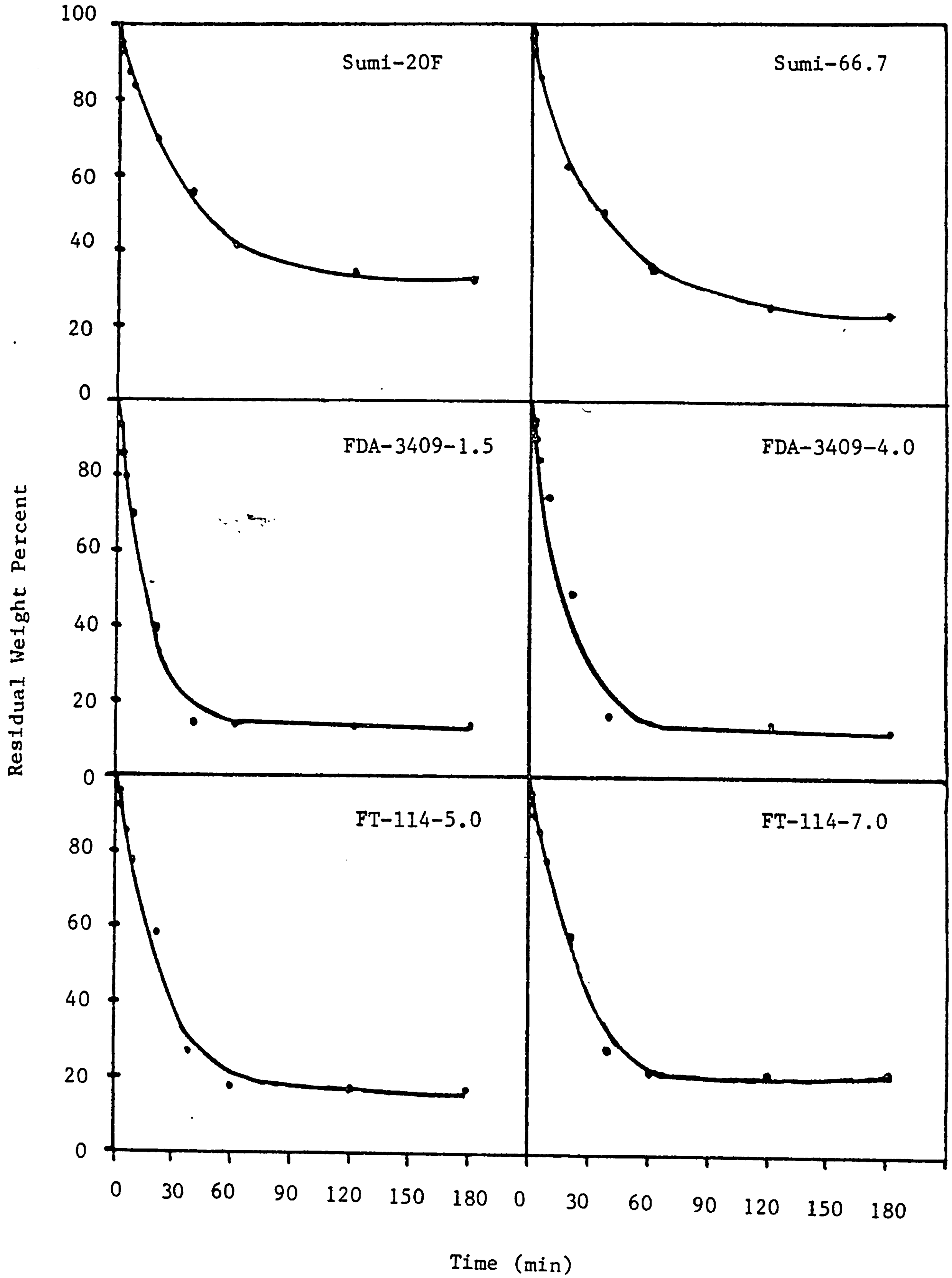

Fig. 1. Variation of Residual Weight Percent with Time 't'

Table 14. Viscosity-temperature relationships of ingredients and spray formulations

Liquid Abbreviation	Linear Regression Equation	R^2 (%)	Intercept* 'I'	Slope* 'S'	D@	E@ (kJ/mol.K)
			Ingredients			
Fe-tech	$\log \eta = -7.64 + 2704\ (1/T)$	99.7	-7.64	2704	0.00×10^{-5}	51.8
Dow-TPM	$\log \eta = -3.80 + 1419\ (1/T)$	99.8	-3.80	1419	16.0	27.2
Atlo-3409	$\log \eta = -22.8 + 7437\ (1/T)$	79.3	-22.8	7437	0.00	142
Trit-114	$\log \eta = -9.44 + 3514\ (1/T)$	99.1	-9.44	3514	0.00	67.3
Water	$\log \eta = -3.14 + 923\ (1/T)$	99.2	-3.14	923	72.4	17.7
			Spray formulations			
Sumi-20F	$\log \eta = -2.55 + 1485\ (1/T)$	96.4	-2.55	1485	282	28.4
Sumi-66.7	$\log \eta = -3.71 + 1582\ (1/T)$	99.3	-3.71	1582	19.5	30.3
FDA-3409-1.5	$\log \eta = -3.83 + 1197\ (1/T)$	99.4	-3.83	1197	14.8	22.9
FDA-3409-4.0	$\log \eta = -5.75 + 1948\ (1/T)$	97.4	-5.75	1948	0.18	37.3
FT-114-5.0	$\log \eta = -3.71 + 1256\ (1/T)$	99.8	-3.71	1256	19.5	24.1

* The intercept 'I' and slope 'S' values are from the linear equation $\log \eta = I + S\ (1/T)$

@ The constants 'D' and 'E' are from the Arrhenius equation $\eta = D\ e^{E/RT}$

Table 15. Exponential Decay Equation* for Evaporation Characteristics of Spray Formulations, Regression Coefficients and Half-lives of Decay.

Liquid abbreviation	Regression Equation	R^2 (%)	A	B	C	$T_{½}$ (min)**
Sumi-20F	$Y = 30.6 + 69.4\, e^{-0.0258\, t}$	99.4	30.6	69.4	0.0258	26.9
Sumi-66.7	$Y = 20.8 + 79.2\, e^{-0.0235\, t}$	97.6	20.8	79.2	0.0235	29.5
FDA-3409-1.5	$Y = 15.0 + 85.0\, e^{-0.0894\, t}$	94.1	15.0	85.0	0.0894	7.75
FDA-3409-4.0	$Y = 16.0 + 84.0\, e^{-0.0484\, t}$	98.7	16.0	84.0	0.0484	14.3
FT-114-5.0	$Y = 19.5 + 80.5\, e^{-0.0569\, t}$	96.4	19.5	80.5	0.0569	12.2
FT-114-7.0	$Y = 21.5 + 78.5\, e^{-0.0615\, t}$	96.4	21.5	78.5	0.0615	11.3

* The decay equation: $Y = A + B\, e^{-C\, t}$ represents the exponential decrease of the residual weight percent with time 't'

** The half-life ' $T_{½}$ ' represents the time required for the volatile portion of the spray formulations to reach 50% of their initial values.

application was made in triplicate, to provide a total of 12 cards for each formulation. The droplet stains were counted using a dissecting microscope at 40X, 100X and 200X magnifications. The data obtained from the 12 cards were grouped according to diameter classes and the cumulative droplet and volume distribution in each diameter class was evaluated using a correction spread factor (SF) for converting stain sizes on the cards into the corresponding aerodynamic droplet sizes. To obtain SF, droplets of uniform diameter were produced and were allowed to impact on Kromekote cards and on glass fibres, and SF was calculated using SF = D/d, where 'D' is the stain diameter on the card and 'd' the droplet diameter on the fibre. Figures 2, 4, 6, 8 and 10 present the frequency and volume distribution percentages as histograms, according to the diameter classes chosen. Figures 3, 5, 7, 9 and 11 present the cumulative number and volume distribution percentages from which, the number and volume median diameters (NMD and VMD respectively) were calculated. Table 16 presents the NMD and VMD values along with the maximum diameter (D_{max}) observed for each formulation.

11. Deposit Assessment on Glass Plates

The spray deposits on the glass plates were eluted using methanol as the solvent, followed by ethyl acetate to remove any traces of fenitrothion remaining. The extracts were flash-evaporated, reconstituted in a suitable solvent and analysed by GLC technique as described by Sundaram and Nott (1985). From the concentration of fenitrothion measured, the volume of the spray deposited per unit area of the glass plates was calculated in litre/ha units (Table 16). From this, the percentage of fenitrothion deposited was estimated.

12. Droplet Impaction, Retention and Adhesion Characteristics on Spruce Needles

Small potted seedlings of white spruce were sprayed in the spray chamber mentioned earlier. Microscopic analysis of the droplets indicated

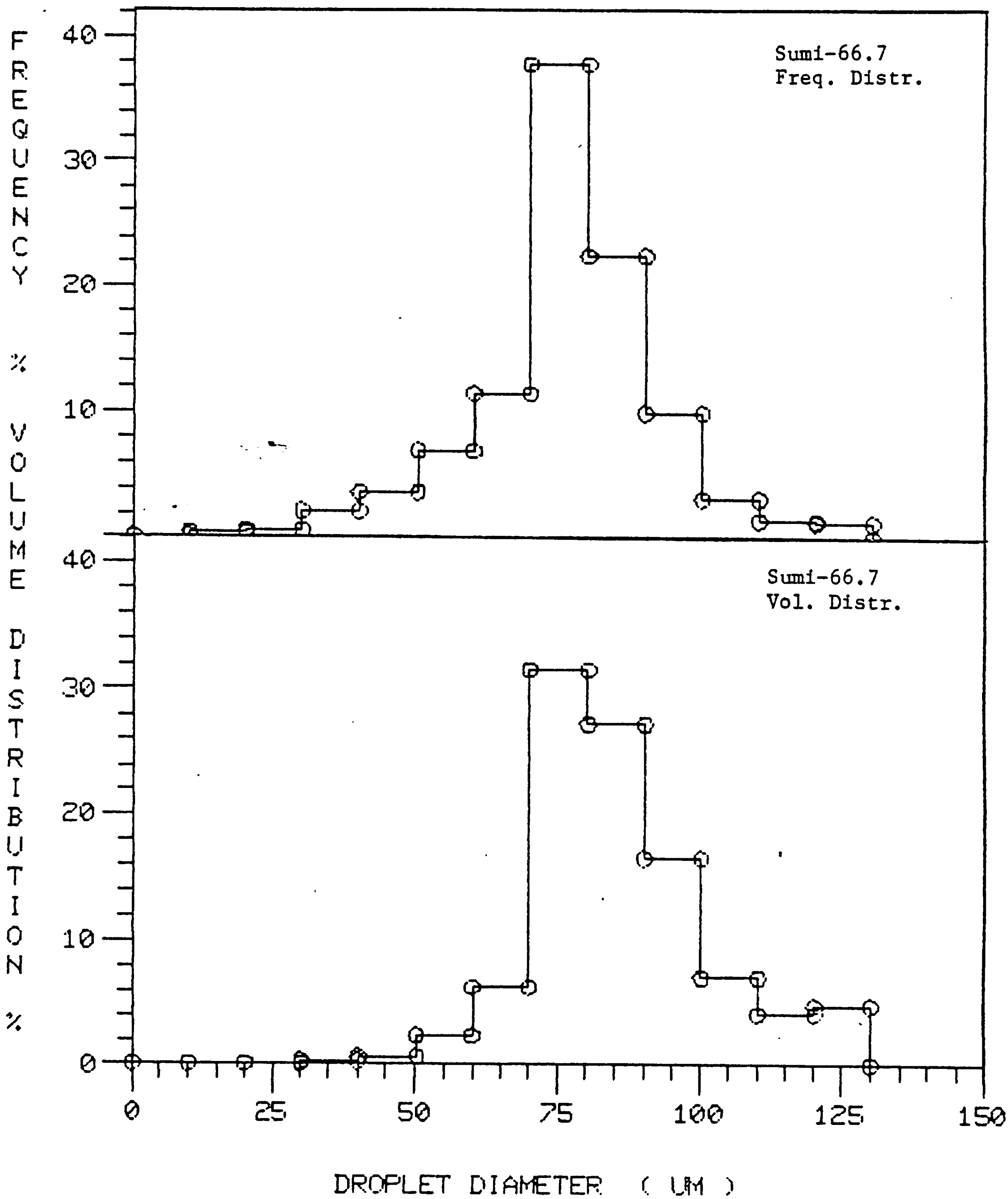
FREQUENCY % VOLUME DISTRIBUTION %
40
30
20
10
Sumi-66.7
Freq. Distr.
40
30
20
10
0
Sumi-66.7
Vol. Distr.
0
25
50
75
100
125
150
DROPLET DIAMETER (UM)

Figure 2.

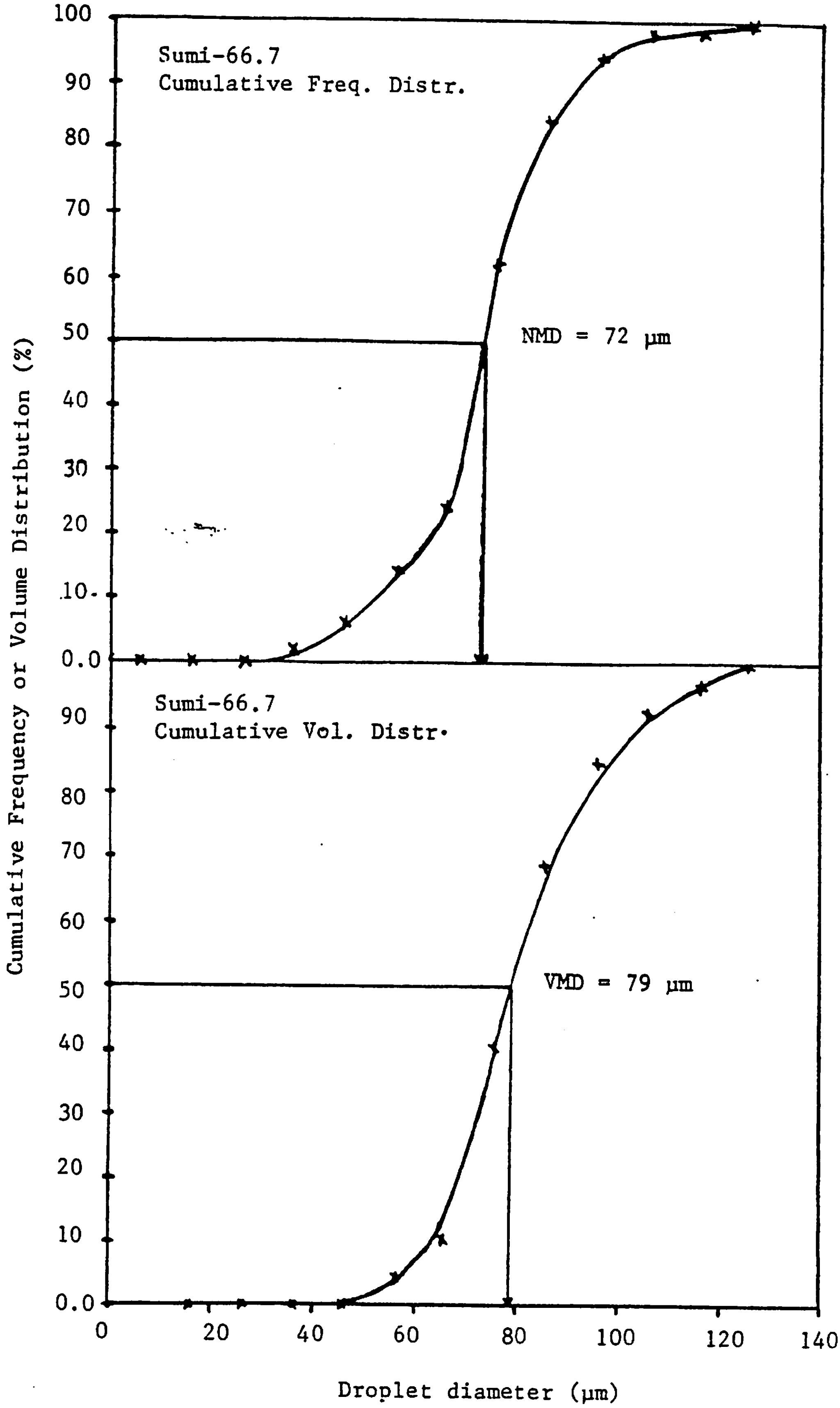
Sumi-66.7
Cumulative Freq. Distr.
NMD = 72 μm
Sumi-66.7
Cumulative Vol. Distr.
VMD = 79 μm
Cumulative Frequency or Volume Distribution (%)
Droplet diameter (μm)
0
20
40
60
80
100
120
140
0.0
10
20
30
40
50
60
70
80
90
100

Figure 3.

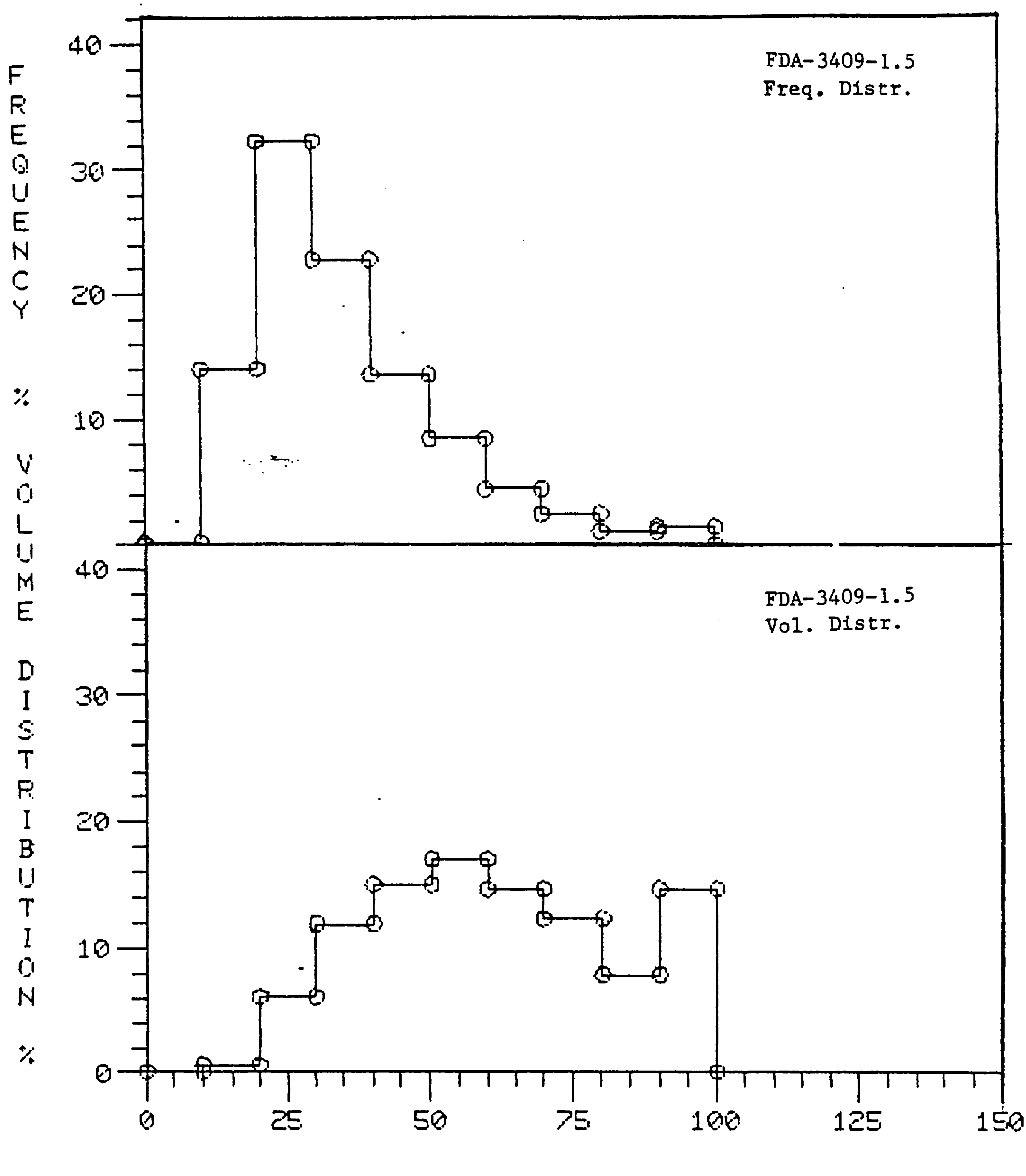
FDA-3409-1.5
Freq. Distr.
FDA-3409-1.5
Vol. Distr.
FREQUENCY % VOLUME DISTRIBUTION %
40
30
20
10
40
30
20
10
0
0
25
50
75
100
125
150
DROPLET DIAMETER (UM)

Figure 4.

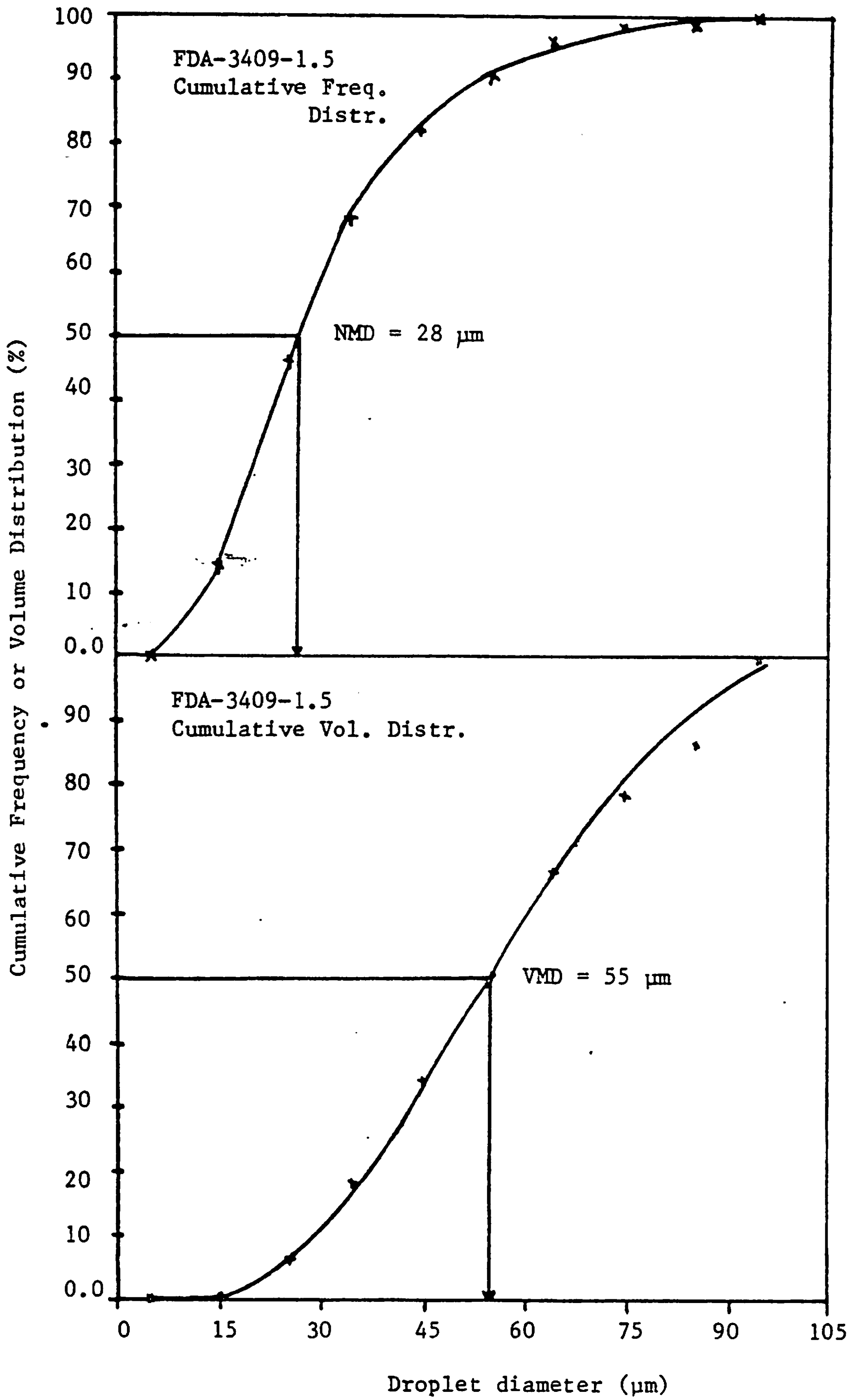
100
90
80
70
60
50
40
30
20
10
0.0
FDA-3409-1.5
Cumulative Freq.
Distr.
NMD = 28 μm
FDA-3409-1.5
Cumulative Vol. Distr.
VMD = 55 μm
Cumulative Frequency or Volume Distribution (%)
0
15
30
45
60
75
90
105
Droplet diameter (μm)

Figure 5.

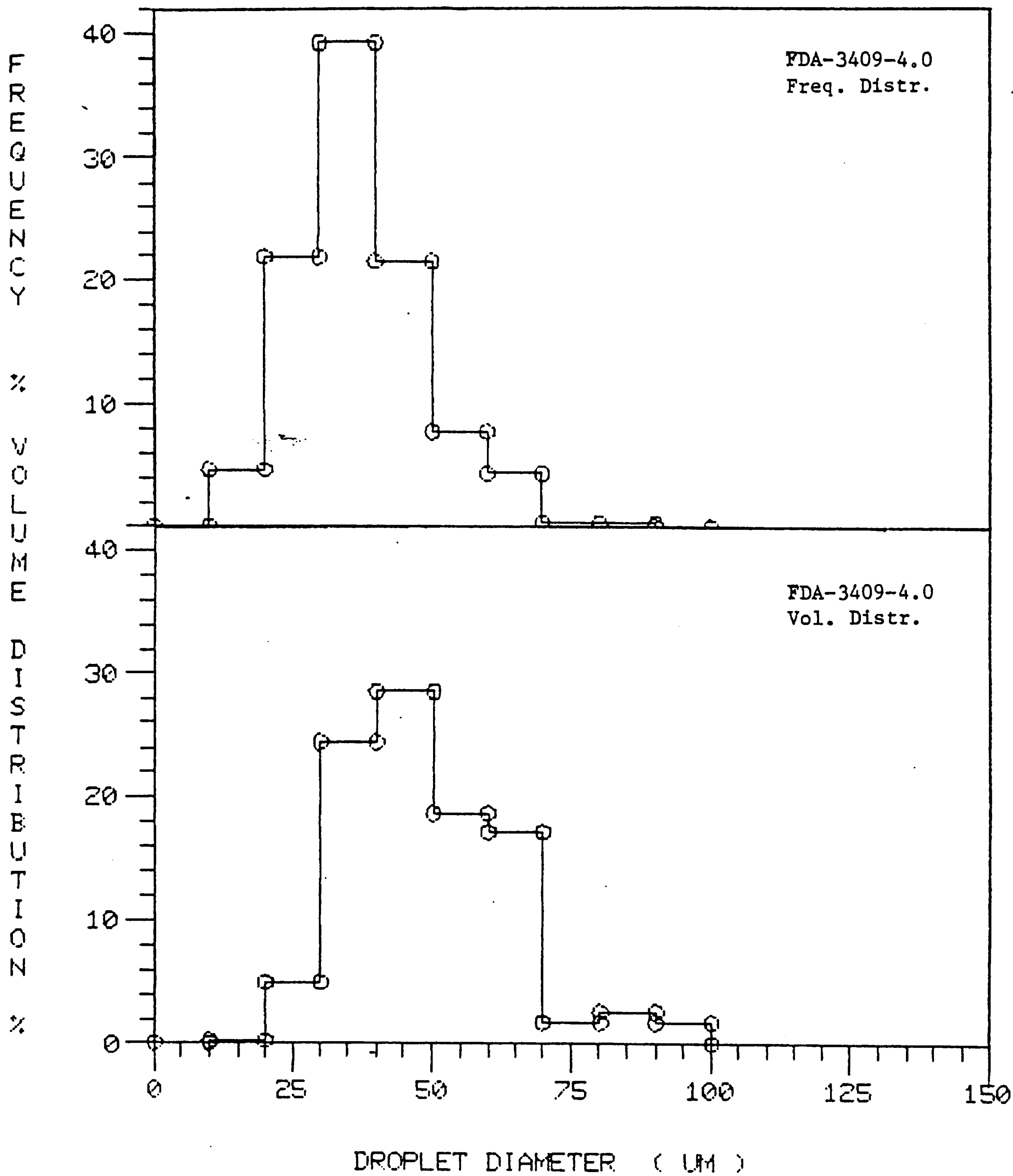
FREQUENCY % VOLUME DISTRIBUTION %
40
30
20
10
FDA-3409-4.0
Freq. Distr.
40
30
20
10
0
FDA-3409-4.0
Vol. Distr.
0
25
50
75
100
125
150
DROPLET DIAMETER (UM)

Figure 6.

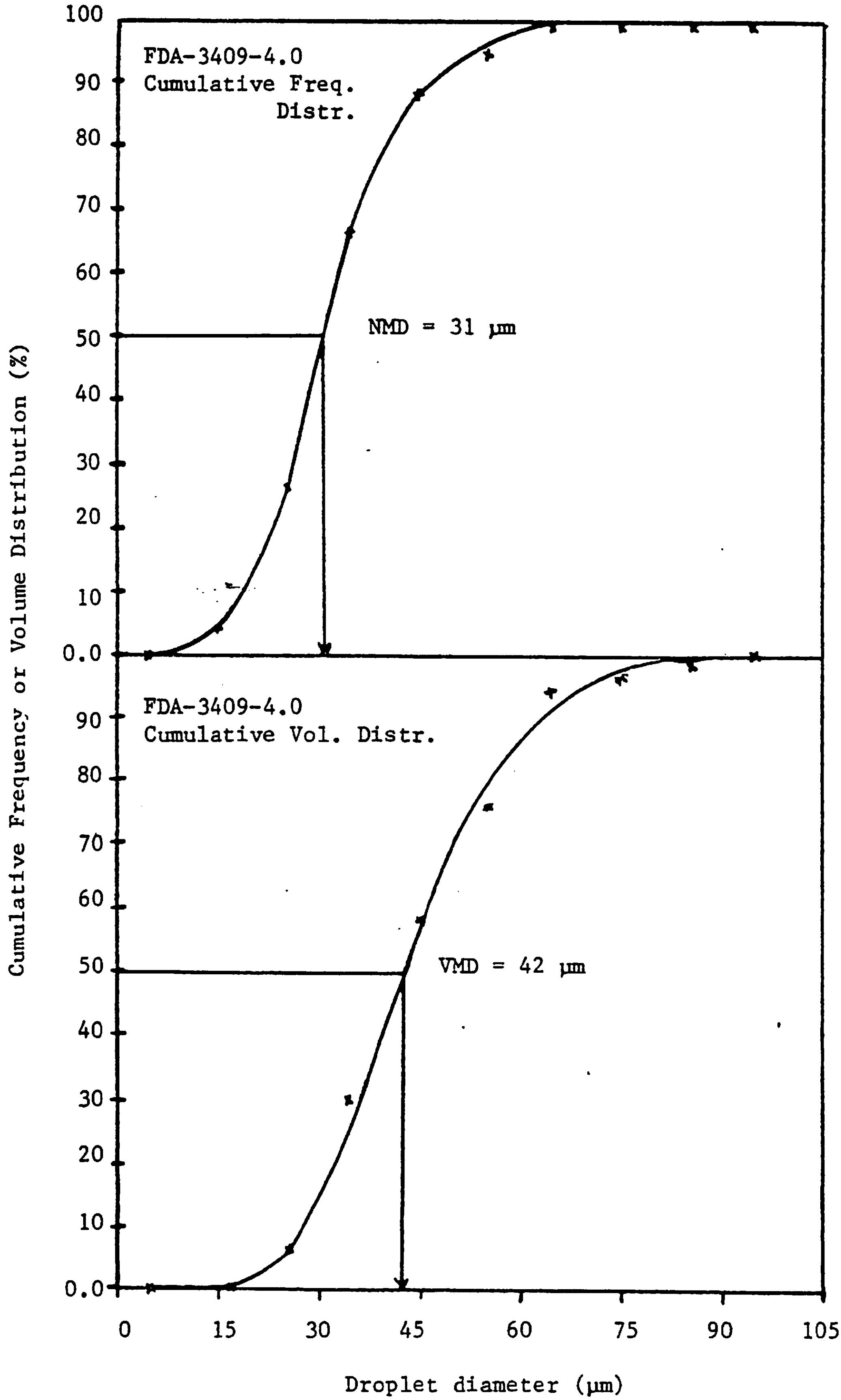
FDA-3409-4.0
Cumulative Freq.
Distr.
NMD = 31 μm
FDA-3409-4.0
Cumulative Vol. Distr.
VMD = 42 μm
Cumulative Frequency or Volume Distribution (%)
100
90
80
70
60
50
40
30
20
10
0.0
0
15
30
45
60
75
90
105
Droplet diameter (μm)

Figure 7.

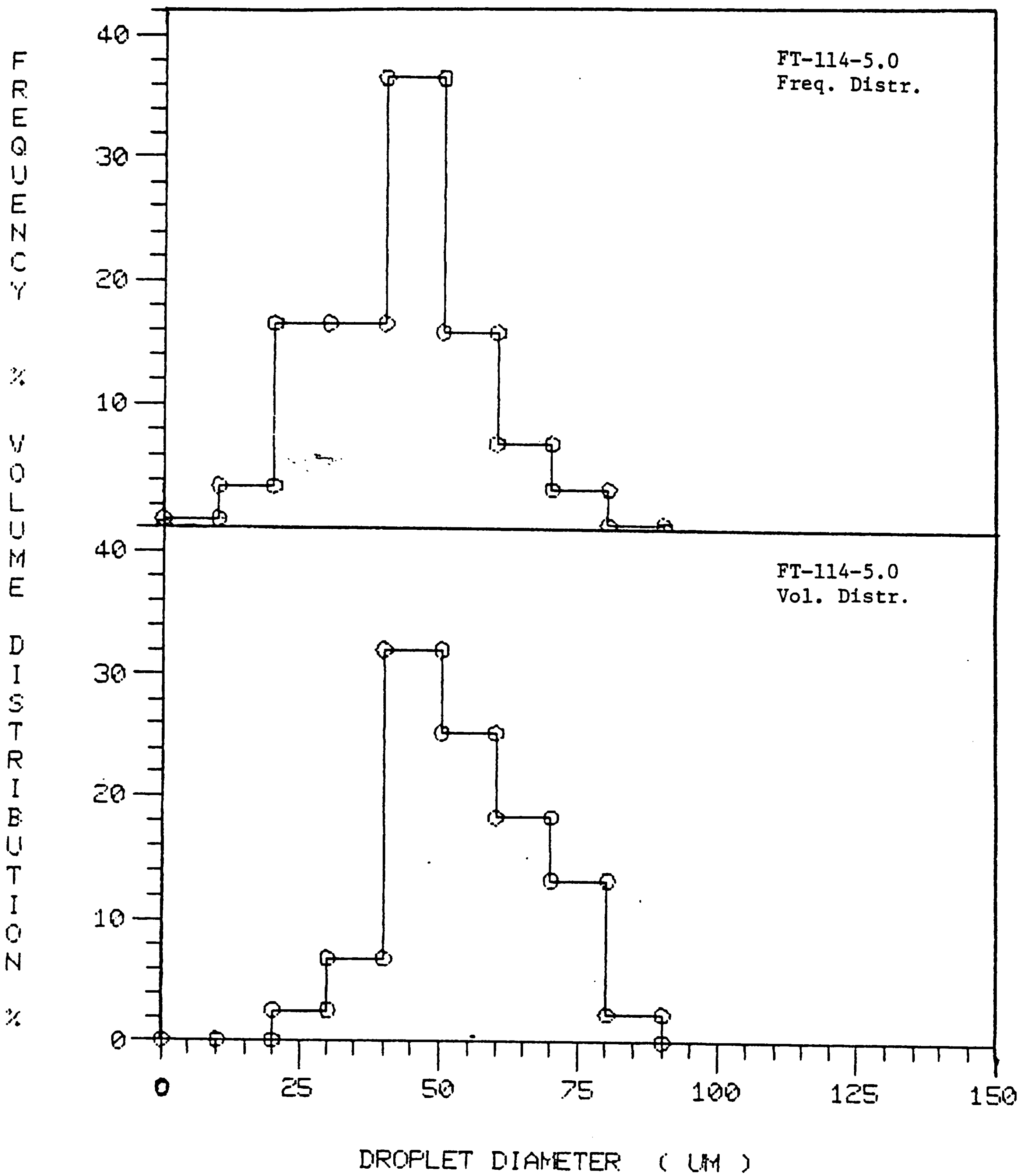
FT-114-5.0
Freq. Distr.
FT-114-5.0
Vol. Distr.
FREQUENCY % VOLUME DISTRIBUTION %
40
30
20
10
0
0
25
50
75
100
125
150
DROPLET DIAMETER (UM)

Figure 8

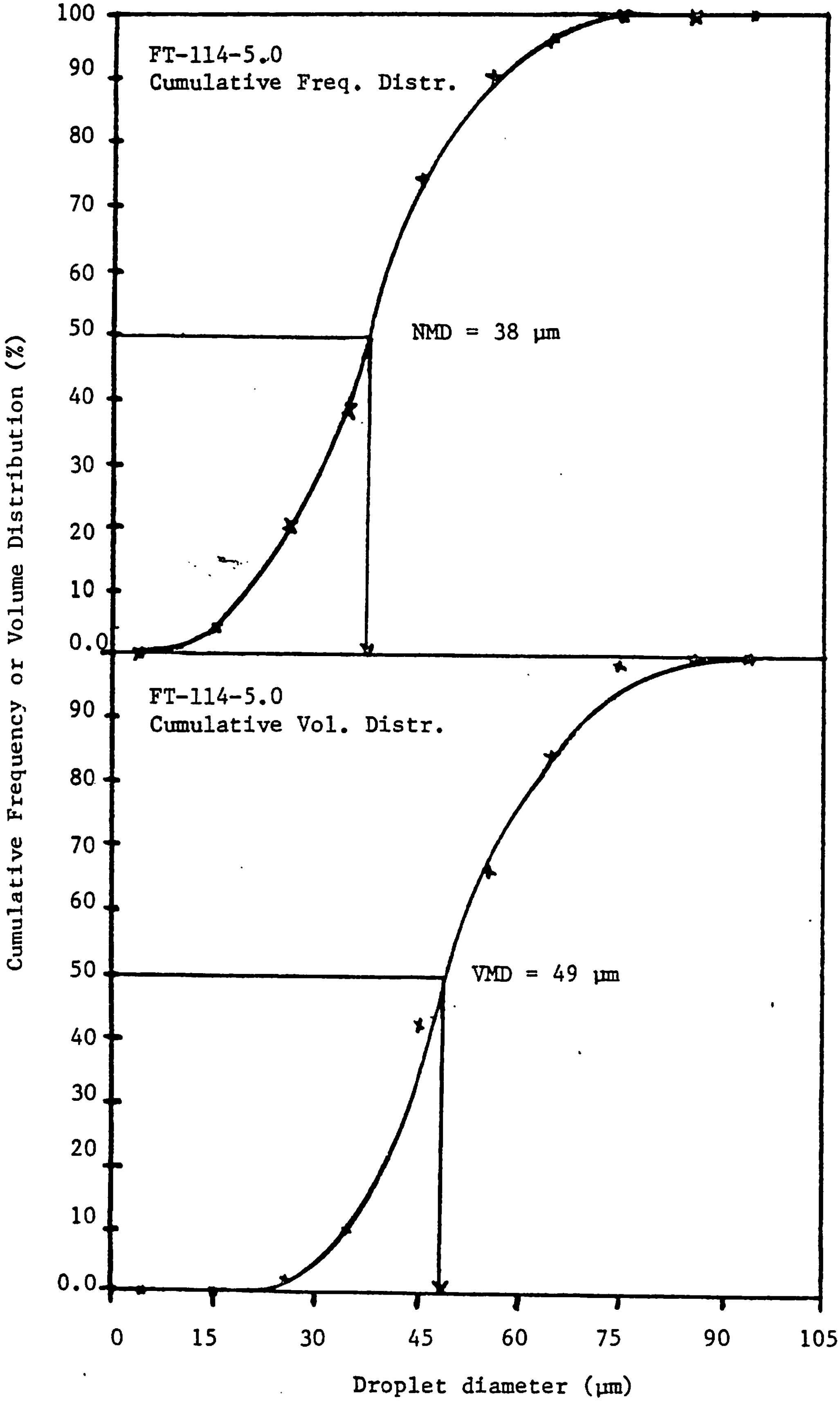
FT-114-5.0
Cumulative Freq. Distr.
NMD = 38 µm
FT-114-5.0
Cumulative Vol. Distr.
VMD = 49 µm
Cumulative Frequency or Volume Distribution (%)
100
90
80
70
60
50
40
30
20
10
0.0
0
15
30
45
60
75
90
105
Droplet diameter (µm)

Figure 9.

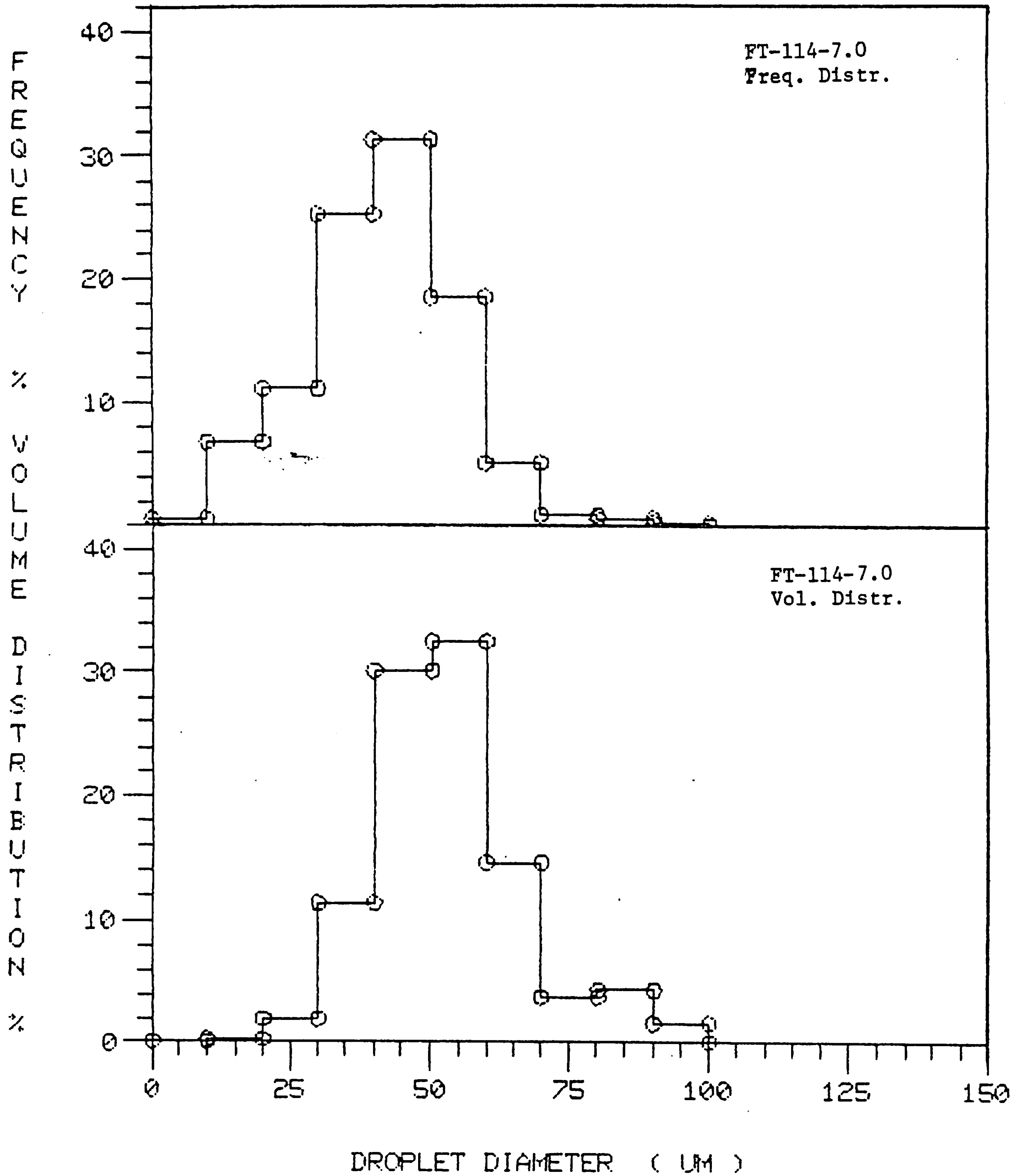
FT-114-7.0
Freq. Distr.
FT-114-7.0
Vol. Distr.
FREQUENCY % VOLUME DISTRIBUTION %
40
30
20
10
0
0
25
50
75
100
125
150
DROPLET DIAMETER (UM)

Figure 10

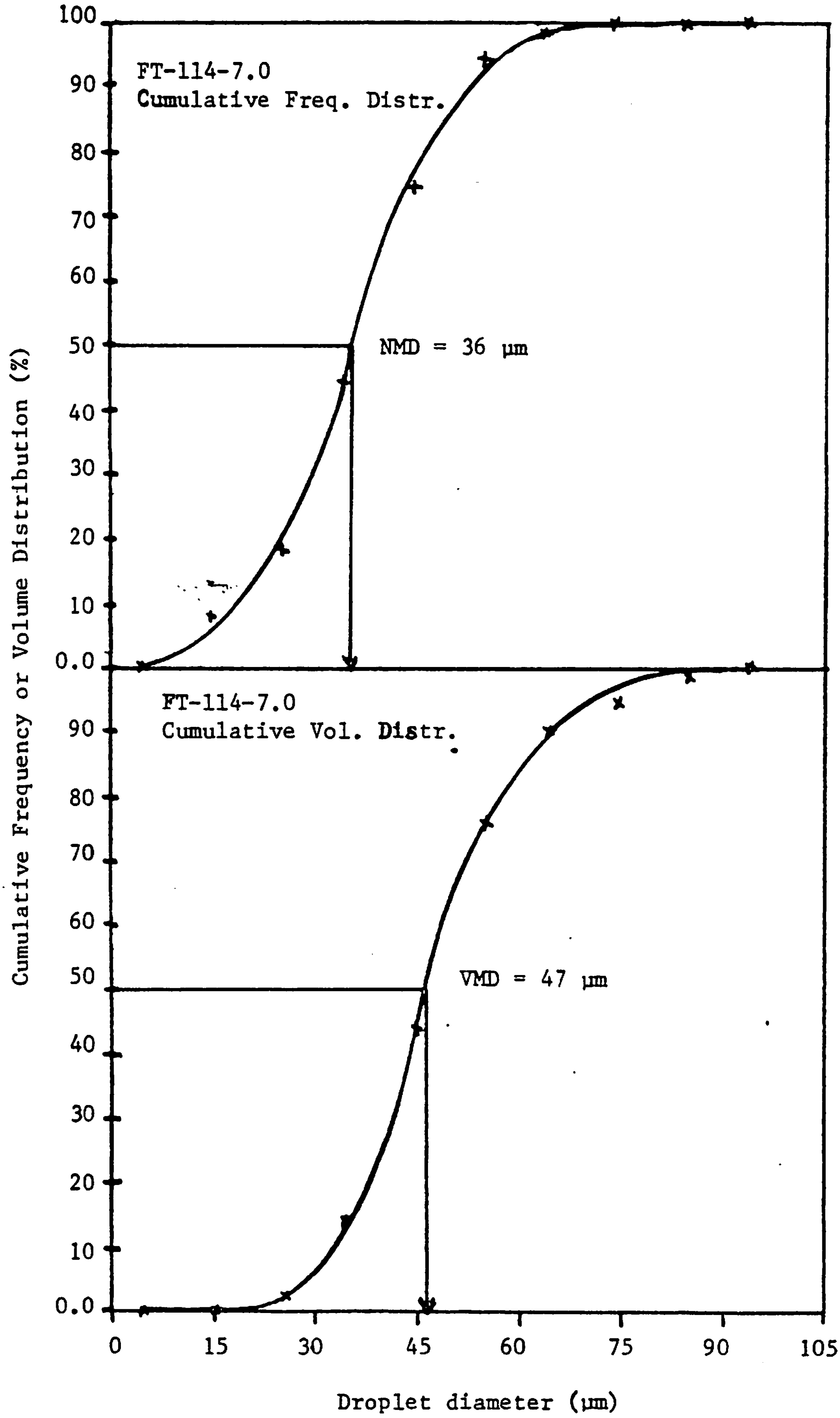
FT-114-7.0
Cumulative Freq. Distr.
NMD = 36 µm
FT-114-7.0
Cumulative Vol. Distr.
VMD = 47 µm
Cumulative Frequency or Volume Distribution (%)
100
90
80
70
60
50
40
30
20
10
0.0
0
15
30
45
60
75
90
105
Droplet diameter (µm)

Figure 11

that all formulations showed comparable droplet impaction characteristics on the conifer needles.

With respect to droplet retention, all formulations except the Sumi-66.7 exhibited cuticular absorption of droplets. However, the droplets of Sumi-66.7 were retained on the surface of the needles without undergoing penetration for a long period of time, even after 30 days post-treatment. This finding indicates that the ingredients in the formulation caused the drying of the droplets from the surface of the needles, thus preventing from cuticular penetration. Because of this, the adhesion characteristics of the droplets were impaired to some extent, especially under low humidity conditions of the ambient air, viz., below ca 60 to 70%. Under these low humidity values, the surface of the conifer needles were very dry and this facilitated the lack of adhesion of droplets. However, if the seedling was stored in moist environment until the time of spray application, and if the spray was applied at low humidity conditions (say, as low as 50%), then the droplets did not show lack of adhesion because the needle surface was moist and assisted in adhesion.

The present study was focussed on the ability of the droplet to show physical adhesion only. No attempts were made to study the rainfastness of these droplets. To improve adhesion under dry weather conditions, about 3 to 4 percent v/v of three oils was investigated, viz., Cyclosol® 63, Sunspray® 6N and canola oil. Droplet adhesion was markedly improved by the addition of the oil matrix. However, this addition is needed only at relative humidity values of below 70%. Any one of the three oils provided sufficient adhesion, although canola oil is preferred because of its low volatility and because it is a vegetable oil .

Table 16. Spray application details, droplet size spectra and deposit assessment of active ingredient

Measurements	Formulation abbreviation				
	Sumi-66.7	FDA-3409-1.5	FDA-3409-4.0	FT-114-5.0	FT-114-7.0
Floor area of the chamber (m^2)	3.87	3.87	3.87	3.87	3.87
Spray nozzle	Spin. disc.*	Spin. disc.	Spin. disc.	Spin. disc.	Spin. disc.
Temperature °C	22 ± 2	22 ± 2	22 ± 2	22 ± 2	22 ± 2
Wind conditions	Still air@	Still air	Still air	Still air	Still air
Relative humidity (%)	50 ± 3	50 ± 3	50 ± 3	50 ± 3	50 ± 3
Application rate (g AI/ha)	210	210	210	210	210
Volume rate (l/ha)	1.5	1.5	1.5	1.5	1.5
Droplets/cm^2	22	36	87	65	75
NMD (μm)	72	28	31	38	36
VMD (μm)	79	55	42	49	47
D_{max}	130	100	100	90	100
Volume deposit (l/ha)**	601	254	425	375	475
Percent deposition**	43.4	16.9	28.3	25.0	31.7

* Flak™ from Micron Corporation, Wingham, Ont., Canada.

@ Still air except for the minor turbulence generated by the moving nozzle along the central rail of the spray chamber.

** Volume deposit was calculated from the AI concentration determined by GLC. From this the percent deposition was estimated.

RESULTS AND DISCUSSION

1. Physical Properties

1.1. Viscosity

Viscosity is the most important intrinsic property that provides information on liquid behaviour under various handling conditions. Formulation stability, pumping and mixing capabilities and spray atomization characteristics are all related to this intrinsic property. Data in Tables 3 to 6 show that the New Sumithion 20% flowable is an excellent formulation that has comparable storage stability of the spray mixtures, winter storage capabilities of the formulation concentrate, pumping and mixing characteristics and re-emulsifiability of the stored mixtures.

1.2. Evaporation Rates

Tables 11, 12 and 13, and Figure 1 provide data on comparative evaporation pattern of the six formulations. The data indicate that the spray mixture Sumi-66.7 prepared from Sumithion 20F has a much better evaporation pattern since it has a lower evaporation rate and a higher residual weight percent. This finding implies that Sumi-66.7 would provide greater spray deposits at the target region than any of the four formulations studied.

2. Spray Droplet Spectra on Kromekote Cards

Table 16 and Figures 2, 4, 6, 8 and 10 provide data on comparative droplet spectra, NMD, VMD and D_{max} for all the five spray mixtures. The data indicate that a significantly higher NMD, VMD and D_{max} values were obtained for the spray mix Sumi-66.7. The reason for this lies in the thixotropic properties of the formulation. Because of this, a significantly lower proportion of the 'driftable' small droplets are produced during atomization of this formution. This provides a great advantage for aerial spraying near the watersheds and other sensitive areas requiring large buffer zones. Use of this formulation would probably reduce the buffer zone, thus providing forest protection to greater areas of forests.

3. Deposit Analysis on Glass Plates

GLC analysis of the deposited fenitrothion (Table 16) provide information on the percentage deposition of the applied formulations. Data in Table 16 indicate a significantly higher deposit value for Sumi-66.7 than for the currently used FDA-3409-1.5, and slightly higher deposit values than for FDA-3409-4.0 and FT-114-7.0 formulations. The reason for this high deposition rate lies in the properties of the formulations, viz., a much better evaporation pattern, thixotropic properties, better atomization with much less proportion of small droplets and a higher surface tension.

CONCLUSIONS

The present study with the Sumithion 20% flowable indicates the following:

a). The formulation concentrate is highly suitable for easy storage under the cold winter climate conditions of Canada. Because of the thixotropic properties, it has excellent flowing properties in the pumps and the pipes during mixing. Even when it is frozen, it poses no difficulty in pumping capabilities, and upon thawing it has an excellent stability with no phase separation of the active ingredient.

b). The spray formulation, which was prepared to provide a concentration of 210 g AI/1.5 litre/ha, exhibits excellent stability properties, re-emulsifiability, appropriate viscosity, better evaporation rate and atomization properties. However, there is a tendency to provide too many large droplets thus resulting in a somewhat low droplet density (droplets/cm^2). This however, can be corrected if the application equipment were calibrated to counteract this minor drawback. The droplet size spectra on Kromekote cards indicated a significantly lower proportion of 'driftable' small droplets, a phenomenon which was caused by the thixotropic properties of the formulation. The spray deposition pattern also indicated that the formulation provided AI deposits similar to the four registered formulations tested.

The droplet adhesion characteristics indicated a somewhat poor adhesion under very dry ambient air conditions, especially if the needle surface was dry before spraying. To improve adhesion on spruce needles under low relative humidity conditions, it is recommended that about 3 to 4 percent v/v of canola oil be added to the spray mixture, at 70% and below humidity values.

ACKNOWLEDGEMENTS

The author wishes to thank Mr. John W. Leung for his technical assistance in carrying out this investigation.

REFERENCES

Sundaram, K.M.S. and Nott, R. Fenitrothion Residues in Selected Components of A Conifer Forest Following Aerial Application of Tank Mixes Containing Triton® X-100. Agric. Can. For. Serv. For. Pest Mangmt. Inst. Inf. Rept. FPM-X-65, 1984, 14 pp.

A P P E N D I X

Droplet Number and Volume Distribution According to Size Category

TABLE 17

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2% . Formulation Sumi-66.7

Droplet number distribution according to size category

Stain diameter range (μm)	Spread factor	Droplet diameter range (μm)	Average droplet diameter (μm)	Total droplets per 48 cm^2	Droplets per cm^2	Frequency (%)	Cumulative frequency (%)
≤ 20	1.80	≤ 10	5	1	0.02	0.10	0.10
21 - 37	1.85	11 - 20	15	3	0.06	0.29	0.38
38 - 56	1.85	21 - 30	25	5	0.11	0.48	0.86
57 - 76	1.90	31 - 40	35	22	0.46	2.10	2.96
77 - 95	1.90	41 - 50	45	37	0.78	3.54	6.50
96 - 114	1.90	51 - 60	55	71	1.49	6.79	13.29
115 - 137	1.95	61 - 70	65	119	2.50	11.38	24.67
138 - 156	1.95	71 - 80	75	393	8.27	37.57	62.24
157 - 176	1.96	81 - 90	85	234	4.92	22.37	84.61
177 - 200	2.00	91 - 100	95	102	2.15	9.75	94.36
201 - 220	2.00	101 - 110	105	32	0.67	3.06	97.42
221 - 240	2.00	111 - 120	115	14	0.29	1.34	98.76
241 - 265	2.05	121 - 130	125	13	0.27	1.24	100.00
				Total = 1046	22.0		

TABLE 18

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2%. Formulation Sumi-66.7

Droplet volume distribution according to size category

Droplet diameter range (μm)	Average droplet diameter (μm)	Volume of one droplet (10^{-8} cc)	Droplets per cm^2	Volume of deposit (mL/ha)	Droplet volume distribution (%)	Cumulative droplet volume distribution (%)
≤ 10	5	0.01	0.02	0.0001	0.00	0.00
11 - 20	15	0.18	0.06	0.0112	0.00	0.00
21 - 30	25	0.82	0.11	0.0860	0.01	0.01
31 - 40	35	2.24	0.46	1.039	0.18	0.19
41 - 50	45	4.77	0.78	3.713	0.64	0.83
51 - 60	55	8.71	1.49	13.01	2.23	3.06
61 - 70	65	14.38	2.50	35.99	6.17	9.23
71 - 80	75	22.09	8.27	182.6	31.31	40.54
81 - 90	85	32.16	4.92	158.3	27.14	67.68
91 - 100	95	44.89	2.15	96.31	16.51	84.19
101 - 110	105	60.61	0.67	40.80	7.00	91.18
111 - 120	115	79.63	0.29	23.45	4.02	95.21
121 - 130	125	102.27	0.27	27.96	4.79	100.00
		Total =	22.0	583.2		

TABLE 19

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2% . Formulation FDA-3409-1.5

Droplet number distribution according to size category

Stain diameter range (μm)	Spread factor	Droplet diameter range (μm)	Average droplet diameter (μm)	Total droplets per 48 cm²	Droplets per cm²	Frequency (%)	Cumulative frequency (%)
≤ 30	3.00	≤ 10	5	3	0.06	0.17	0.17
31 - 60	3.03	11 - 20	15	238	4.99	13.87	14.04
61 - 90	3.04	21 - 30	25	552	11.58	32.17	46.21
91 - 120	3.04	31 - 40	35	390	8.18	22.73	68.94
121 - 150	3.05	41 - 50	45	231	4.85	13.46	82.40
151 - 180	3.05	51 - 60	55	144	3.02	8.39	90.79
181 - 210	3.05	61 - 70	65	75	1.57	4.37	95.16
211 - 240	3.05	71 - 80	75	41	0.86	2.39	97.55
241 - 270	3.05	81 - 90	85	18	0.38	1.05	98.60
271 - 300	3.05	91 - 100	95	24	0.50	1.40	100.00
301 - 335	3.06	101 - 110	105	--	--	--	--
336 - 370	3.07	111 - 120	115	--	--	--	--
371 - 400	3.08	121 - 130	125	--	--	--	--
			Total =	1716	36.00		

TABLE 20

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2%. Formulation FDA-3409-1.5

Droplet volume distribution according to size category

Droplet diameter range (μm)	Average droplet diameter (μm)	Volume of one droplet (10^{-8} cc)	Droplets per cm^2	Volume of deposit (mL/ha)	Droplet volume distribution (%)	Cumulative droplet volume distribution (%)
≤ 10	5	0.01	0.06	0.0004	0.00	0.00
11 - 20	15	0.18	4.99	0.8823	0.57	0.57
21 - 30	25	0.82	11.58	9.474	6.13	6.70
31 - 40	35	2.24	8.18	18.37	11.89	18.59
41 - 50	45	4.77	4.85	23.12	14.96	33.55
51 - 60	55	8.71	3.02	26.32	17.03	50.58
61 - 70	65	14.38	1.57	22.62	14.64	65.22
71 - 80	75	22.09	0.86	19.00	12.29	77.52
81 - 90	85	32.16	0.38	12.14	7.86	85.37
91 - 100	95	44.89	0.50	22.60	14.63	100.00
101 - 110	105	60.61	--	--	--	--
111 - 120	115	79.63	--	--	--	--
121 - 130	125	102.27	--	--	--	--
		Total =	36.00	154.5		

TABLE 21

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2% . Formulation FDA-3409-4.0

Droplet number distribution according to size category

Stain diameter range (μm)	Spread factor	Droplet diameter range (μm)	Average droplet diameter (μm)	Total droplets per 8 cm^2	Droplets per cm^2	Frequency (%)	Cumulative frequency (%)
⩽ 22	2.15	⩽ 10	5	0	0.00	0.00	0.00
23 - 60	3.00	11 - 20	15	32	4.00	4.60	4.60
61 - 100	3.33	21 - 30	25	152	19.00	21.84	26.44
101 - 140	3.50	31 - 40	35	273	34.13	39.22	65.66
141 - 180	3.60	41 - 50	45	150	18.75	21.55	87.21
181 - 220	3.62	51 - 60	55	54	6.75	7.76	94.97
221 - 255	3.65	61 - 70	65	30	3.75	4.31	99.28
256 - 300	3.70	71 - 80	75	2	0.25	0.29	99.57
301 - 335	3.72	81 - 90	85	2	0.25	0.29	99.86
336 - 375	3.75	91 - 100	95	1	0.13	0.14	100.00
376 - 415	3.78	101 - 110	105	-	--	--	--
416 - 455	3.80	111 - 120	115	-	--	--	--
456 - 495	3.82	121 - 130	125	-	--	--	--
			Total =	696	87.00		

TABLE 22

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2%. Formulation FDA-3409-4.0

Droplet volume distribution according to size category

Droplet diameter range (μm)	Average droplet diameter (μm)	Volume of one droplet (10^{-8} cc)	Droplets per cm^2	Volume of deposit (mL/ha)	Droplet volume distribution (%)	Cumulative droplet volume distribution (%)
≤ 10	5	0.01	0.00	0.0000	0.00	0.00
11 - 20	15	0.18	4.00	0.7069	0.22	0.22
21 - 30	25	0.82	19.00	15.54	4.95	5.17
31 - 40	35	2.24	34.13	76.61	24.38	29.55
41 - 50	45	4.77	18.75	89.46	28.47	58.02
51 - 60	55	8.71	6.75	58.80	18.71	76.74
61 - 70	65	14.38	3.75	53.92	17.16	93.90
71 - 80	75	22.09	0.25	5.522	1.76	95.66
81 - 90	85	32.16	0.25	8.039	2.56	98.21
91 - 100	95	44.89	0.13	5.612	1.79	100.00
		Total =	87.00	314.2		

TABLE 23

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2% . Formulation FT-114-5.0

Droplet number distribution according to size category

Stain diameter range (μm)	Spread factor	Droplet diameter range (μm)	Average droplet diameter (μm)	Total droplets per 8 cm²	Droplets per cm²	Frequency (%)	Cumulative frequency (%)
≤ 25	2.45	≤ 10	5	4	0.50	0.77	0.77
26 - 50	2.55	11 - 20	15	18	2.25	3.45	4.22
51 - 80	2.65	21 - 30	25	86	10.73	16.51	20.73
81 - 110	2.75	31 - 40	35	86	10.73	16.51	37.24
111 - 145	2.90	41 - 50	45	190	23.70	36.47	73.70
146 - 180	3.00	51 - 60	55	82	10.23	15.74	89.44
181 - 215	3.07	61 - 70	65	36	4.49	6.91	96.35
216 - 245	3.10	71 - 80	75	17	2.12	3.26	99.62
246 - 280	3.11	81 - 90	85	2	0.25	0.38	100.00
281 - 320	3.20	91 - 100	95	--	--	--	--
			Total =	521.00	65.00		

TABLE 24

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2%. Formulation FT-114-5.0

Droplet volume distribution according to size category

Droplet diameter range (μm)	Average droplet diameter (μm)	Volume of one droplet (10^{-8} cc)	Droplets per cm^2	Volume of deposit (mL/ha)	Droplet volume distribution (%)	Cumulative droplet volume distribution (%)
≤ 10	5	0.01	0.50	0.0033	0.00	0.00
11 - 20	15	0.18	2.25	0.3968	0.11	0.11
21 - 30	25	0.82	10.73	8.778	2.47	2.59
31 - 40	35	2.24	10.73	24.09	6.79	9.37
41 - 50	45	4.77	23.70	113.1	31.86	41.24
51 - 60	55	8.71	10.23	89.12	25.11	66.34
61 - 70	65	14.38	4.49	64.58	18.20	84.54
71 - 80	75	22.09	2.12	46.85	13.20	97.74
81 - 90	85	32.16	0.25	8.024	2.26	100.00
91 - 100	95	44.89	--	--	--	--
		Total =	65.00	354.9		

TABLE 25

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2% .. Formulation FT-114-7.0

Droplet number distribution according to size category

Stain diameter range (μm)	Spread factor	Droplet diameter range (μm)	Average droplet diameter (μm)	Total droplets per 8 cm^2	Droplets per cm^2	Frequency (%)	Cumulative frequency (%)
≤ 25	2.45	≤ 10	5	4	0.50	0.67	0.67
26 - 50	2.55	11 - 20	15	40	5.03	6.71	7.38
51 - 80	2.60	21 - 30	25	66	8.31	11.07	18.46
81 - 105	2.65	31 - 40	35	150	18.88	25.17	43.62
106 - 130	2.75	41 - 50	45	186	23.41	31.21	74.83
131 - 170	2.83	51 - 60	55	110	13.84	18.46	93.29
171 - 210	3.00	61 - 70	65	30	3.78	5.03	98.32
211 - 250	3.12	71 - 80	75	5	0.63	0.84	99.16
251 - 290	3.22	81 - 90	85	4	0.50	0.67	99.83
291 - 330	3.30	91 - 100	95	1	0.13	0.17	100.00
			Total =	596	75.00		

TABLE 26

Kromekote card data. Spray chamber study. Temp. °C = 22 ± 1.5

Relative humidity = 45 ± 2%. Formulation FT-114-7.0

Droplet volume distribution according to size category

Droplet diameter range (μm)	Average droplet diameter (μm)	Volume of one droplet (10^{-8} cc)	Droplets per cm^2	Volume of deposit (mL/ha)	Droplet volume distribution (%)	Cumulative droplet volume distribution (%)
≤ 10	5	0.01	0.50	0.0033	0.00	0.00
11 - 20	15	0.18	5.03	0.8895	0.24	0.24
21 - 30	25	0.82	8.31	6.795	1.82	2.06
31 - 40	35	2.24	18.88	42.375	11.38	13.45
41 - 50	45	4.77	23.41	111.7	29.99	43.44
51 - 60	55	8.71	13.84	120.6	32.39	75.82
61 - 70	65	14.38	3.78	54.28	14.58	90.40
71 - 80	75	22.09	0.63	13.90	3.73	94.14
81 - 90	85	32.16	0.50	16.19	4.35	98.48
91 - 100	95	44.89	0.13	5.650	1.52	100.00
		Total =	75.00	372.3		