



# Department of Pesticide Regulation



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## MEMORANDUM

TO: Randy Segawa, Senior Environmental Research Scientist (Supervisor)  
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SUBJECT: CALCULATION OF A TOLERANCE INTERVAL FOR A TOWNSHIP LIMIT  
ON METHYL BROMIDE USE TO CONTROL SUBCHRONIC EXPOSURE

### Background

Methodology for determining a methyl bromide use cap by employing tolerance interval statistical techniques was referenced in Frank (2003). The purpose in such a calculation is to establish a use limit, which assures that when methyl bromide use is less than such a limit, then one-month, or two-month average ambient air concentrations will also be below a corresponding health limit of nine parts per billion (ppb). Further examples and discussion of tolerance intervals can be found in Hahn and Meeker (1991).

Li et al. (2001) provided a methodology for systematically exploring the empirical relationship between methyl bromide use density and methyl bromide subchronic air concentrations. Li et al. (2001) was based on monitoring conducted in the summer and fall of 2000 in Monterey, Santa Cruz and Kern Counties (Mongar and Lew 2000, Mongar and Lew 2001). Li et al. (2001) organized the analysis by systematic examination of regressions over stepwise changes in both spatial and temporal scales in order to find a 'best' relationship between measured air concentrations and methyl bromide use surrounding each monitoring site. Li et al. (2001) examined many spatial and temporal scales. The general criterion for evaluating the relationships was based on the  $r^2$  value. Higher  $r^2$  values were viewed as better within this context.

In line with Frank (2003), the focus in this analysis will be at the township scale. The township consists of a 6 x 6 square mile area. The basic regressions developed in this memorandum utilize weekly average use per township. The original use estimates bracketed the 6 x 6 township areas with 5 x 5 and 7 x 7 square miles. In order to estimate the 6 x 6 square mile use, linear interpolation was utilized as follows:

$$U(6x6) = \left[ \frac{(36 - 25)}{(49 - 25)} * (U(7x7) - U(5x5)) \right] + U(5x5) \quad (0.1)$$

where U (i x i) stands for weekly average use over an i x i square mile area.



Subsequent to Li et al. (2001), further monitoring results have become available. These results consist of Air Resources Board (ARB) monitoring in 2001 in Kern and Monterey (Cook et al. 2002, Mongar et al. 2002), Alliance for Methyl Bromide Industry (AMBI) monitoring in Santa Maria and Ventura in 2001 (Winegar 2002), and AMBI monitoring in Monterey and Ventura in 2002 (Winegar 2003).

The purpose of this memorandum is to discuss the studies which will be used to examine the relationship between air concentration and use, to determine that relationship, and to utilize the tolerance interval statistical procedure to calculate a use limit which will assure air concentrations remain substantively below nine ppb, a level promulgated in Frank (2003).

*Note on use information.* The regressions have been conducted on methyl bromide use calculated as weekly average pounds of methyl bromide use per township. In the case of the one-month analysis, the weekly average use was based on an approximately four-week period, during which the monitoring occurred. For the two-month analysis, the weekly average use was based on the approximately eight-week period of monitoring. The one-month analysis used 62 pairs of concentration versus weekly average use values, while the two-month analysis used 31 pairs of concentration versus weekly average use values. For each monitoring station, the two-month average weekly use is approximately the average of the two four-week periods. However, these relationships are not exact because some 'months' were comprised of only three weeks of monitoring. To convert from a weekly average use to a monthly use, we have assumed 30 day months and used the fraction  $(30/7)=4.286$  to convert between use on weekly and monthly basis. That is, 10,000 pounds of weekly average use is equivalent to 42,860 pounds of monthly use. Linear transformation of these use amounts does not affect any underlying linear regression relationship between concentration and use.

### **Discussion of studies**

Four studies of ambient air monitoring for methyl bromide have been conducted in recent years: ARB 2000 (Mongar and Lew 2000, Mongar and Lew 2001), ARB 2001 (Cook et al. 2002, Mongar et al. 2002), AMBI 2001 (Winegar 2002), and AMBI 2002 (Winegar 2003a). Based on the Department of Pesticide Regulation's (DPR's) review of the studies, three of these studies were used for the regression analysis. The AMBI 2001 data are still questionable, and have not been used in this analysis. AMBI's response (Winegar 2003b) to DPR's review (Segawa 2002) still leaves some issues unresolved and raises new issues. The identification of valid and invalid samples due to airflow deviations remains unresolved. The AMBI 2001 study uses airflow deviation of  $\pm 50\%$  as the criteria for invalid samples, while the other three studies use  $\pm 25\%$ . DPR believes that all studies should use the same criteria. The quality control results also indicate questionable data. All trip blanks were positive. AMBI's June 3, 2003 response explains that "the likely cause for the problem was the site for sample media receipt, which was near a high methyl bromide use area. Small vacuum leaks could have contributed to the

inadvertent collection of a small amount of contaminated ambient air.” Since all of the trip blanks were positive, this indicates a systematic error in the sampling, with a variable effect on field samples. If this is the cause of contamination, it indicates that field samples containing high concentrations were inadvertently diluted due to the leaks. The AMBI 2001 (Winegar 2002) study should not be used unless these and other issues affecting the data can be resolved.

### **Analysis of relationship between use and air concentration**

As described in Frank (2003), two regressions analyses were conducted: one based on month-long average air concentrations and one based on the entire period (mostly two-month) average air concentrations. Most periods were eight weeks, however, a few were seven weeks. The data sets utilized for regression are shown in Tables 1 and 2. Although both regressions were statistically significant, the fit was poor with  $r^2$  values of 28% and 32%, respectively, for one and two-month regressions (Table 3).

For the one-month average concentrations, the basic regression line and points are shown in Figure 1A. The residuals tend to show more uniform scatter around the axis for larger use values, than smaller use values (Figure 1B). The regression of standardized residuals on rankits (Figure 1C) was highly significant ( $p < .001$ ) and gave an intercept of 0 and a slope of 0.91, which was not significantly different from 1.0 ( $p > 0.05$ ). Sokal and Rohlf (1981) suggest that this line should be linear if the residuals are normally distributed (pg 122). Glantz and Slinker (1990, page 130) suggest performing a significance test on the correlation coefficient to determine if the residuals are normally distributed. This is equivalent to a significant linear regression. These rankit regression results strongly imply normality of residuals. Therefore, these residuals are sufficiently normal for the regression analysis.

Similar results were obtained for the two-month analysis (Figures 2ABC). The fit was slightly better with an  $r^2$  of 32%, but still poor. For the standardized residual analysis (Figure 2C), the intercept was zero and slope was not significantly different from one ( $p > 0.05$ ), with a highly significant relationship ( $p < .001$ ). As in the one-month analysis, these residuals appear to be sufficiently normal for the basic regression.

### **Determination of tolerance limit**

Under typical regression assumptions,  $x$  is fixed, and the variance in any particular estimated value at  $x$  ( $y(x) = bx + a$ ) depends on various regression statistics as well as the value of  $x$  where the estimate is made. For a given value of  $x$  (in this case, methyl bromide use in pounds per month), one would expect (under the typical regression assumptions) that an observed concentration at that level of use would be greater than the predicted concentration about half of the time, because the predicted concentration represents the mean of a normal distribution centered vertically over  $y(x)$ .

Given a hypothetical  $x$  level of use, what is an interval based on the normal distribution of individuals around  $y(x)$ , which would capture with some level of confidence the 95<sup>th</sup> percentile concentration value? The goal is to find some value (in this case, ambient air concentration of methyl bromide), call it  $T$ , which is the bound for a one-sided interval such that 90% of the time this interval would contain (i.e. be greater than) the true 95<sup>th</sup> percentile of the distribution of points at  $c(x)$ . This is called a tolerance interval.

Vardeman (1994) gives a procedure for determining tolerance limits for regressions. The relevant case in his formulation is

$$(-\infty, a + bX + \tau s_{xy}) \quad (0.2)$$

Expression 1.2 defines a one sided (upper) tolerance limit for a fraction  $p$  of responses, which correspond to a particular value of the  $x$  variable. This range goes from negative infinity (which in the practical case at hand is zero) to some value above the regression line at the point  $x$ . In expression 1.2,  $a$  is the  $y$  intercept and  $b$  is the slope of the regression. The value  $\tau$  is calculated according to Vardeman (1994) as

$$\tau = \frac{Q_z(p) + A\lambda \sqrt{1 + \frac{1}{1(n-2)} \left( \frac{Q_z^2(p)}{A^2} - \lambda^2 \right)}}{1 - \frac{\lambda^2}{2(n-2)}} \quad (0.3)$$

In equation 1.3,  $Q_z$  is the inverse cumulative normal distribution,  $A$  is defined below and  $\lambda$  is a value which can be approximated by  $Q_z(\gamma)$ , where  $\gamma$  is the desired level of confidence for the tolerance interval. In the case where  $\gamma=0.95$ , Vardeman (1994) presents a more exacting method for calculating  $\tau$  which involves calculating an intermediate value and then performing an interpolation to estimate  $\lambda$  from tabulated values in an Appendix (Vardeman 1994, Table D-8, page A79). A sample calculation using both the interpolation and estimation methods for calculating  $\lambda$  yielded 2.43 and 2.42, respectively, for  $\tau$ . Therefore, the approximation appears sufficiently accurate. The definition for  $A$  in equation 1.3 is shown below in equation 1.4.

$$A = \sqrt{\frac{1}{n} + \frac{x_0^2}{\sum_i x_i^2}} \quad (0.4)$$

where  $x_0$  is a particular level of methyl bromide use normalized by subtracting the average of the x values used in the regression ( $x_0 = X_0 - \bar{X}$ ), n is the number of points in the regression, and  $x_i$  are the individual x values used in the regression, each with the mean x value subtracted from it ( $x_i = X_i - \bar{X}$ ).

Putting together equation 1.2 and the goal of determining the upper tolerance limit, results in equation 1.5 below:

$$T_{upper} = a + bX_0 + \tau s_{yx} \quad (0.5)$$

where  $T_{upper}$  is the upper tolerance limit on the 95<sup>th</sup> percentile at  $X_0$ , a is the y intercept ( $a = \bar{y} - b\bar{x}$ ), b is the slope, and the capitalized letter,  $X_0$ , represents use without subtracting the mean use.

As described in Frank (2003), the reference level is nine ppb. This means that the goal of the calculations in this memorandum is to find the X-value such that the 90% confidence 95<sup>th</sup> percentile estimate is nine ppb. This is working backwards given a reference level of nine ppb to find a corresponding use level such that there will be 90% confidence that 95<sup>th</sup> upper percentile estimated at this use level, will not exceed nine ppb. Another way to say this is that ninety times out of 100 in long term theoretical sampling this interval can be expected to capture the true population 95<sup>th</sup> percentile air concentration at that X-value. Substituting the assumed limit of nine ppb and the slope, intercept and regression error (Table 3), for illustration, from the case of the one-month regression analysis into equation 1.4 above, gives the following equation

$$9 = 0.868 + 0.0000624X_0 + \tau(1.634) \quad (0.6)$$

The problem becomes one of inverse estimation of  $X_0$ . There is an implicit contribution of  $X_0$  to  $\tau$  via equations 1.3 and 1.4. Consequently, solving for the appropriate value of  $X_0$  is not trivial.

For each of the two cases, a spreadsheet analysis using Excel (2000) was created. The spreadsheet contained the formulas listed above, the basic regression statistics, and used the 'Solver' feature to determine a level of use ( $X_0$ ) that resulted in a nine ppb tolerance limit.

Figure 3 depicts the regression line and the upper 90% tolerance bound on the 95th percentile at each x value for the one-month analysis. Based on the one-month analysis, the spreadsheet solver routine locates the weekly average use at 70,560 lbs/week-township, equivalent

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to 302,420 lbs/month-township. This solution lies outside the range of measured use values and should therefore be regarded with some caution. The two axes for use in Figure 3 reflect weekly average use (as found in Table 1) and the equivalent month use.

A single point lies above the nine ppb level (Figure 3). This point was generated during ARB monitoring in Monterey during 2000. The weekly average use corresponding to this point was 25786 (Table 1), which is equivalent to a monthly use of 110,511 lbs per township. The existence of this measured value underscores the nature of this analysis and associated policy, which in theory and in reality allows for the possibility of exceeding the reference concentration. The majority of points, however, do fall below the nine ppb level.

Figure 4 depicts the solution for the two-month case. The general features are similar to those in one-month case (Figure 3). The solution, however, is smaller at 62,108 lbs/week-township, or the equivalent of 266,194 lbs/month-township. As in the one-month case, the solution lies outside the range of measured use. None of the two month concentrations exceed nine ppb, though two values exceed the upper 90% tolerance bound on the 95th percentile. Both of these points were measurements taken in Monterey by the ARB (Table 2).

In accordance with Frank (2003), the more conservative of the two equations is recommended for use in establishing a township cap. Hence, the recommended township cap is 266,194 lbs/month. This number may be rounded down in order to simplify and consistent with the fact that the smaller place holding digits are not significant.

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Sally Powell, Senior Environmental Research Scientist

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Table 1. One-month data used for regression analysis. Linear interpolation used to derive 6x6 square mile estimate. Site codes available in various monitoring study reports. The designators 1 and 2 indicate months 1 or 2. Zones are mon=Monterey/Santa Cruz, ker=Kern, ven=Ventura.

Site Code	Zone	Year	Monitor	Weekly Average Use (lbs/week)			Concentration (ppb)
				5x5	7x7	6x6 square	
				square miles	square miles	miles (Interpolated)	
SAL1	mon	2000	ARB	6404	20925	13059	1.32
SAL2	mon	2000	ARB	2300	8772	5266	1.26
OAS1	mon	2000	ARB	0	0	0	0.35
OAS2	mon	2000	ARB	1183	4613	2755	0.43
CHU1	mon	2000	ARB	2404	6346	4211	0.69
CHU2	mon	2000	ARB	0	4375	2005	0.64
LJE1	mon	2000	ARB	13468	32310	22104	5.43
LJE2	mon	2000	ARB	3713	14950	8863	2.32
PMS1	mon	2000	ARB	34689	67713	49825	6.12
PMS2	mon	2000	ARB	17963	35031	25786	9.35
SES1	mon	2000	ARB	16842	30895	23283	3.55
SES2	mon	2000	ARB	11380	17727	14289	1.68
ARB1	ker	2000	ARB	0	0	0	0.26
ARB2	ker	2000	ARB	0	0	0	0.10
SHA1	ker	2000	ARB	0	0	0	0.91
SHA2	ker	2000	ARB	0	2229	1022	0.64
CRS1	ker	2000	ARB	10474	10474	10474	2.88
CRS2	ker	2000	ARB	8079	8079	8079	1.19
MVS1	ker	2000	ARB	0	0	0	0.08
MVS2	ker	2000	ARB	0	0	0	0.11
VSD1	ker	2000	ARB	0	0	0	0.10
VSD2	ker	2000	ARB	0	0	0	0.10
SAL1	mon	2001	ARB	11879	16509	14001	0.90
SAL2	mon	2001	ARB	3733	5234	4421	1.92
MES1	mon	2001	ARB	25908	47166	35651	6.72
MES2	mon	2001	ARB	6455	10578	8345	5.56
CHU1	mon	2001	ARB	1122	3556	2238	0.52
CHU2	mon	2001	ARB	1512	3393	2374	0.65
LJE1	mon	2001	ARB	10362	18424	14057	1.41
LJE2	mon	2001	ARB	4675	9217	6757	4.30
PMS1	mon	2001	ARB	31192	64122	46285	3.26
PMS2	mon	2001	ARB	3685	12340	7652	3.38
SES1	mon	2001	ARB	19305	30017	24215	1.09
SES2	mon	2001	ARB	5245	8841	6893	1.19
ARB1	ker	2001	ARB	0	0	0	0.09
ARB2	ker	2001	ARB	0	0	0	0.15
ARV1	ker	2001	ARB	0	0	0	0.06
ARV2	ker	2001	ARB	0	0	0	0.09
CRS1	ker	2001	ARB	6258	6258	6258	2.83
CRS2	ker	2001	ARB	2300	2300	2300	2.71
MVS1	ker	2001	ARB	0	0	0	0.05
MVS2	ker	2001	ARB	0	0	0	0.10
VSD1	ker	2001	ARB	0	0	0	0.05
VSD2	ker	2001	ARB	0	0	0	0.10
MAQ1	mon	2002	AMBI	11526	22644	16622	1.06
MAQ2	mon	2002	AMBI	2506	7776	4921	1.19
BBC1	mon	2002	AMBI	20636	31641	25680	2.34
BBC2	mon	2002	AMBI	12006	23219	17145	1.82
WAT1	mon	2002	AMBI	28586	66368	45903	4.94
WAT2	mon	2002	AMBI	22635	47889	34210	2.63
FRM1	mon	2002	AMBI	22553	61159	40247	3.69
FRM2	mon	2002	AMBI	20762	40395	29760	1.54
CPW1	mon	2002	AMBI	13180	35750	23525	2.69
CPW2	mon	2002	AMBI	9920	24207	16468	1.43
ABD1	ven	2002	AMBI	26405	38460	31930	0.51
ABD2	ven	2002	AMBI	36136	73318	53178	1.01
SHA1	ven	2002	AMBI	1424	7611	4260	0.17
SHA2	ven	2002	AMBI	11787	39219	24360	1.00
PVW1	ven	2002	AMBI	7531	23204	14714	0.56
PVW2	ven	2002	AMBI	35297	84843	58006	2.68
UWC1	ven	2002	AMBI	29267	42345	35261	1.45
UWC2	ven	2002	AMBI	36635	78562	55852	2.99

**Table 2. Two-month data used for regression analysis. Linear interpolation used to derive 6x6 square mile estimate. Site codes available in various monitoring reports. Zones are mon=Monterey/Santa Cruz, ker=Kern, ven=Ventura.**

Site Code	Zone	Year	Monitor	Weekly Average Use (lbs/week)			Concentration (ppb)
				5x5 square miles	7x7 square miles	6x6 square miles (interpolated)	
SAL	mon	2000	ARB	4352	14848	9163	1.29
OAS	mon	2000	ARB	591	2306	1377	0.39
CHU	mon	2000	ARB	1202	5360	3108	0.67
LJE	mon	2000	ARB	8590	23630	15483	3.88
PMS	mon	2000	ARB	26326	51372	37805	7.73
SES	mon	2000	ARB	14111	24311	18786	2.61
ARB	ker	2000	ARB	0	0	0	0.19
SHA	ker	2000	ARB	0	955	438	0.79
CRS	ker	2000	ARB	9448	9448	9448	2.16
MVS	ker	2000	ARB	0	0	0	0.09
VSD	ker	2000	ARB	0	0	0	0.10
SAL	mon	2001	ARB	7806	10747	9154	1.41
MES	mon	2001	ARB	16181	28869	21996	6.14
CHU	mon	2001	ARB	1317	3475	2306	0.58
LJE	mon	2001	ARB	7518	13819	10406	2.86
PMS	mon	2001	ARB	17435	38228	26965	3.31
SES	mon	2001	ARB	12275	19429	15554	1.14
ARB	ker	2001	ARB	0	0	0	0.12
ARV	ker	2001	ARB	0	0	0	0.08
CRS	ker	2001	ARB	4059	4059	4059	2.76
MVS	ker	2001	ARB	0	0	0	0.08
VSD	ker	2001	ARB	0	0	0	0.08
MAQ	mon	2002	AMBI	7016	15210	10771	1.12
BBC	mon	2002	AMBI	16321	27430	21412	2.08
WAT	mon	2002	AMBI	25611	57129	40056	3.78
FRM	mon	2002	AMBI	21658	50777	35004	2.62
CPW	mon	2002	AMBI	11550	29978	19996	2.06
ABD	ven	2002	AMBI	31271	55889	42554	0.76
SHA	ven	2002	AMBI	6605	23415	14310	0.59
PVW	ven	2002	AMBI	21414	54023	36360	1.62
UWC	ven	2002	AMBI	32951	60453	45556	2.22

Table 3. Regression results for one- and two-month data sets. Variables are y=concentration (ppb) and x=weekly average use of methyl bromide (lbs/week-township)

	<b>One-month</b>	<b>Two-month</b>
equation	$y=0.868+0.0000624x$	$y=0.732+0.0000721x$
$r^2$	28%	32%
p	<.001	<.001
n	62	31
syx	1.634	1.489

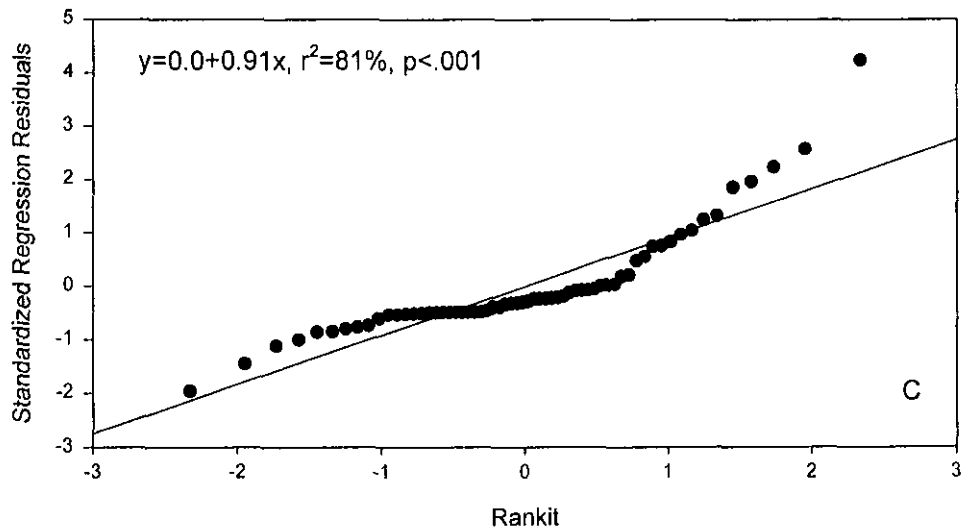
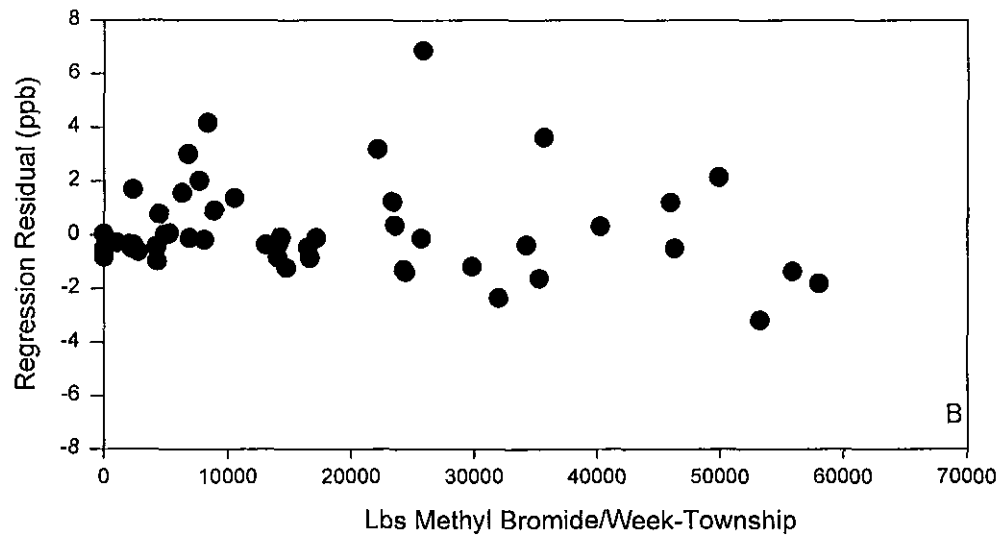
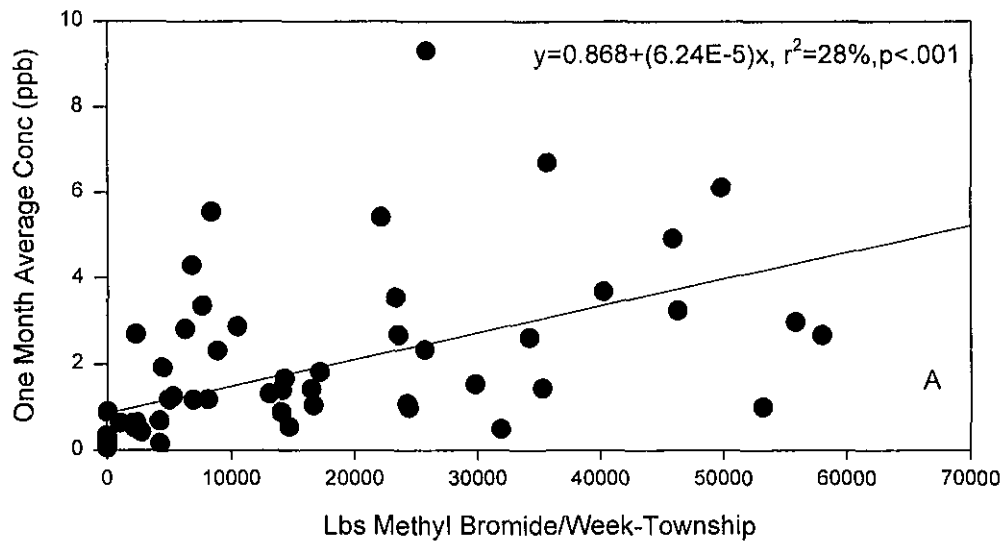


Figure 1. Regression of one-month average concentrations on average weekly use. A. Regression line and measured values. B. Residual plot. C. Regression of sorted, standardized residuals on rankit values.

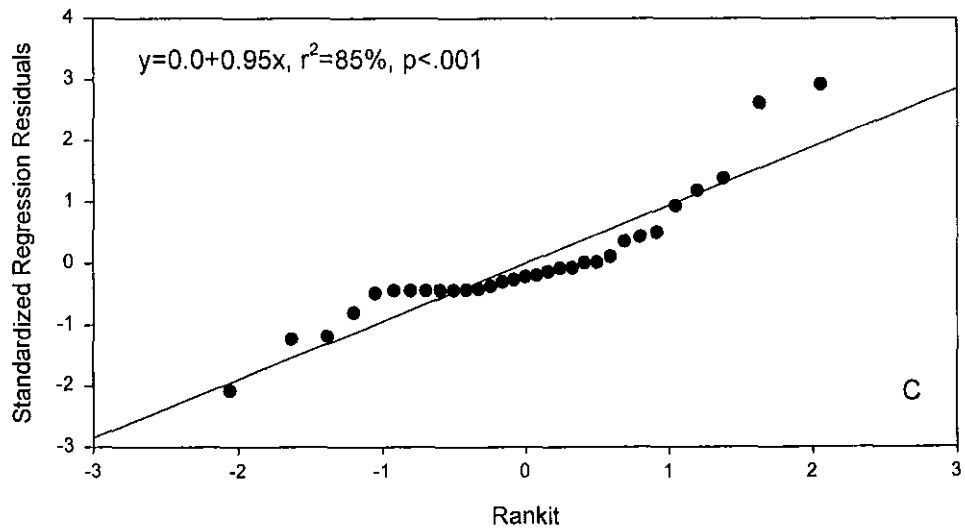
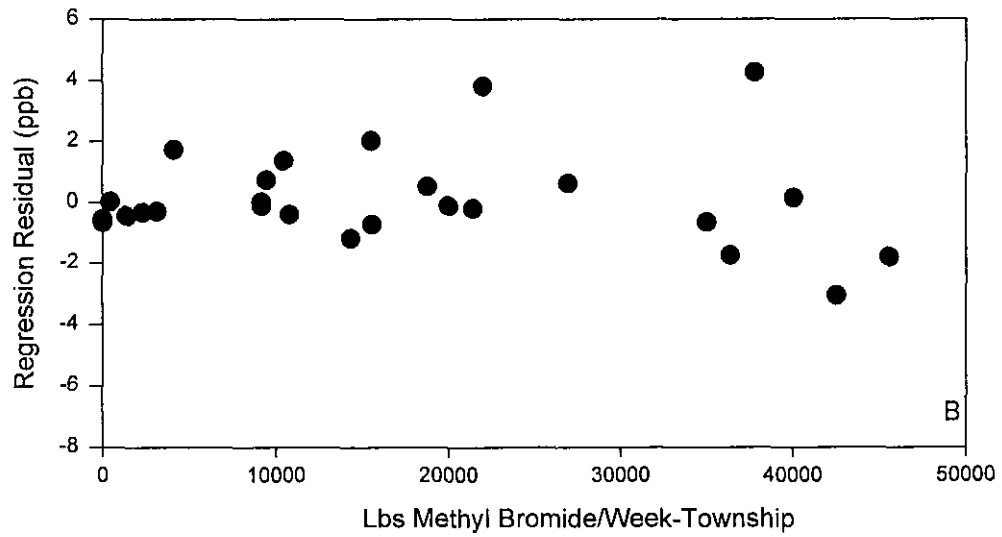
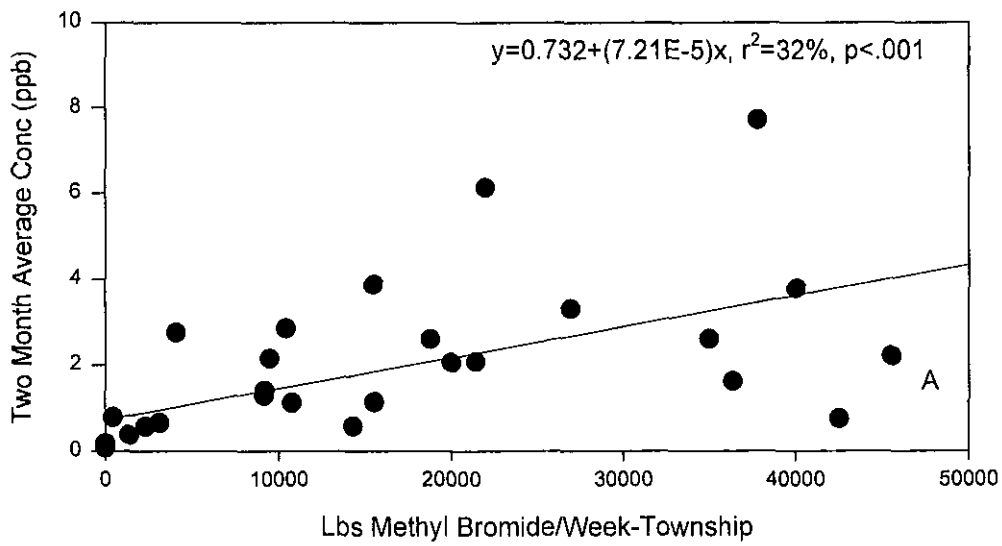


Figure 2. Regression of two-month average concentrations on average weekly use. A. Regression line and measured values. B. Residual plot. C. Regression of sorted, standardized residuals on rankit values.

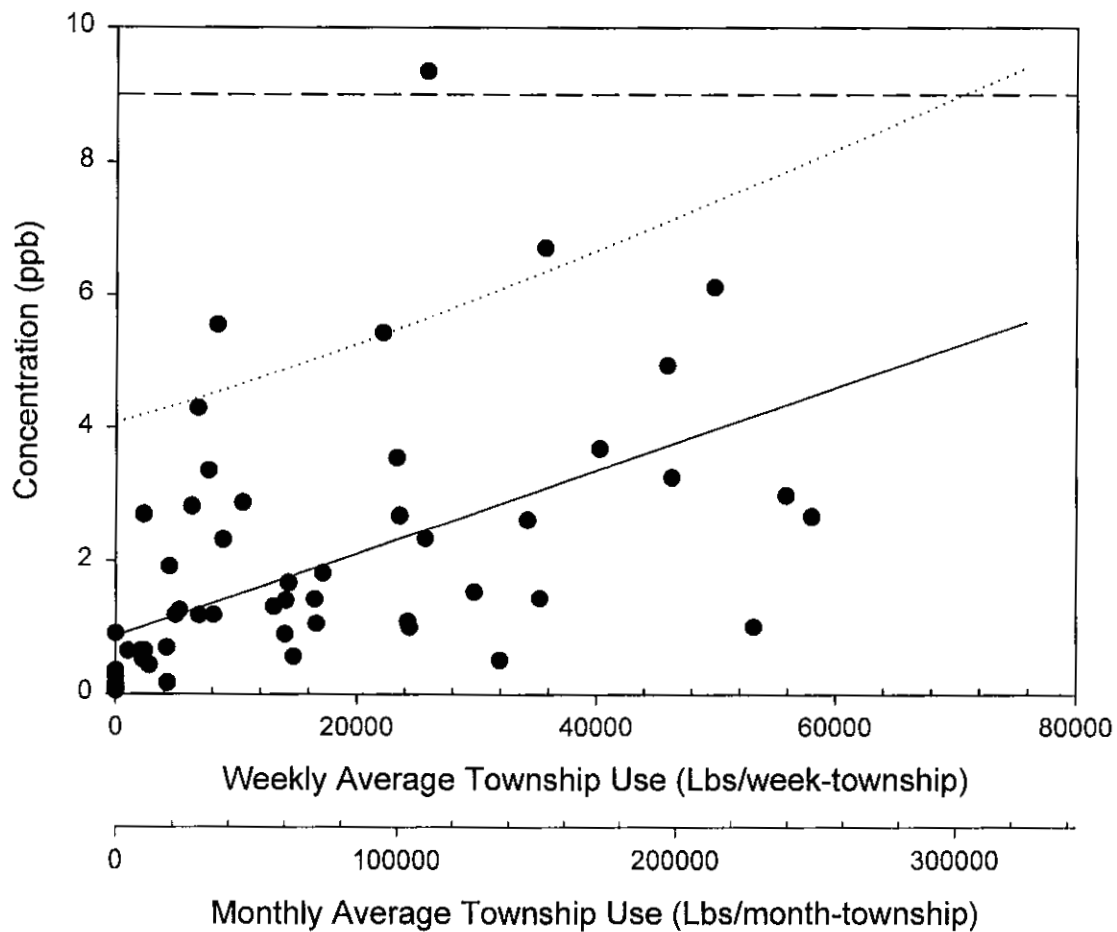


Figure 3. Regression line (solid) and 90% tolerance interval line (dotted) for the 95th percentile for the one-month case. Dashed reference line indicates 9 ppb level and use level below the intersection of tolerance line is 70,560 lbs/week-township or 302,420 lbs/month-township.

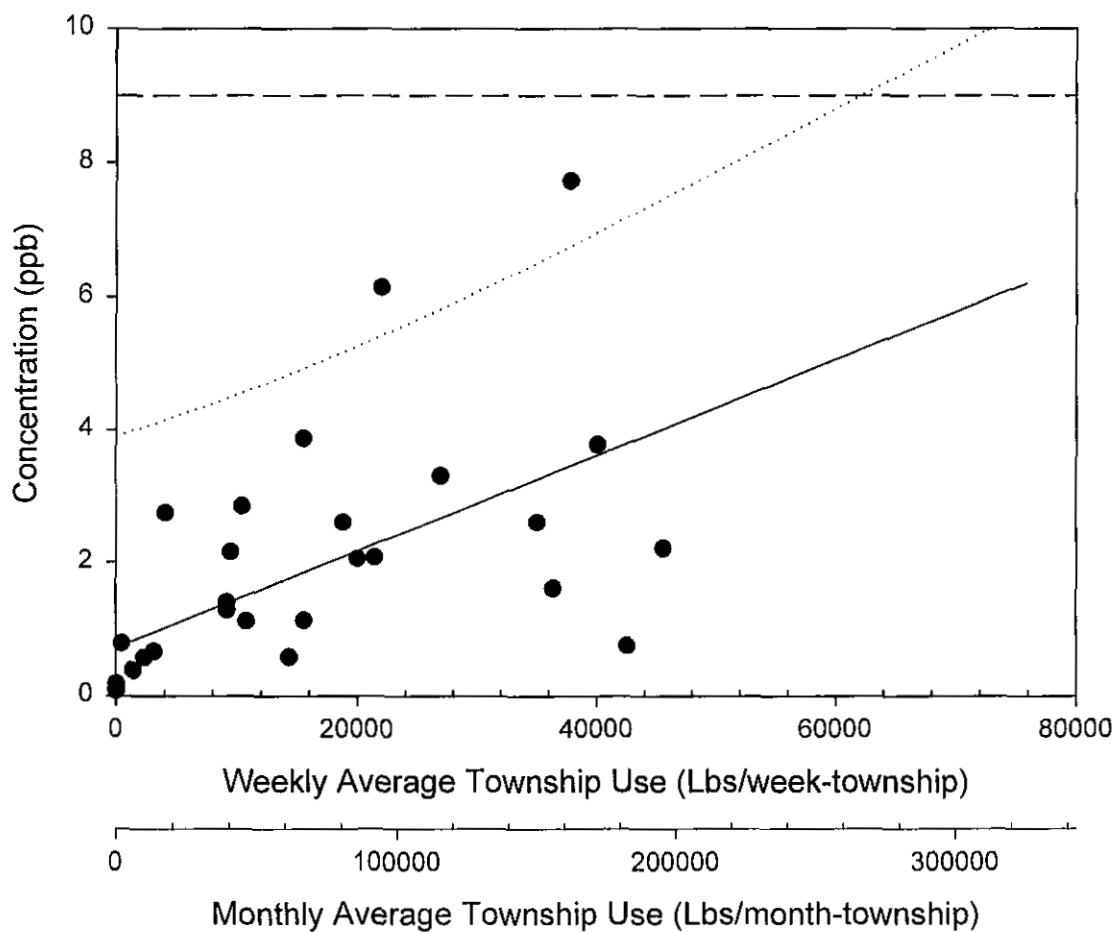


Figure 4. Regression line (solid) and 90% tolerance interval line (dotted) for the 95th percentile for the two-month case. Dashed reference line indicates 9 ppb level and use level below the intersection of tolerance line is 62,108 lbs/week-township or 266,194 lbs/month-township.