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DATE: February 23, 2010

SUBJECT: METHODOLOGY TO DETERMINE METHYL BROMIDE USE CAP

Introduction

This memorandum provides a detailed explanation of the method used to determine the monthly use limit (township cap) to control seasonal exposure to methyl bromide. As discussed in the interagency workgroup and the memorandum on methyl bromide regulations development (January 29, 2010 memorandum to Andrews and Verder-Carlos), this method has been revised from the one used to support the earlier regulations.

In 2004, the Department of Pesticide Regulation (DPR) established an ambient air concentration limit of 9 ppb (as a monthly average) to control subchronic exposure for health concerns in areas of heavy agricultural use (Title 3, California Code of Regulations section 6447[h]). DPR and county agricultural commissioners limit the use of methyl bromide to no more than 270,000 pounds in any township (6 x 6 mile area) in any month as the primary means to ensure that the 9 ppb limit is not exceeded. DPR determined this “township cap” of 270,000 pounds using linear regression to correlate monitored methyl bromide ambient air concentrations and reported use intensity (Li, et al., 2005) and calculating a tolerance interval (Johnson and Li, 2003). The tolerance interval was calculated using data from both Air Resource Board (ARB) monitoring in 2000 and 2001 (ARB, 2001a; ARB, 2001b; ARB, 2002a; ARB, 2002b) and Alliance of Methyl Bromide Industry (AMBI) monitoring in 2002 (AMBI, 2003) studies.

Subsequent to the 2004 regulations, the regression and tolerance interval approach (Johnson and Li 2003) was reviewed by Dr. Robert Spear (Spear, 2004), Dr. Shu Geng (Geng, 2004) and

Dr. Neil Willits (Willits, 2004), University of California peer reviewers. Willits identified several problems: (1) residuals not normally distributed, (2) heterodasticity, and (3) spatial correlation between nearby monitoring sites. For issues (1) and (2), since the time of the original effort (Johnson and Li 2003) to relate subchronic exposures to use information, DPR requested ARB to conduct ambient air monitoring of methyl bromide in Ventura County to evaluate the effectiveness of the township cap. In response ARB conducted monitoring in 2005 and 2006 at sites in areas of high methyl bromide use. These methyl bromide monitoring data (ARB, 2006; ARB, 2007) were added to increase sample size. To address issue (3), nearby monitoring sites were aggregated in this data analysis. In addition, the aggregation of neighboring sites may help to smooth the data in order to satisfy (1) and (2). A drawback to aggregating monitoring sites is the potential loss of degrees of freedom in the statistical analysis and consequent failure to extract trends and information from the data.

Statistical Consultation

In 2009, DPR further consulted Willits on this data analysis for three questions:

1. Were there better statistical methods for our data analysis? Instead of grouping monitoring concentration and methyl bromide use data sets, Willits suggested and performed a mixed regression model ANOVA with covariance structure using 34 individual date sets (Appendix 1). This independent data analysis showed a significant effect of methyl bromide use on ambient air concentration ($P=0.0192$).

2. In 2001 Kern County monitoring study, four out of six sites had no reported methyl bromide use in a township area and one has irregular township size. This resulted in only one data set at site CRS being used in this data analysis (Table 1). The ratio of air concentration to methyl bromide use at this site was much higher than the rest of data. Was it legitimately correct to treat this data set as an outlier? Willits looked into three residuals (observed, studentized, and predicted) and performed with t-test, sign test, and signed rank test for each of them (Appendix 1). All results showed that residual distribution was significantly abnormal (all $P<0.04$) and the site CRS in Kern County in 2001 was not an extreme value (all $P >0.4$). Therefore, the CRS data was included in this data analysis.

3. How could we compare the relationships between methyl bromide use and ambient air concentration before and after implementing the monthly use township cap in 2004 with limited data sets? Willits used another mixed model ANCOVA for shifts in slope analysis to identify any significant changes by introducing a factor of period (before or after 2004). The results showed a significant covariate interaction ($P=0.0336$) of methyl bromide use and period (Appendix 1). However, the data after 2004 were only from Ventura County. Considering the geographical and atmospheric complications in addition to limited sample size, it is not correct to simply conclude that the implementation of monthly township use cap

since 2004 has reduced ambient air concentration. Also, the data from Ventura in 2002 was within the same range as in 2005 and 2006. Therefore, this data analysis attempted to combine both data sets before and after 2004.

Methods and Materials

Grouping monitoring sites

The raw data used in this regression analysis includes ARB and AMBI monitoring studies in 2000, 2001, 2002, 2005, and 2006 (ARB, 2001a; ARB, 2001b; ARB, 2002a; ARB, 2002b; AMBI, 2003; ARB, 2006; and ARB, 2007). A total of 30 monitoring sites are shown on the monitoring site maps (Figures 1, 2, and 3). Twenty three of the 30 sites were used for regression analysis. Six sites, LS and TO in Ventura County (Figure 1), ARB, MVS, VSD, and ARV in Kern County (Figure 3b) were excluded because there was no methyl bromide use in a township area during the monitoring period (LS, TO, and ARB were the selected background sites). The other site, MET (Figure 3b), was also excluded because the sections around MET site were irregularly shaped (Li, et al., 2005).

Figure 1. Monitoring sites in 2005-2006 ARB studies

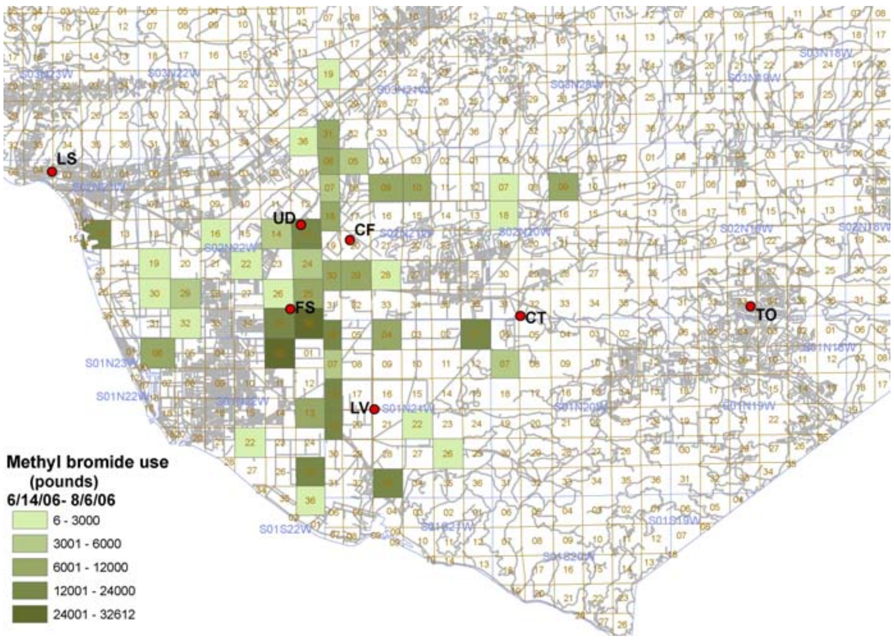
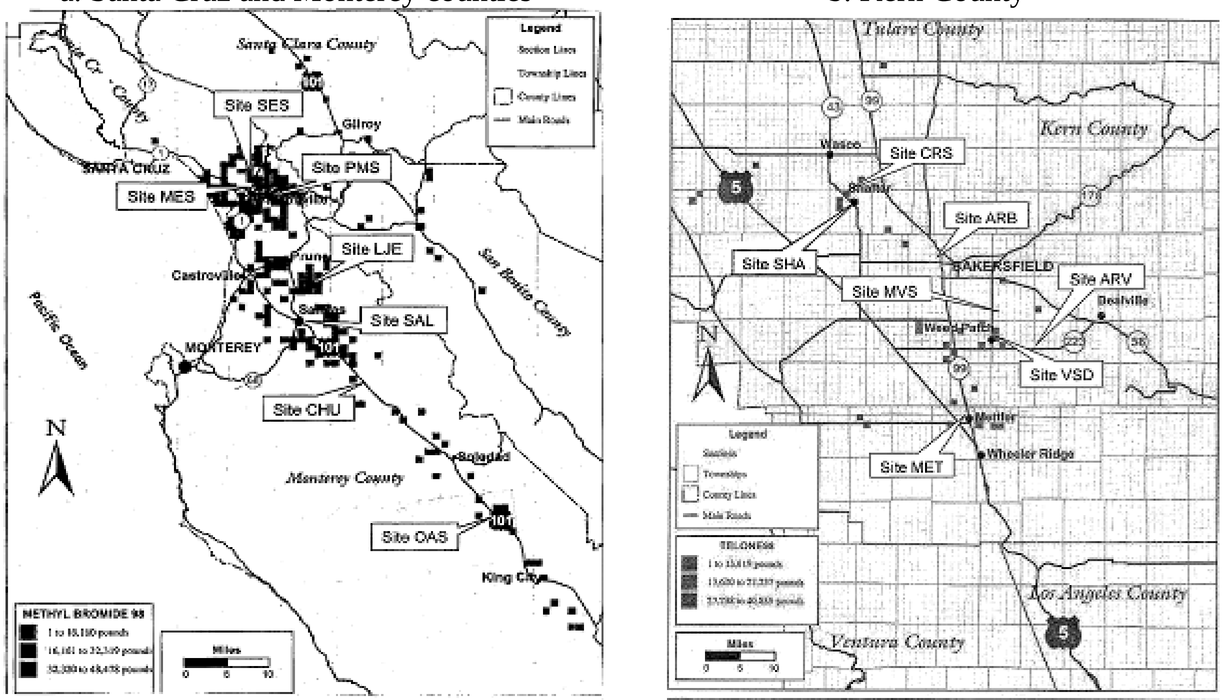


Figure 2. Monitoring sites in 2000-2001 ARB studies

a. Santa Cruz and Monterey counties b. Kern County



Modified from ARB, 2002a *from ARB, 2002b*

Figure 3. Monitoring sites in 2002 ARBI study

a. Ventura County

From AMBI. 2003

b. Santa Cruz County

c. Monterey County

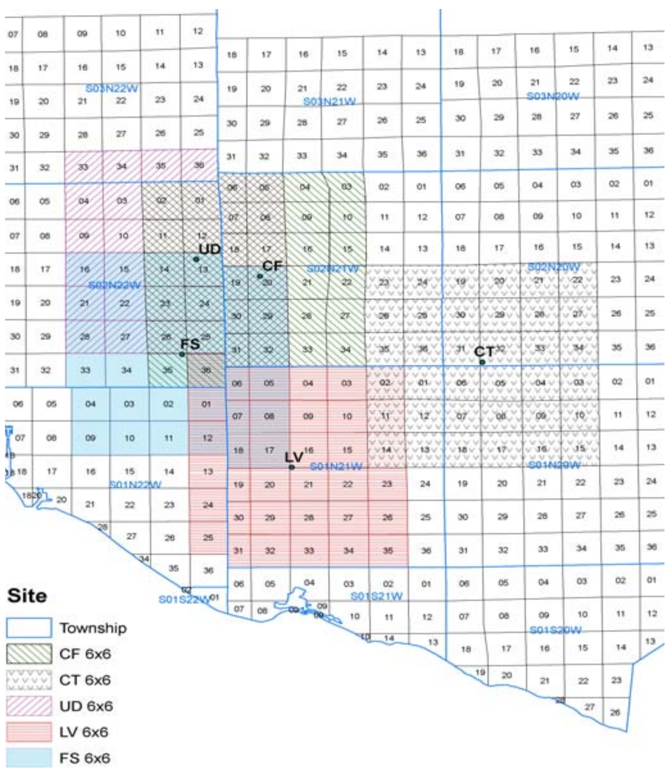
Modified from AMBI, 2003

Modified from AMBI, 2003

Data from different years was assumed to be independent. Eleven of the 23 sites were monitored for multiple years. The total sites x years of monitoring resulted in 34 individually measured ambient air concentrations.

Data from different sites were aggregated when a six mile by six mile township-sized area centered on the monitoring site overlapped with an adjacent six mile by six mile township-sized area for a nearby monitoring site (Figure 4). Li et al. (2005) found that the highest correlations between use and ambient air concentrations occurred with domains approximately the size of townships (6x6 mile square areas). The use of the word township in this memorandum refers to surrounding 6x6 square mile area around each monitoring location. The word township can also refer to formal administrative land survey units within California which are used to identify parcels of land. The use of the word township in this memorandum is clear from the context.

Figure 4. Area of 6x6 sections around each monitoring site



The 23 different site locations were aggregated into 9 locations (see number code in Group column in Table 1) based on overlap of the township-sized areas. This resulted in 9 distinct site locations, and 14 ambient air concentrations for the aggregated sites x years data (Table 2).

Ambient air concentration

The methyl bromide air concentration for each group (Table 2) is the average of the average air concentrations at all sites in that group (Table 1). The air concentration at each site is the average of either weekly average air concentrations (Johnson and Li, 2003) or daily monitoring air concentrations over the monitoring period (Fan, 2008). Daily concentrations were 24-hour ambient air samples collected at each monitoring site on four consecutive days a week for six to eight weeks during methyl bromide peak use seasons (ARB, 2001a; ARB, 2001b; ARB 2002a; ARB, 2002b; AMBI, 2003; ARB, 2006; ARB, 2007).

Agricultural use

Methyl bromide use for each group (Table 2) is the average of the normalized monthly use data at all sites in that group (Table 1). The individual site use data were queried from DPR's Pesticide Use Report database (DPR, 2003; DPR, 2008) with the exclusion of outliers (Wilhoit, 1998). The use data in the database is reported by section that contains the field which received the application. A section is usually a square mile. DPR regulates methyl bromide through the use of limits on application within an administrative township based on the California land survey system.

For 2000-2002 data, weekly methyl bromide use was obtained for 5x5 and 7x7 square mile areas centered on the monitoring sites for the monitoring time periods. The use in 6x6 sections around each monitoring site was estimated by the following formula (Johnson and Li, 2003):

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

where $Use_{(ixi)}$ *refers to reported methyl bromide use in areas of (ixi) sections.*

The monthly use was normalized by:

$$Use_{monthly} = \frac{30}{7} * Use_{weekly}$$

where Use_{weekly} *was the average of the weekly uses during a monitoring period in a township area.*

For 2005-2006 data, methyl bromide use was queried in 6x6 mile areas centered on the monitoring site for each entire monitoring period. The monthly use was normalized by:

$$Use_{monthly} = \frac{30}{days(entire\_monitoring\_period)} * Use_{entire\_monitoring\_period}$$

Table 1. Monitoring air concentration and monthly use in a township-sized area for each site x year combination, showing the Group codes for grouping as explained in the text

County	Year	Site	Group	Concentration (ppb)	Monthly use (pounds)
Monterey	2000	SAL	2	1.29	39268
Monterey	2000	OAS	3	0.39	5903
Monterey	2000	CHU	4	0.67	13319
Monterey	2000	LJE	2	3.88	66358
Santa cruz	2000	PMS	1	7.73	162024
Santa cruz	2000	SES	1	2.61	80511
Kern	2000	SHA	5	0.79	1876
Kern	2000	CRS	5	2.16	40490
Monterey	2001	SAL	2	1.41	39231
Santa cruz	2001	MES	1	6.14	94270
Monterey	2001	CHU	4	0.58	9883
Monterey	2001	LJE	2	2.86	44597
Santa cruz	2001	PMS	1	3.31	115565
Santa cruz	2001	SES	1	1.14	66660
Kern	2001	CRS	5	2.76	17396
Monterey	2002	MAQ	6	1.12	46163
Monterey	2002	BBC	6	2.08	91768
Santa cruz	2002	WAT	7	3.78	171670
Santa cruz	2002	FRM	7	2.62	150016
Santa cruz	2002	CPW	7	2.06	85698
Ventura	2002	ABD	8	0.76	182374
Ventura	2002	SHA2	8	0.59	61328
Ventura	2002	PVW	8	1.62	155828
Ventura	2002	UWC	8	2.22	195241
Ventura	2005	UD	9	0.29	132800
Ventura	2005	CF	9	0.39	122102
Ventura	2005	FS	9	0.17	175564
Ventura	2005	LV	9	0.22	57560
Ventura	2005	CT	9	0.14	54878
Ventura	2006	UD	9	0.57	51149
Ventura	2006	CF	9	0.52	85545
Ventura	2006	FS	9	0.80	97693
Ventura	2006	LV	9	0.88	73867
Ventura	2006	CT	9	0.45	13716

Table 2. Average air concentration and monthly use after grouping overlapped sites

Site group	County	Year	Concentration (ppb)	Monthly use (pounds)
1	Santa cruz	2000	5.17	121267
1	Santa cruz	2001	3.53	92165
2	Monterey	2000	2.58	52813
2	Monterey	2001	2.14	41914
3	Monterey	2000	0.39	5903
4	Monterey	2000	0.67	13319
4	Monterey	2001	0.58	9883
5	Kern	2000	1.48	21183
5	Kern	2001	2.76	17396
6	Monterey	2002	1.60	68965
7	Santa cruz	2002	2.82	135795
8	Ventura	2002	1.29	148693
9	Ventura	2005	0.24	108581
9	Ventura	2006	0.64	64394

Results and Discussion

Regression Analysis

Using the grouped data sets, a parametric linear regression of the ambient air concentration to the methyl bromide use was attempted:

$$Y = a + bX$$

where Y is the methyl bromide air concentration (ppb); X is the monthly methyl bromide use (pounds/month) during the monitoring period; a and b are regression coefficients, intercept and slope, respectively.

Three regression analyses were conducted using Minitab 14 software (Minitab, 2003): A. using all 14 group data sets, B. 3 data sets from Ventura County only, and C. 11 nonVentura County data. The analysis using data sets C was the only one that resulted in a significant regression (P=0.005) (Table 3 and Figure 4C).

Regression A is not statistically significant (P=0.163). There was no correlation between use and air concentration (Table 3, Figure 4A) and a township cap cannot be calculated.

Regression B also shows no statistical significance (P=0.597) (Table 3). Air concentrations detected in Ventura were significantly lower than other areas with comparable use regardless monitoring years (Figures 4A and 4B, additional analysis in Appendix 2). Ventura concentrations could be lower due to differences in application methods between the regions, weather, or other factors, such as geographic location facing the ocean and frequent strong wind blowing during application season. Minimal data size limits any firm conclusions for the low concentrations detected in Ventura.

Regression C is statistically significant (P=0.005) when excluding the data from Ventura County (Table 3, Figure 4C). The coefficient of determination (R^2) for this regression was the highest (0.61). It measured 61% of the air concentration (*Y*)'s variation that was explained by the monthly methyl bromide use (*X*).

Therefore the linear regression model C:

*Air concentration (ppb) = 0.88 + 0.000024 * township use (pounds/month)*

was used for further data analysis. Its residues were plotted in Figure 5 and coefficient estimates were listed in Table 4.

The Ryan-Joiner test, similar to Shapiro-Wilk test (Minitab 14), was performed to test normality of the regression residuals (Figure 6). The test failed to reject normality (P>0.100).

Homogeneity of variance was tested by the Levene test which looked for a linear relationship between the absolute values of the residual errors and the predictor variable (Willits, 2004). The tested linear relationship was not significant (P=0.136) as shown in Table 5. The test failed to establish heterodasticity.

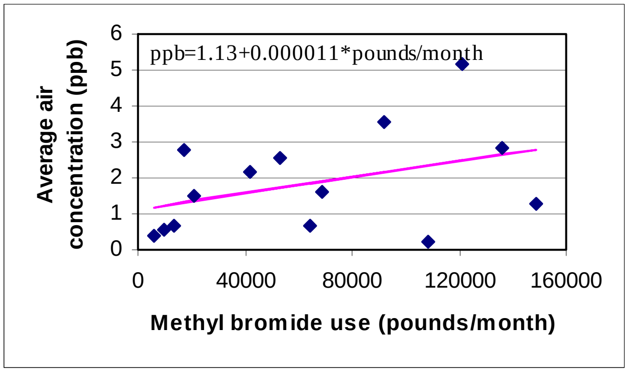
Due to the sample size, these tests are generally lacking in power to definitively reject normality or heterodasticity. At issue in the case of heterodasticity is whether the residuals show a fan-shaped pattern (residuals versus fitted values). If the large residual at x=17396 is removed, the plot suggests increasing variance with x (Figure 5). Thus while the technical requirements for regression are satisfied, the small sample size and a suggestive residual plot weaken confidence that the regression assumptions are satisfied.

Table 3. Regression Coefficient of determination (R^2) and mean square error (MSE)

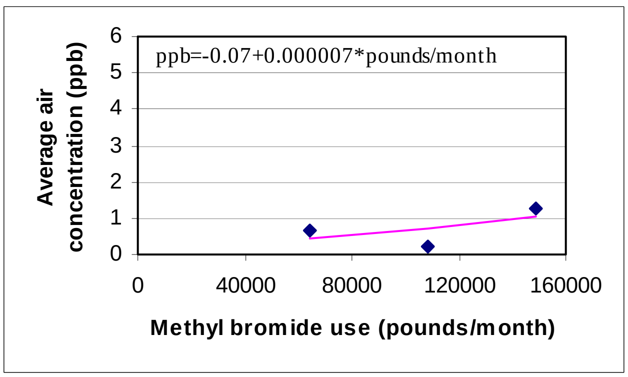
Data source	DF of residual error	R^2	MS		P
			regression	residual error	
A. all group data	12	0.16	4.027	1.826	0.163
B. only Ventura data	1	0.35	0.196	0.366	0.597
C. without Ventura data	9	0.61	12.46	0.897	0.005

Figure 4. Linear regression analyses using group data sets

A. all group data sets



B. only Ventura County data sets



C. excluding Ventura County data sets

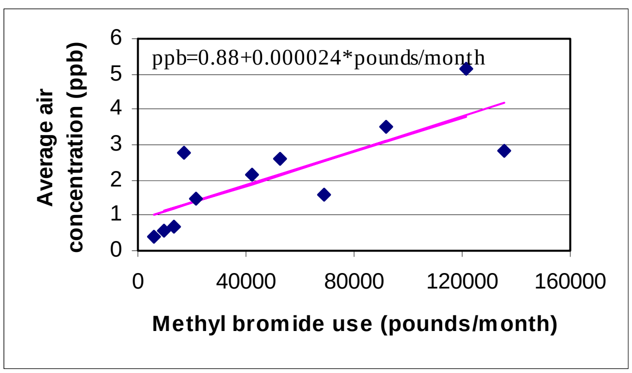


Table 4. Coefficient estimates for regression model C

	Coefficients	Standard Error	t Stat	P	Lower 95%	Upper 95%
Intercept	0.88	0.446	1.98	0.079	-0.13	1.89
Slope	0.000024	0.000006	3.73	0.005	0.000009	0.000039

Figure 5. Residual plots for the regression model C

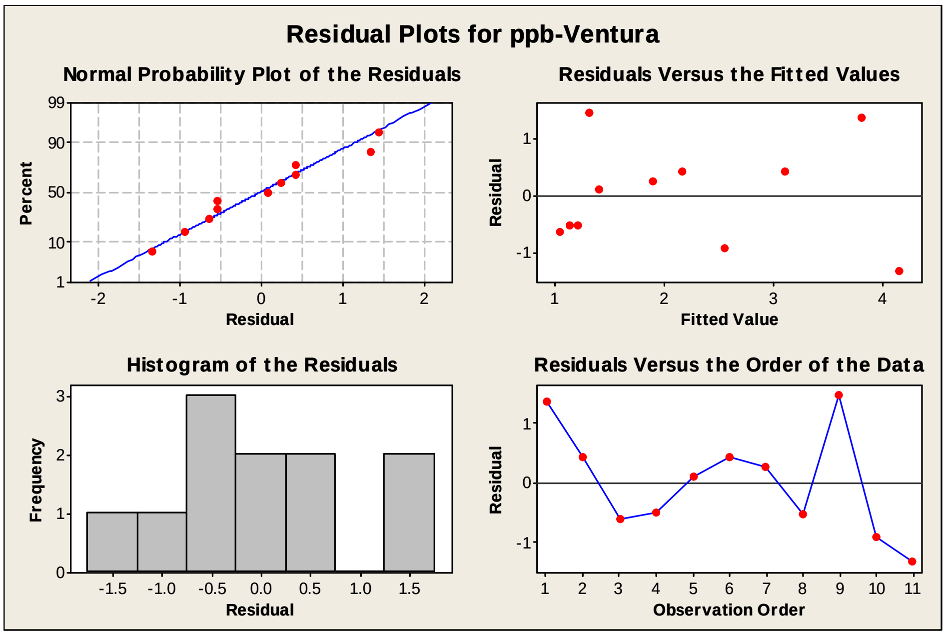


Figure 6. Ryan-Joiner Normality test for residues of regression model C

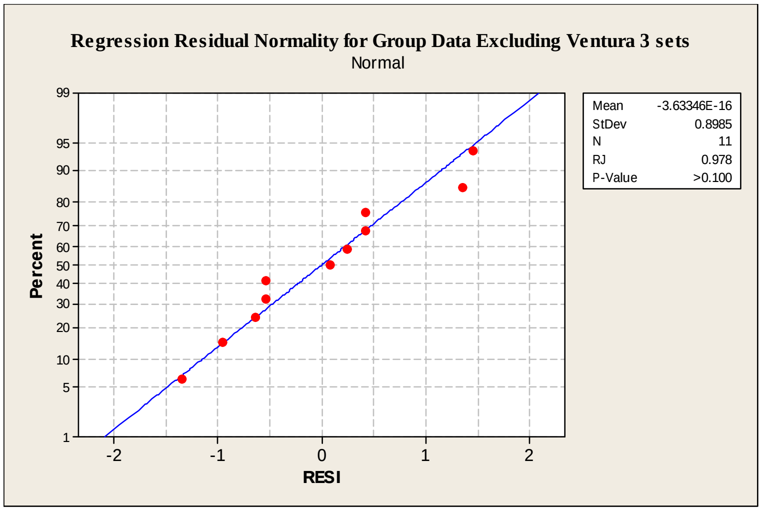


Table 5. ANOVA of the Levene test for linear regression residuals

Source	DF	SS	MS	F	P
Regression	1	0.521	0.521	2.6	0.136
Residual Error	9	1.748	0.194		
Total	10	2.269			

Methyl bromide monthly township use caps

The relationship between use and air concentrations for Ventura was different from the other areas (Figure 4). Air concentrations detected in Ventura (2002, 2005 and 2006) were significantly lower than other areas with comparable use levels. DPR staff are uncertain if the lower Ventura concentrations are due to differences in application methods between the regions or other factors. Ventura data were excluded from the regression used to determine township caps for several reasons. First, by excluding the Ventura data, the correlation between use and air concentration becomes statistically significant and a township cap can be calculated. Second, the data are more representative of the area affected by the township cap. Nine of the 11 data points represent monitoring in Monterey and Santa Cruz Counties. Most if not all townships affected by the cap are in Monterey and Santa Cruz Counties. Third, the slope of the regression line without Ventura data is steeper than the regression line including Ventura data. This results in more health-protective township caps for use levels greater than 60,000 pounds in a township. Fourth, there were several laboratory problems during the 2005 monitoring. Repeated instrument failures caused some samples to be held past the verified hold time, laboratory control samples were not included in the initial sets of samples because a gas standard was unavailable, and other problems. Some of the 2005 data may have questionable validity. DPR staff have determined the recommended township cap from the regression that excludes Ventura data (model C). As specified in the risk management directive, the regulatory target level should be in the range of 2-9 ppb (Reardon 2009). Accordingly, the monthly township use caps can be derived based on regression model C (Table 6).

Table 6. Methyl bromide monthly township use caps

Ambient air concentration limits (ppb)	Township use derived by model (pounds/month)
2	46,667
3	88,333
4	130,000
5	171,667
6	213,333
7	255,000
8	296,667
9	338,333

Of the 202 California townships which had some methyl bromide use in 2008, DPR staff compiled the highest methyl bromide monthly use figures based on the 2008 Pesticide Use Report (Table 7). These are the townships that would most likely be affected by a cap on the township use. The township, county, reported use and corresponding regression model C concentration estimates are shown in Table 7.

Table 7. Highest monthly township methyl bromide use from 2008 Pesticide Use Report

Township	County	2008 Max Monthly Use (lbs)	Estimated Concentration (Model C - ppb)
M12S02E	MONTEREY/SANTA CRUZ	241,140	6.7
M14S02E	MONTEREY	196,620	5.6
M46N01W	SISKIYOU	153,084	4.6
M15S03E	MONTEREY	138,542	4.2
S10N34W	SANTA BARBARA	124,524	3.9
M12S01E	MONTEREY/SANTA CRUZ	79,650	2.8
S01N21W	VENTURA	79,080	2.8
S10N33W	SANTA BARBARA/SAN LUIS OBISPO	70,166	2.6
M14S03E	MONTEREY	67,083	2.5
S11N35W	SAN LUIS OBISPO	62,191	2.4
M47N01E	SISKIYOU	58,968	2.3
M11S02E	SANTA CRUZ	55,116	2.2
M14N03E	SUTTER	50,820	2.1
M20S17E	FRESNO	50,352	2.1

Conclusion

This data analysis found a linear regression model between monthly methyl bromide use and ambient air concentrations using aggregated data sets excluding data from Ventura County. The model weakly satisfied the regression assumptions of residual distribution normality and homogeneity of variance. The regression model was used to calculate monthly township caps based on various levels of concentration and to calculate monthly concentrations corresponding to 2008 monthly use in high methyl bromide use townships in California. The exclusion of Ventura data results in more health-protective township caps for use levels greater than 60,000 pounds in a township.

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Appendix 1
Statistical consultation summary from Willits (2009)

Here are the analyses I ran for you. I've attached four files. The first is a Word-formatted file containing the SAS output from those analyses. The others are probability plots for various forms of the residual errors from those analyses. The mixed model ANOVAs that I ran were primarily to see if the outliers in the analyses she had run were due to the fact that a few surprisingly large observations were also ones for which there was less replication (averaging over usage sites within a township) going on. These analyses included the township (called group) in the model and so if there was appreciable site to site variability within a township, it would "expect" an underreplicated observation to be more variable than one that was based on a greater number of replicate usage sites. In the first proc mixed, the estimated township to township variability was comparable in magnitude to the residual error, which means that those underreplicated townships _would_ be seen as less extreme. In this analysis, there was a significant usage effect (p = .0192), which just means that there seems to be a non-zero relationship between usage and ambient concentrations.

The next part of the output (starting on SAS page 3 of the output -- look above the date to find these page numbers). Since there are two sources of randomness, there are several ways of calculating the residual errors, but all three of them are telling pretty much the same story. The outliers that came to Shifang's attention don't look that extreme in the residual plots. As such, it's hard to argue that they should be deleted because they're so extreme. (Mind you, I've said in the past that I don't like doing this, but even if I did, it would be hard to justify here.) At the same time, the residual distributions are significantly non-normal (all three have p-values in the range of .01 to .02) so the application of a normal tolerance interval to these data is hard to justify. There are nonparametric alternatives to parametric tolerance intervals, but they require a larger sample size than she has available.

The final part of the output (starting on SAS' page 12) contains the results of another mixed model analysis that looks at the question of whether the relationship between usage and ambient levels has changed since the latest set of regulations were implemented. The answer to this is "yes", as reflected by the significant interaction between the covariate (i.e., usage) and the ambient level (p = .0336).

If you have any questions about these results, please let me know.

-Neil Willits
Department of Statistics.

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regression with random group effect                              1
                                                                09:45
Thursday, December 10, 2009

                                       The Mixed Procedure

                                       Model Information

                     Data Set                        WORK.SFAN
                     Dependent Variable              ppb
                     Covariance Structure            Variance Components
                     Estimation Method               REML
                     Residual Variance Method        Profile
                     Fixed Effects SE Method         Model-Based
                     Degrees of Freedom Method       Containment


                                     Class Level Information

                        Class    Levels    Values

                        year          5    2000 2001 2002 2005 2006
                        group         9    1 2 3 4 5 6 7 8 9


                                           Dimensions

                              Covariance Parameters            2
                              Columns in X                     2
                              Columns in Z                     9
                              Subjects                         1
                              Max Obs Per Subject             34


                                     Number of Observations

                          Number of Observations Read             34
                          Number of Observations Used             34
                          Number of Observations Not Used          0


                                        Iteration History

                   Iteration    Evaluations    -2 Res Log Like
Criterion

                           0              1       152.07188922
                           1              3       140.36597859
0.02861658
```

2	2	138.81819670	0.00287888
3	2	138.74834094	0.00047299
4	1	138.72794164	0.00000815
5	1	138.72761193	0.00000000

Convergence criteria met.

regression with random group effect

The Mixed Procedure

Covariance Parameter Estimates

Cov Parm	Estimate
group	1.2166
Residual	1.3064

Fit Statistics

-2 Res Log Likelihood	138.7
AIC (smaller is better)	142.7
AICC (smaller is better)	143.1
BIC (smaller is better)	143.1

Type 3 Tests of Fixed Effects

Effect	Num DF	Den DF	F Value	Pr > F
usage	1	24	6.30	0.0192

normality of residuals

The UNIVARIATE Procedure

Variable: Resid (Residual)

Moments

N	34	Sum Weights	34
Mean	0	Sum Observations	0
Std Deviation	1.02202975	Variance	1.04454482
Skewness	1.01723541	Kurtosis	2.83517555
Uncorrected SS	34.469979	Corrected SS	34.469979
Coeff Variation	.	Std Error Mean	0.17527666

Basic Statistical Measures

Location		Variability	
Mean	0.00000	Std Deviation	1.02203
Median	-0.06758	Variance	1.04454
Mode	.	Range	5.46902
		Interquartile Range	1.05097

Tests for Location: Mu0=0

Test	-Statistic-		-----p Value------	
Student's t	t	0	Pr > |t|	1.0000
Sign	M	-3	Pr >= |M|	0.3915
Signed Rank	S	-37.5	Pr >= |S|	0.5295

Tests for Normality

Test	--Statistic---		-----p Value------	
Shapiro-Wilk	W	0.919589	Pr < W	0.0157
Kolmogorov-Smirnov	D	0.154101	Pr > D	0.0398
Cramer-von Mises	W-Sq	0.169067	Pr > W-Sq	0.0132
Anderson-Darling	A-Sq	0.984658	Pr > A-Sq	0.0125

Quantiles (Definition 5)

Quantile	Estimate
100% Max	3.2194067
99%	3.2194067

```
                                    95%            2.4258410
                                    90%            1.0809277
                                    75% Q3         0.3964934
                                    50% Median    -0.0675824
                                     normality of residuals
4
                                                                  09:45
Thursday, December 10, 2009

                                    The UNIVARIATE Procedure
                                  Variable:  Resid  (Residual)

                                    Quantiles (Definition 5)

                                    Quantile        Estimate

                                    25% Q1        -0.6544774
                                    10%           -0.9424248
                                    5%            -1.4659792
                                    1%            -2.2496092
                                    0% Min        -2.2496092


                                       Extreme Observations

                              ------Lowest------        -----Highest-----

                                  Value      Obs           Value      Obs

                              -2.249609       14         0.71713       12
                              -1.465979       28         1.08093       15
                              -1.225760       21         1.48133        4
                              -0.942425        6         2.42584       10
                              -0.843297       25         3.21941        5


                           Stem Leaf                     #             Boxplot
                              3 2                        1                0
                              2
                              2 4                        1                0
                              1 5                        1                |
                              1 1                        1                |
                              0 677                      3                |
                              0 0112244                  7             +--+--+
                             -0 3332111110              10             *-----*
                             -0 9887776                  7             +-----+
                             -1 2                        1                |
                             -1 5                        1                |
                             -2 2                        1                0
                                ----+----+----+----+
```

```
                                     normality of residuals
5
                                                               09:45
Thursday, December 10, 2009

                                    The UNIVARIATE Procedure
                                  Variable:  Resid  (Residual)

                                         Normal Probability Plot
                      3.25+                                              *
                          |
+
                          |                                         *  +++++
                          |                                       +++++
                          |                                  ++*+*
                          |                             ++++***
                          |                        +++******
                          |                   ********
                          |            *******+
                          |       *  *++++
                          |     +++++
                     -2.25+ +++*+
                           +----+----+----+----+----+----+----+----+----+----
+
                               -2        -1        0         +1        +2

                                     normality of residuals
6
                                                               09:45
Thursday, December 10, 2009

                                    The UNIVARIATE Procedure
                             Variable:  StudentResid  (Studentized Residual)

                                             Moments

                 N                           34    Sum Weights
34
                 Mean                -0.0020973    Sum Observations    -
0.0713096
                 Std Deviation       1.00727083    Variance
1.01459452
                 Skewness            1.05564029    Kurtosis
2.9236927
                 Uncorrected SS      33.4817686    Corrected SS
33.4816191
```

```
                 Coeff Variation    -48026.071    Std Error Mean
0.17274552


                                   Basic Statistical Measures

                         Location                    Variability

                     Mean     -0.00210     Std Deviation            1.00727
                     Median   -0.06235     Variance                 1.01459
                     Mode       .          Range                    5.40551
                                           Interquartile Range      0.99796


                                   Tests for Location: Mu0=0

                        Test           -Statistic-    -----p Value------

                        Student's t    t  -0.01214    Pr > |t|    0.9904
                        Sign           M        -3    Pr >= |M|   0.3915
                        Signed Rank    S     -39.5    Pr >= |S|   0.5076


                                      Tests for Normality

                   Test                  --Statistic---    -----p Value------

                   Shapiro-Wilk          W     0.919914    Pr < W      0.0161
                   Kolmogorov-Smirnov    D     0.150955    Pr > D      0.0476
                   Cramer-von Mises      W-Sq  0.164148    Pr > W-Sq   0.0157
                   Anderson-Darling      A-Sq  0.958119    Pr > A-Sq   0.0150


                                    Quantiles (Definition 5)

                                    Quantile        Estimate

                                    100% Max       3.2237208
                                    99%            3.2237208
                                    95%            2.3287671
                                    90%            1.0994548
                                    75% Q3         0.3679625
                                    50% Median    -0.0623469
                                     normality of residuals
7
                                                                09:45
Thursday, December 10, 2009

                                    The UNIVARIATE Procedure
                         Variable:  StudentResid  (Studentized Residual)
```

```
                    Quantiles (Definition 5)

                    Quantile        Estimate

                    25% Q1        -0.6300004
                    10%           -0.9075477
                    5%            -1.4600988
                    1%            -2.1817892
                    0% Min        -2.1817892


                       Extreme Observations

           ------Lowest------        -----Highest-----

               Value      Obs            Value      Obs

           -2.181789       14          0.70353       12
           -1.460099       28          1.09945       15
           -1.233748       21          1.45484        4
           -0.907548        6          2.32877       10
           -0.790778       25          3.22372        5


      Stem Leaf                     #             Boxplot
         3 2                        1                0
         2
         2 3                        1                0
         1 5                        1                |
         1 1                        1                |
         0 677                      3                |
         0 0112244                  7             +-----+
        -0 4332111110              10             *--+--*
        -0 9887766                  7             +-----+
        -1 2                        1                |
        -1 5                        1                |
        -2 2                        1                0
           ----+----+----+----+
```



```
                    The UNIVARIATE Procedure
       Variable:  StudentResid  (Studentized Residual)

                         Normal Probability Plot
```

```
                   3.25+                                     *
                       |
+
                       |                                   *  +++++
                       |                                +++++
                       |                             ++*+*
                       |                          ++++***
                       |                       ++******
                       |                    ********
                       |              *******+
                       |          *  *++++
                       |        +++++
                  -2.25+ +++*+
                        +----+----+----+----+----+----+----+----+----+----
+
                             -2        -1         0         +1        +2

                                   normality of residuals
9
                                                                   09:45
Thursday, December 10, 2009

                                  The UNIVARIATE Procedure
                         Variable:  PearsonResid  (Pearson Residual)

                                           Moments

                 N                          34    Sum Weights
34
                 Mean                        0    Sum Observations
0
                 Std Deviation       0.8941795    Variance
0.79955698
                 Skewness           1.01723541    Kurtosis
2.83517555
                 Uncorrected SS     26.3853804    Corrected SS
26.3853804
                 Coeff Variation             .    Std Error Mean
0.15335052


                                 Basic Statistical Measures

                       Location                    Variability

                    Mean      0.00000     Std Deviation            0.89418
                    Median   -0.05913     Variance                 0.79956
                    Mode       .          Range                    4.78487
                                          Interquartile Range      0.91950
```

Tests for Location: Mu0=0

Test	-Statistic-		-----p Value------	
Student's t	t	0	Pr > |t|	1.0000
Sign	M	-3	Pr >= |M|	0.3915
Signed Rank	S	-37.5	Pr >= |S|	0.5295

Tests for Normality

Test	--Statistic---		-----p Value------	
Shapiro-Wilk	W	0.919589	Pr < W	0.0157
Kolmogorov-Smirnov	D	0.154101	Pr > D	0.0398
Cramer-von Mises	W-Sq	0.169067	Pr > W-Sq	0.0132
Anderson-Darling	A-Sq	0.984658	Pr > A-Sq	0.0125

Quantiles (Definition 5)

Quantile	Estimate
100% Max	2.8166768
99%	2.8166768
95%	2.1223817
90%	0.9457096
75% Q3	0.3468942
50% Median	-0.0591282

normality of residuals

10

09:45

Thursday, December 10, 2009

The UNIVARIATE Procedure
Variable: PearsonResid (Pearson Residual)

Quantiles (Definition 5)

Quantile	Estimate
25% Q1	-0.5726059
10%	-0.8245327
5%	-1.2825934
1%	-1.9681956
0% Min	-1.9681956

```
                                Extreme Observations

                        ------Lowest------        ------Highest-----

                            Value      Obs            Value      Obs

                        -1.968196       14         0.627421       12
                        -1.282593       28         0.945710       15
                        -1.072425       21         1.296028        4
                        -0.824533        6         2.122382       10
                        -0.737805       25         2.816677        5


                      Stem Leaf                     #             Boxplot
                         2 8                        1                0
                         2 1                        1                0
                         1
                         1 3                        1                |
                         0 5669                     4                |
                         0 0111234                  7             +--+--+
                        -0 3322111100              10             *-----*
                        -0 8776665                  7             +-----+
                        -1 31                       2                |
                        -1
                        -2 0                        1                0
                           ----+----+----+----+
```

```
                                 normality of residuals
11
                                                                   09:45
Thursday, December 10, 2009

                                The UNIVARIATE Procedure
                         Variable:  PearsonResid  (Pearson Residual)

                                    Normal Probability Plot
                     2.75+                                                *
                         |                                            *
+++
                         |                                            ++++++
                         |                                       ++*++
                         |                                 +++****
                     0.25+                           +++******
                         |                      ********
                         |               *******
                         |          *++*++
                         |     +*++++
                    -2.25++++
```

```
                          +----+----+----+----+----+----+----+----+----+----
+
                              -2        -1         0        +1        +2
```


The Mixed Procedure

Model Information

Data Set	WORK.RESIDUALS
Dependent Variable	ppb
Covariance Structure	Variance Components
Estimation Method	REML
Residual Variance Method	Profile
Fixed Effects SE Method	Model-Based
Degrees of Freedom Method	Containment

Class Level Information

Class	Levels	Values
year	5	2000 2001 2002 2005 2006
period	2	older recent
group	9	1 2 3 4 5 6 7 8 9

Dimensions

Covariance Parameters	2
Columns in X	6
Columns in Z	9
Subjects	1
Max Obs Per Subject	34

Number of Observations

Number of Observations Read	34
Number of Observations Used	34
Number of Observations Not Used	0

Iteration History

Iteration	Evaluations	-2 Res Log Like	Criterion
0	1	160.17554669	
1	3	151.23560173	0.00002232
2	1	151.23455688	0.00000003
3	1	151.23455535	0.00000000

Convergence criteria met.

The Mixed Procedure

Covariance Parameter Estimates

Cov Parm	Estimate
group	1.2661
Residual	1.0885

Fit Statistics

-2 Res Log Likelihood	151.2
AIC (smaller is better)	155.2
AICC (smaller is better)	155.7
BIC (smaller is better)	155.6

Solution for Fixed Effects

Effect	period	Estimate	Standard Error	DF	t Value	Pr > |t|
Intercept		0.5461	1.3348	7	0.41	0.6947
usage		-1.19E-6	7.375E-6	23	-0.16	0.8731
period	older	0.05892	1.4647	23	0.04	0.9683

period	recent	0	.	.	.	.
usage*period	older	0.000021	9.158E-6	23	2.26	0.0336
usage*period	recent	0	.	.	.	.

Type 3 Tests of Fixed Effects

Effect	Num DF	Den DF	F Value	Pr > F
usage	1	23	4.00	0.0575
period	1	23	0.00	0.9683
usage*period	1	23	5.11	0.0336

Appendix 2. Comparison of data from Ventura to data from other counties

Data from Ventura County were different from other counties regardless monitoring year. The three group data sets, one was in 2002 monitored by AMBI and the other two were in 2005 and 2006 monitored by ARB, generally showed high use but low air concentrations.

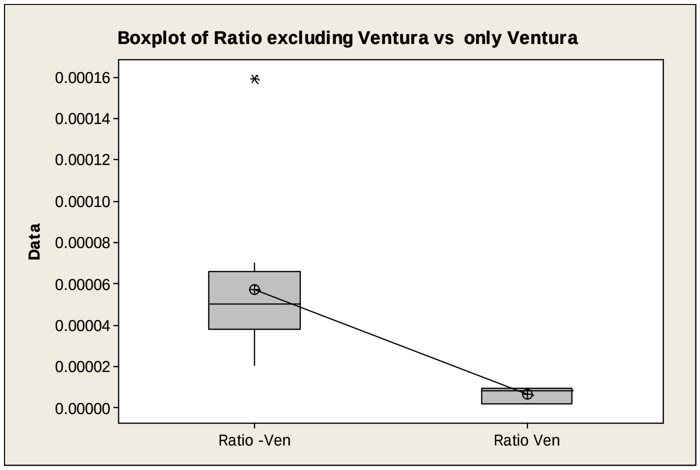
To statistically compare data from Ventura to those from other counties, the ratio of the air concentration to the normalized monthly township use was calculated for each group (Table 1). Student t-test was performed to compare the mean of the ratios for data excluding Ventura County vs. those of only Ventura County. The results showed significant difference at P=0.04 (Table 1). Figure 1 is a boxplot of this comparison.

Table 1. t-test for the ratio of air concentration/monthly township use using group data sets

	Excluding Ventura		**Only Ventura**
	County	**Ratio**	**Ratio**
Ratio of concentration/use (ppb/pounds per month)	Santa Cruz	4.26E-05	8.68E-06
	Santa Cruz	3.83E-05	2.21E-06
	Monterey	4.89E-05	9.94E-06
	Monterey	5.11E-05	
	Monterey	6.61E-05	
	Monterey	5.03E-05	
	Monterey	5.87E-05	
	Kern	6.99E-05	
	Kern	1.59E-04	
	Monterey	2.32E-05	
	Santa Cruz	2.08E-05	

	Excluding Ventura	Only Ventura
N	11	3
Mean	5.72E-05	6.95E-06
Standard deviation	3.72E-05	4.15E-06
SE mean	1.10E-05	2.40E-06
DF	12	
T-value	2.27	
P-value	0.04	

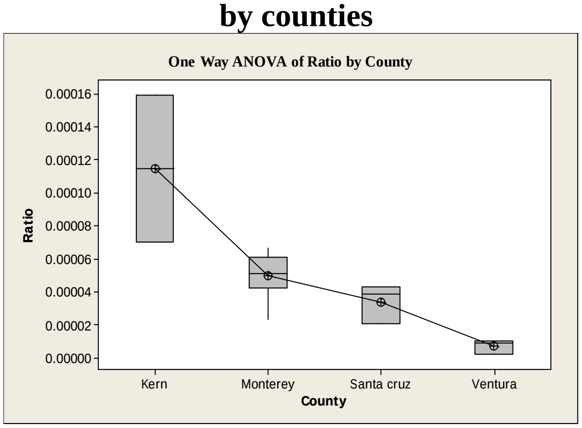
Figure 1. Boxplot comparison of excluding Ventura versus Ventura only



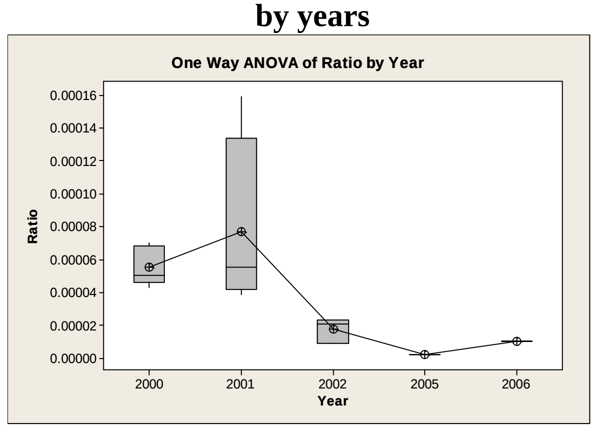
Further analysis of One Way ANOVA compared the ratio of the grouped air concentration to the grouped normalized monthly township use by counties and years (Figure 2), respectively (The data were unbalanced, so not suitable for Two Way ANOVA). The results showed a significant difference between counties (P=0.003), and the result of residual normality test was P=0.07. However, there was no significant difference among years (P=0.143), and the residues was not normally distributed ($p<0.01$). Figure 2 showed that the ratio of the air concentration to the normalized monthly township use for Ventura County was much lower than all other counties.

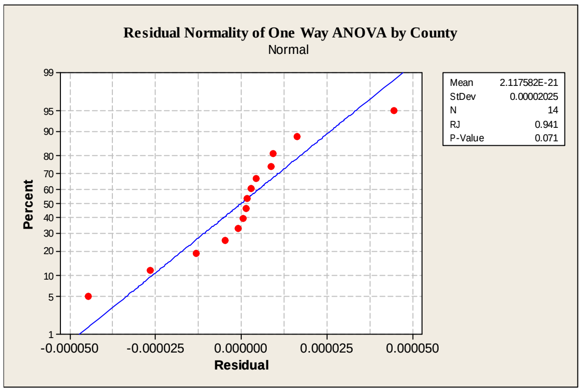
Figure 2. One Way ANOVA for the ratio of the air concentration to the normalized monthly township use:

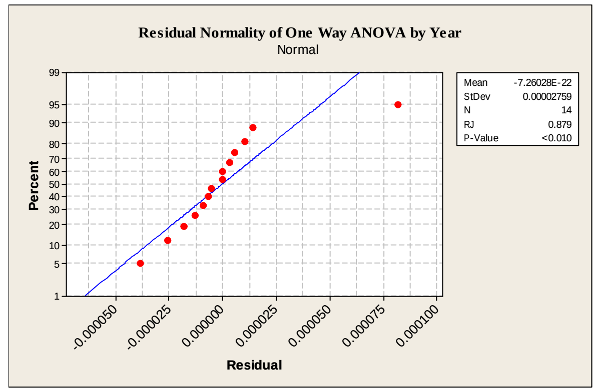
by counties



by years




Residual Normality of One Way ANOVA by County
Normal
Percent
99
95
90
80
70
60
50
40
30
20
10
5
1
-0.000050
-0.000025
0.000000
0.000025
0.000050
Residual
Mean 2.117582E-21
StDev 0.00002025
N 14
RJ 0.941
P-Value 0.071


Residual Normality of One Way ANOVA by Year
Normal
Percent
99
95
90
80
70
60
50
40
30
20
10
5
1
-0.000050
-0.000025
0.000000
0.000025
0.000050
0.000075
0.000100
Residual
Mean -7.26028E-22
StDev 0.00002759
N 14
RJ 0.879
P-Value <0.010