Analysis of methyl bromide ambient air concentration data monitored by the Air Resources Board and the Alliance of Methyl Bromide Industry in year 2001

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1. Introduction

In recent years, the Department of Pesticide Regulation (DPR) has evaluated seasonal exposure risks of people to the soil fumigant methyl bromide. The Air Resources Board (ARB) monitoring in Monterey/Santa Cruz Counties in 2000 indicated that subchronic concentrations of methyl bromide in several monitored areas were higher than the 1 ppb reference concentration established by the Department of Pesticide Regulation (DPR)[1,2,3]. In order to understand causes of these ambient air concentrations, DPR staff analyzed the ARB's air monitoring data, Pesticide Use Reports(PUR), and weather conditions during the monitoring period of the year 2000[4,5]. Some empirical relationships were established between the air concentration and the methyl bromide use in various areas surrounding the monitoring sites, and it was concluded that use was responsible for the concentrations observed [4,5].

DPR has taken a series of steps and measures to address this issue, including requesting additional air monitoring by ARB in 2001 and initiating a reevaluation. As a result, the ARB conducted air monitoring in 2001 in the same area and period as in 2000, and the Alliance of the Methyl Bromide Industry (AMBI) conducted air monitoring in two other high use areas within Ventura and Santa Barbara counties.

The ARB recently completed reports which summarized monitoring results in Monterey/Santa Cruz counties and Kern County in 2001[6,7]. The AMBI also submitted a final report of their monitoring results conducted in Oxnard/Camarillo of Ventura County and Santa Maria of Santa Barbara County in August-October, 2001 [8]. Based on the monitoring results and PUR data, the DPR staff established regression models that link the air concentration to the nearby methyl bromide use. This report documents the procedure and results of this data analysis. The objectives of this study are (1) to characterize the causal relationship between the ambient air concentration and methyl bromide use in surrounding areas, (2) to validate the empirical models based on the ARB 2000 monitoring data, and (3) to explore whether or not adjustments to the use by time, distance and wind direction improve regressions between air concentration and use.

2. Methods and Materials

2.1 Location of Monitoring Sites

The ARB conducted air monitoring at six sites in each area, and five of them were the same as used in year 2000 (Figures 1, 2 and 3). The new site in Monterey/Santa Cruz area was MacQuiddy Elementary School (MES), and the Arvin High School (ARV) in Kern County. The location references (Meridian, township, range and section [MTRS]) of monitoring sites in the Public Land Survey System (PLSS) are listed in Table 1.

Table 1a: Location of ARB air monitoring sites in 2001

ID	County	Site Name	Symbol	MTRS	Note
1	Monterey	MBUAPCD Ambient Monitoring Station	SAL	M14S03E22	old
2	Monterey	Chualar School	CHU	M16S04E03	old
3	Monterey	La Joya Elementary	LJE	M14S03E10	old
4	Monterey	Pajaro Middle School	PMS	M12S02E09	old
5	Santa Cruz	Salsepuedes Elementary School	SES	M11S02E22	old
6	Santa Cruz	MacQuiddy Elementary School	MES	M11S02E33	new
7	Kern	ARB Ambient Air Monitoring Station	ARB	M29S27E34	old
8	Kern	Arvin High School	ARV	M31S29E23	new
9	Kern	Cotton Research Station	CRS	M27S25E33	old
10	Kern	Mountain View School	MVS	M30S29E30	old
11	Kern	Vineland School District- Sunset School	VSD	M31S29E19	old
12	Kern	Mettler Fire School	MET	S11N20W01	old

Because the MET site was on the boundary of two meridian systems (Mount Diablo and San Bernardino), and the surrounding use areas did not consist of regular squares of 1x1 mile^2, it was dropped from regression analysis.

The AMBI monitoring sites in Santa Barbara and Ventura Counties are shown in Figures 4 and 5. The actual PLSS sections in Ventura County do not follow the regular 1x1 grid pattern. The county agricultural commissioner's office uses a normalized grid layout (as shown in Figure 5) to document locations of pesticide applications in preparing pesticide use reports. Therefore, this report will use the normalized grid to denote MTRS locations of the monitoring sites in Ventura County (Table 1b).

Table 1b: Location of AMBI air monitoring sites in 2001

ID	County	Site Name	Symbol	MTRS	Note
1	Santa Barbara	Plantell Nursery	PLN	S10N33W32	PNT
2	Santa Barbara	Edward Community Center	EDW	S10N34W12	
3	Santa Barbara	Agriculture Commissioner's Office	AGC	S09N34W03	
4	Santa Barbara	Blosser Road	BLO	S10N34W09	
5	Ventura	Sharps Automotive	SHA	S01N22W22	
6	Ventura	Abandoned Building	ABD	S02N22W27	
7	Ventura	United Water Conservation District	UWC	S02N21W32	
8	Ventura	Pleasant Valley Water District pump station	PVW	S01N21W03	

2.2 Methyl Bromide Air Concentrations

The ARB air sampling took place between September 08th and November 07th in Monterey/Santa Cruz Counties, and between June 30th and August 30th in Kern County. The AMBI monitoring in Santa Barbara and Ventura Counties was conducted from August 15th to October 10th. The daily average air concentrations for the ARB and AMBI monitoring are summarized in Tables 2a and 2b.

Table 2a. Summary of ARB 2001 methyl bromide monitoring results for Monterey/Santa Cruz and Kern Counties (ppb).

Week	Monterey/Santa Cruz								Kern							
	Date	CHU	LJE	MES	PMS	SAL	SES	Jday*	Date	ARB	ARV	CRS	MET	MVS	VSD	Jday
1	09/08/01	0.33	0.59	0.07	0.44	0.48	0.13	251	06/30/01	0.04	0.03	0.26				181
	09/09/01	0.94	6.22	3.79	4.19	0.99	1.55	252	07/01/01	0.03		0.11	0.02	0.02		182
	09/10/01	0.74	4.83			4.31		253	07/02/01	0.31		0.07	0.02			183
	09/11/01		5.39					254								
2	09/17/01	0.24	0.31	1.09	1.16	0.17	1.14	260	07/06/01	0.03	0.05	0.06	0.03			187
	09/18/01	0.2	0.24	0.76	0.74	0.21	1	261	07/07/01	0.02	0.02	0.02	0.02		0.02	188
	09/22/01	0.48	1.19	1.01	1.71	0.55	2.83	265	07/08/01	0.02	0.02	0.02	0.02	0.02	0.02	189
	09/23/01	0.25	0.52	0.69	1.14	0.32	1.04	266								
3	09/24/01	0.33	0.76	6.58	2.56	0.46	1.55	267	07/13/01	0.15	0.13	25.13	0.06		0.13	194
	09/25/01	1.42	0.69	19.05	13.15	1.21	0.5	268	07/14/01	0.07	0.03	0.17			0.04	195
	09/26/01	0.48	0.26	10.78	3.25	0.28	2.32	269	07/15/01	0.04		0.26	0.02	0.03		196
	09/27/01	0.49	0.22	18.7	13.11	0.31	1.13	270	07/16/01	0.15	0.06		0.04	0.13	0.04	197
4	10/03/01	0.35	0.43	0.37	0.62	0.33	0.44	276	07/21/01	0.16	0.21	1.22	0.15	0.17	0.23	202
	10/04/01		0.17	1.9	0.8	0.12	0.77	277	07/22/01	0.05	0.05	8.27	0.05	0.04	0.05	203
	10/05/01	0.24	0.32	2.14	1.61	0.3	1.09	278	07/23/01	0.11		0.53		0.09	0.06	204
	10/06/01	0.73	0.39	36.65	2.93	2.42	0.32	279	07/24/01	0.04	0.04	0.36	0.03	0.05	0.04	205
5	10/11/01	0.97	1.47	23.61	2.83	2.45	1.36	284	07/29/01	0.05	0.03	0.66	0.03	0.03	0.02	210
	10/12/01	1.18	2.9		21.09	8.91		285	07/30/01	0.11	0.05	0.07	0.03	0.04	0.06	211
	10/13/01	1.84	7.41		10.01	9.25		286	07/31/01	0.04	0.04	0.71	0.04	0.04	0.04	212
	10/14/01	0.66	6.43	4.54	7.95	4.14		287	08/01/01	0.07	0.03	0.06	0.03	0.04	0.03	213
6	10/19/01	0.8	2.59	2.56	2.01	1.24	2.03	292	08/06/01	0.29	0.22	25.34	0.25	0.23	0.21	218
	10/20/01		0.43	0.35	0.38	0.36	0.24	293	08/07/01	0.23		4.38	0.12	0.16		219
	10/21/01	0.25	0.14	1.72	0.39	0.14	0.6	294	08/08/01	0.12	0.1	10.59	0.09		0.09	220
	10/22/01	1.01	9.24	7.65	2.06	0.81	5.31	295	08/09/01	0.12	0.06	0.59	0.06	0.06	0.06	221
7	10/27/01	0.57	4	4.38	2.44	1.01	0.34	300	08/14/01	0.18	0.15	0.47	0.13	0.17	0.14	226
	10/28/01	0.72	13.4	3.12	0.53	0.58		301	08/15/01	0.24		0.94	0.17	0.15	0.23	227
	10/29/01	0.58	14.9	3.98	1.56	0.6	0.49	302	08/16/01	0.15		0.64	0.07	0.11	0.1	228
	10/30/01	0.28	4.13		0.34	0.48	0.07	303	08/17/01		0.12	0.62	0.13	0.11	0.11	229
8	11/04/01	0.29	0.79		0.15	0.3		308	08/22/01	0.2	0.1	5.75	0.07	0.09	0.1	234
	11/05/01	0.11	0.35	0.95	0.35	0.16	0.19	309	08/23/01	0.13	0.13	1.77	0.19	0.11	0.14	235
	11/06/01	0.11	0.39	2.48	1.25	0.1	1.03	310	08/24/01	0.08	0.05	0.15	0.04	0.07	0.06	236
	11/07/01	0.27	0.35	0.31	0.41	0.17	1.86	311	08/25/01	0.06	0.06	0.17	0.05	0.06	0.07	237
9									08/28/01	0.08		0.09		0.07		240
									08/29/01	0.31	0.08	0.71	0.08	0.1	0.12	241
									08/30/01	0.08	0.08	0.13	0.06	0.08	0.07	242

* Jday is the Julian day of the monitoring date.
Blank cells indicate no data available on those dates.

Table 2b. Summary of 2001 AMBI methyl bromide monitoring results in Ventura and Santa Barbara (ppb)

Week	Ventura						Santa Barbara					
	DATE	PVW	UWC	SHA	ABD	Jday*	DATE	BLO	AGC	EDW	PLN	Jday
1	08/15/01	1.82	2.58	0.69		226	08/23/01	0.04	0.03	0.02		234
	08/16/01	1.05	1.85	0.17		227	08/24/01	0.03	0.13	1.02		235
	08/17/01	3.17	1.80	0.18		228	08/25/01	0.68	0.11	0.69		236
							08/26/01	3.46	0.13	1.33	0.34	237
2	08/21/01	0.50	1.53			232	08/27/01	2.09	0.14	0.98	0.68	238
	08/22/01	1.91	0.45			233	08/28/01	0.19	0.06	0.44	0.10	239
	08/23/01	2.49	4.35	2.94		234	08/29/01	0.34	0.02	0.32	1.29	240
	08/24/01		2.01	3.38		235	08/30/01	0.30	0.06	0.58	1.68	241
	08/25/01	0.81	0.25	1.09		236						
3	08/28/01	0.12	0.21	1.09		239	09/04/01	0.07	0.05	0.30	0.22	246
	08/29/01	0.15	0.10	0.07		240	09/05/01	0.17	0.05	0.09	0.43	247
	08/30/01	0.28	0.35	0.56	0.44	241	09/06/01	0.21	0.13	0.59	0.51	248
	08/31/01	0.15	0.18			242	09/07/01	0.11		0.20		249
4	09/06/01	0.20		0.04	0.05	248	09/11/01	1.47	0.15	1.30	1.81	253
	09/07/01	0.10		0.03	0.13	249	09/12/01		0.21	0.68	0.78	254
	09/08/01	0.07		0.05	0.13	250	09/13/01	0.40	0.21	0.64	0.59	255
	09/09/01	0.16		0.23	0.39	251	09/14/01	0.51	0.20	1.01	1.07	256
5	09/13/01	0.17		0.38	0.07	255	09/16/01	0.78				258
	09/14/01	0.15		0.07	0.10	256	09/17/01	0.31	0.14	0.54	0.57	259
	09/15/01			0.13	0.15	257	09/18/01	0.33	0.37	0.83		260
	09/16/01	0.29		0.11	0.11	258	09/19/01	0.42	0.30	0.49		261
6	09/17/01	0.35		0.13	0.14	259	09/24/01	2.22	0.20	4.09	1.24	266
	09/18/01					260	09/25/01	1.12		7.08		267
	09/19/01	0.18	0.18	0.10	0.10	261	09/26/01	0.34	0.42	11.15	0.55	268
	09/20/01	0.59	0.30	0.36	0.11	262	09/27/01	1.20	0.72	4.05	0.83	269
7	09/26/01	0.17	0.60	0.45	0.25	268	09/30/01	4.55		6.08	2.69	272
	09/27/01	0.08		0.09	0.12	269	10/01/01	0.24	0.90	0.38	1.98	273
	09/28/01	0.08		0.10	0.15	270	10/02/01	0.52	1.16	0.68	1.85	274
	09/29/01	0.11		0.19	0.19	271	10/03/01	0.24	0.48	0.22	1.43	275
	09/30/01	0.15		0.07	0.06	272						
8	10/07/01	0.06	0.07	0.10	0.04	279	10/06/01	0.58	0.08	0.36	0.82	278
	10/08/01	0.05	0.05	0.05	0.06	280	10/07/01	0.52	0.21		0.93	279
	10/09/01	0.09	0.07	0.07	0.10	281	10/08/01	0.21	0.17	0.26	0.21	280
	10/10/01	0.10	0.07	0.11	0.11	282	10/09/01	1.04	0.39	0.82	2.26	281

* Jday is the Julian day of the monitoring date.
Blank cells indicate no data available on those dates.

The emission of methyl bromide could last serval days after application, and about 98% escaped from treated soil within a week[9]. The air concentration detected instantaneously likely resulted from applications in various days of the previous week, and an application could influence air concentration measurements on many days in the following week. Therefore, instead of analyzing daily air concentrations, we used one week as the basic time unit for this analysis. For this reason, monitoring days were grouped into concentration weeks(Tables 2a and 2b) that were compatible to the week definition of data analysis of 2000 air monitoring[4]. Unlike the ARB 2000 air monitoring[1,2] where air sampling was conducted from Monday through Thursday, the 2001 air monitoring[6,7] included some weekends and did not follow a regular pattern. Instead of using a calendar week, a concentration week was organized based on clusters of sampling days, which might be consecutive or separated. Most weeks contained four consecutive sampling days. However, there were a few weeks containing less or more sampling days, and in one extreme case, only one sampling day. Weeks that had no data were dropped from this analysis.

The weekly average air concentration was calculated as the arithmetic average of daily concentrations in that week. The mean of weekly average air concentrations was defined as the sum of weekly averages divided by the number of weeks. The objective of this analysis was to correlate air concentrations with methyl bromide use, weekly and seasonal air concentrations calculated here might differ from those of DPR's exposure and risk assessments.

2.3 Methyl Bromide Use and Adjustments

It was assumed that methyl bromide use amount in a certain size of area and during a certain period of time was responsible for the variation of observed air concentrations. Therefore, specification of use period and use area was essential to quantify the use, and to correlate the air concentration to the use. One problem is, however, we usually don't know the spatial and temporal scales at which the cause-effect relationship operates best. Our approach to this problem is to aggregate or integrate methyl bromide use gradually both over time and space, and to conduct correlation and regression analysis between use and air concentration at each step during the aggregation process. This assessment leads to the definition of the best spatial-temporal scales depending on the most explanatory regression results.

2.3.1 Use Week

Because the soil flux of methyl bromide might last several days after application, a use week was defined as a concentration week plus three days prior to the first sampling day of the concentration week. In most cases, a concentration week consisted of four consecutive monitoring days, therefore, a use week consisted of seven days started three days earlier than the concentration week. In this study, these weeks are called regular weeks. There were a few weeks that did not satisfy this requirement (Tables 2a and 2b). In Monterey/Santa Cruz counties, for example, week 2 had two samples in the first two days, and two samples in the last two days of the week. Therefore, the use week covered 7 days starting from the first sampling day to the last sampling day. As the emission of methyl bromide from soil declines quickly with time, shift of a use week relative to a concentration week might cause some noise in correlation between use and concentration. This would be especially true when there was only one or two sampling days in a concentration week. In all cases, a use week always contained seven days, while the number of sampling days in a concentration week varied.

2.3.2 Use Area

Methyl bromide use surrounding a monitoring site was aggregated by square areas(Figure 6). The size of squares ranged from 1x1, 3x3, ..., 15x15 mile^2, all using the monitoring site as the centroid [4]. This arrangement of use areas was determined by section, the basic unit of location in the PUR database. A section was referenced by meridian base, township, range and section number (MTRS) in the PLSS system.

2.3.3 Use Adjustments

When summarizing methyl bromide use over an area and a time period, each application was treated equally. For example, the total use in the 5x5 area as shown in Figure 7 was calculated as the sum of each individual applications. However, closer applications had greater influence on measured air concentrations than more distant applications. Similarly, applications that occur immediately before or during the air monitoring had more impact on monitoring. The application site - monitoring site alignment relative to the wind direction was another important factor. Upwind applications would have more influences on measured air concentrations than those occurred at downwind locations. Therefore, when integrating use over time and space, each use needs to be weighted specifically based on its application time, location and wind direction. This study also explored adjustments to use amount by time, distance and wind direction to help explain the variations in measured ambient air concentrations.

2.3.3.1 Time Adjustment

Methyl bromide soil emissions last several days after an application, but decline quickly with time. The decline pattern can be affected by many factors, such as application methods, weather conditions and soil conditions. Table 3 described decline patterns of daily average emission fractions of methyl bromide under various application methods[9].

Table 3. Daily average emission fractions of methyl bromide from four application methods

application methods	**days after application**								
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**
shallow notarp	0.37	0.32	0.15	0.07	0.04	0.02	0.01	0.01	0.01
tarp	0.24	0.21	0.10	0.05	0.02	0.01	0.01	0.01	
bedtarp	0.80	0.10	0.04	0.02	0.01	0.01	0.01		
deep notarp	0.32	0.25	0.14	0.08	0.05	0.04	0.03		
Average	0.433	0.220	0.108	0.055	0.030	0.020	0.015	0.010	0.010

In the study areas, all four application methods might exist, and the percentage of each application method employed was not known. Therefore, average emission fractions from the four application methods were used to characterize the rate of soil emission. On average, emissions in the first three days accounted for 75% of total soil emissions, and 98% in one week after application.

As described in section 2.3.1, a regular concentration week usually consisted of four monitoring days, while the responsible use week consisted of seven days. Table 4 illustrated the percentage of emission that could be captured by air sampling from applications at various days in the use week. The time weight coefficient of a use (f_t) was defined as the sum of its daily average emission fractions over the monitoring period. Because different emission days as well as different number of emission days were covered by the monitoring period, applications in various days relative to the monitoring days had different weighting coefficients. For example, emission from an application in the first day was not measured until day 4 when emission became very weak. Although emissions of this application were sampled in the rest four days, the sum of emission fractions remained small ($f_t = 0.12$). Therefore, the use in the first day had minimum impact on the weekly average air concentration. Another example was for an application in the fourth day of a use week. Because the air sampling started on the same day, emissions from this application had the maximum potential to be measured in the following four days. Therefore, it had the maximum impact on the measured weekly average air concentration ($f_t = 0.815$). The influence of an application decreased gradually if it was applied after the monitoring started (Table 4):

Table 4. Daily average emission fractions and time weight coefficients (f_t) for applications occurred in various days of a use week

monitoring	**no**	**no**	**no**	**yes**	**yes**	**yes**	**yes**	
day in a use week	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**f_t***
1	0.433	0.220	0.108	0.055	0.030	0.020	0.015	**0.120**
2		0.433	0.220	0.108	0.055	0.030	0.020	**0.213**
3			0.433	0.220	0.108	0.055	0.030	**0.413**
4				0.433	0.220	0.108	0.055	**0.815**
5					0.433	0.220	0.108	**0.760**
6						0.433	0.220	**0.653**
7							0.433	**0.433**

Note: f_t is the time weight coefficient, measuring the impact of an application on the weekly average air concentration measurements. It was calculated as the sum of daily average emission fractions over the monitoring days (from day 4 to day 7).

It should be noted that f_t represents the degree of impact of a use on the weekly average air concentration. Also, the time adjustment based on Table 4 works better with a regular concentration week and a regular use week.

2.3.3.2 Distance Adjustment

Applications closer to a monitoring site have more impact on the monitored ambient concentration than those that occurred far away from the monitoring site. The distance in miles (D) from the center of an application section to the center of the monitoring section was calculated, and the weight coefficient for distance was defined as:

$$f_d = 1/D \qquad (1)$$

D was assigned 0.5 mile if a use was in the same section as the monitoring site, and thus the weight coefficient was 2. An example was given in Figure 8 to illustrate the procedure of distance adjustment.

2.3.3.4 Wind Direction Adjustment

An application upwind from the monitoring site should have more impacts on the air sampling. Wind direction was the direction from which wind blew, and was measured in degrees clockwise from true north. In order to evaluate the wind direction effects, the orientation of each section in the use area relative to the monitoring site was to be determined. The orientation of a use section was defined as the clockwise angle in degrees from true north of the use section with the monitoring section at the center of the circle. For example, sections due north of the monitoring site were 0^o or 360^o, and due east were 90^o. Because daily average wind direction might cancel important hours of potential contribution, hourly wind direction was used to calculate the wind direction adjustment coefficient:

$$f_w = \frac{1}{24}\sum_{h=0}^{23} \max[0, \cos(\alpha_h - \beta)] \qquad (2)$$

where f_w was the daily average wind direction adjustment coefficient, α_h was the hourly average wind direction and β was the orientation of a use section respectively. $\cos(\alpha_h - \beta)$ was a positive number if $|\alpha_h - \beta|$ was in the range of (0, 90) or (270, 360), otherwise $\cos(\alpha_h - \beta) < 0$. $\cos(\alpha_h - \beta)$ decreased correspondingly when the difference between wind direction and section direction $(\alpha_h - \beta)$ increased from 0 to 90^o. As defined by equation (2), f_w changed between 0 and 1. $f_w = 1$ only if wind blew from the section of application in all 24 hours during that day (i.e., $\alpha_h = \beta$, and $h = 0, 1, 2, ..., 23$).

If an application was in the same section of the monitoring site, its location relative to the monitoring site and the wind direction couldn't be determined from the PUR report. For this special case, $f_w = 0.5$. Figure 9 illustrated the calculation of wind direction adjusted use.

Hourly average wind direction data was obtained from CIMIS (California Irrigation Management Information System) weather stations. A weather station at North Salinas (station 116) was used for Monterey/Santa Cruz area, which on average was the closest weather station to all monitoring sites in the area[5]. For monitoring sites in Kern County, weather station 125 was used. Choosing representative weather stations was often compromised by many factors, such as proximity to the monitoring sites and availability of records for the period of concern.

2.3.3.5 Adjustment by All Factors

A multiplicative model was used to account for the combined effects of all three factors:

$$f = f_t \times f_d \times f_w \qquad (3)$$

2.4 Relate Air Concentration to the Methyl Bromide Use

Linear regression model was used to relate the air concentration to the methyl bromide use:

$$Y = a + bX \tag{4}$$

where Y was the average air concentration over certain periods (1 week, 4 weeks and 8 weeks), and X was the weekly average methyl bromide use over various areas in those periods. Depending on the analysis, X could be use pounds, or use pounds modified by various factors as described in section 2.3.3. For instance, considering a use area of *m*x*m* mile^2 and in a period of one week, the total adjusted use in this spatial-temporal domain was calculated as:

$$X = \sum_{i=1}^{m} \sum_{j=1}^{m} \sum_{k=1}^{7} f_t(i, j, k) \times f_d(i, j, k) \times f_w(i, j, k) \times U(i, j, k) \tag{5}$$

here *i* and *j* denote the row and the column of a section in the *mxm* area, and *k* denotes the day of use. Therefore, $U(i, j, k)$ represents use pounds in the section (i, j) that occur on the use day *k*. $f_t(i, j, k)$, $f_d(i, j, k)$ *and* $f_w(i, j, k)$ represent respectively adjustment factors of use time, distance and wind direction.

The ARB and AMBI monitoring data were used separately for the regression analysis. The least squares method was used to estimate regression coefficients a and b for using adjusted and unadjusted use over various spatial and temporal domains. R^2 and the Mean Error Square(EMS) measured the relative fitness of the linear regression models.

2.5 Comparison of ARB 2000 and 2001 Regression Models

Regression models based on the ARB 2001 monitoring data were compared to the regression models using the ARB 2000 monitoring data. Regression lines from the year 2000 data and their 95% confidence intervals were plotted. New data from year 2001 and the corresponding regression analyses were overlaid on the same graphs and compared. Also, parameters of regression models for the two years were compared. Confidence intervals for predicted values and model parameters were calculated using methods described in [10]. If regression models using ARB 2001 monitoring data are not significantly different from those based on ARB 2000 monitoring data, data from two years will be pooled together to conduct regression analysis.

3. Results

3.1 Air Concentration

Weekly average air concentrations in various sites were calculated (Tables 5a and 5b). The mean of weekly average air concentrations was also calculated over a period of 8 or 9 weeks.

Table 5a. Weekly average air concentrations, and the mean of weekly average air concentrations over the 8/9 week period in Monterey/Santa Cruz and Kern counties(ppb)

Week	Monterey/Santa Cruz						Kern					
	CHU	**LJE**	**MES**	**PMS**	**SAL**	**SES**	**ARB**	**ARV**	**CRS**	**MET**	**MVS**	**VSD**
1	0.67	4.26	1.93	2.32	1.93	0.84	0.13	0.03	0.15	0.02	0.02	n/a*
2	0.29	0.57	0.89	1.19	0.31	1.50	0.02	0.03	0.04	0.03	0.02	0.02
3	0.68	0.49	13.78	8.02	0.56	1.37	0.10	0.07	8.52	0.04	0.08	0.07
4	0.44	0.33	10.26	1.49	0.79	0.66	0.09	0.10	2.60	0.08	0.09	0.09
5	1.16	4.55	14.07	10.47	6.19	1.36	0.07	0.04	0.37	0.03	0.04	0.04
6	0.69	3.10	3.07	1.21	0.64	2.05	0.19	0.12	10.22	0.13	0.15	0.12
7	0.54	9.09	3.83	1.22	0.67	0.30	0.19	0.13	0.67	0.12	0.13	0.15
8	0.19	0.47	1.25	0.54	0.18	1.03	0.12	0.08	1.96	0.09	0.08	0.09
9	n/a	n/a	n/a	n/a	n/a	n/a	0.16	0.08	0.31	0.07	0.08	0.10
Mean	0.58	2.86	6.14	3.31	1.41	1.14	0.12	0.08	2.76	0.07	0.08	0.09

n/a: none data available

Table 5b. Weekly average air concentrations, and the mean of weekly average air concentrations over the 8 week period in Ventura and Santa Barbara counties(ppb)

Week	Ventura				Santa Barbara			
	PVW	**UWC**	**SHA**	**ABD**	**BLO**	**AGC**	**EDW**	**PLN**
1	2.01	2.08	0.35	n/a*	1.05	0.10	0.76	0.34
2	1.43	1.72	2.47	n/a	0.73	0.07	0.58	0.94
3	0.18	0.21	0.57	0.44	0.14	0.08	0.30	0.39
4	0.13	n/a	0.09	0.18	0.79	0.19	0.91	1.06
5	0.20	n/a	0.17	0.11	0.46	0.27	0.62	0.57
6	0.37	0.24	0.20	0.12	1.22	0.45	6.59	0.87
7	0.12	0.60	0.18	0.16	1.39	0.85	1.84	1.99
8	0.07	0.06	0.08	0.08	0.59	0.21	0.48	1.06
Mean	0.56	0.82	0.51	0.18	0.80	0.28	1.51	0.90

n/a: none data available

The maximum weekly average air concentration was 14.07ppb at the MacQuiddy Elementary School (MES) of Santa Cruz County in week 5. The maximum mean of weekly average air concentrations over an eight-week period was 6.14 ppb and also occurred at MES, followed by Pajaro Middle School (PMS) with 3.31 ppb. The air concentrations monitored by AMBI in Santa Barbra and Ventura Counties were much lower than those by ARB in Monterey/Santa Cruz and Kern Counties.

The weekly average air concentrations of the ARB 2001 monitoring were also shown on Figure 10. The weekly average air concentrations in four sampling sites of the Monterey/Santa Cruz counties peaked in week 5. The air concentrations at MES and PMS sites were consistently higher than other sites in Monterey/Santa Cruz counties, and those at CHU and SES were consistently lower. Air concentrations in CRS of the Kern County were consistently higher.

3.2 Use Amounts at Various Areas

As expected, methyl bromide use amount increased with the size of area(Tables 6a and 6b). There were very few applications within monitoring sections (1x1 area). The MES site had uses in the monitoring section also exhibited the highest average air concentration over 8 weeks. As the spatial scale increased, differences in use between monitoring sites diminished. In Table 5a, for example, the high and the low use amounts differed by a factor of about 10 for the 3x3 area, but a factor of about 3 over the 15x15 area.

Table 6a: The means of weekly average air concentrations and the average of weekly methyl bromide use (lb/week) over various spatial scales centered on monitoring sites in Monterey/Santa Cruz and Kern Counties

County	Site	Concentration (ppb)	Weekly Methyl Bromide Use (lb/week)							
			1x1	3X3	5X5	7X7	9X9	11X11	13X13	15X15
Monterey	SAL	1.41	0	1417	7806	10872	23476	36672	46794	51452
Santa Cruz	MES	6.14	3034	7224	16181	28872	39391	52252	55096	58744
Monterey	CHU	0.58	0	561	1317	3475	6912	13551	18529	21538
Monterey	LJE	2.86	0	3150	7518	13820	23889	31082	40790	46707
Monterey	PMS	3.31	0	4458	17439	38231	57111	63801	66301	67926
Santa Cruz	SES	1.14	0	5452	12275	19429	27580	41171	44346	53127
Kern	ARB	0.12	0	0	0	0	0	0	0	0
Kern	ARV	0.08	0	0	0	0	0	0	0	0
Kern	CRS	2.76	0	2737	4059	4059	7452	7452	9438	9438
Kern	MVS	0.08	0	0	0	0	1570	1570	1570	1570
Kern	VSD	0.08	0	0	0	0	0	0	0	0

Table 6b: The means of weekly average air concentrations and the average of weekly methyl bromide use (lb/week) over various spatial scales centered on monitoring sites in Santa Barbara and Ventura Counties

County	Site	Concentration (ppb)	Weekly Methyl Bromide Use (lb/week)							
			1x1	3X3	5X5	7X7	9X9	11X11	13X13	15X15
Santa Barbara	PLN	0.90	156	4394	17365	17899	22195	24389	31280	36642
Santa Barbara	EDW	1.51	3828	4159	10708	21727	28773	34334	36437	36642
Santa Barbara	AGC	0.28	0	526	4922	19356	22889	29296	35687	35778
Santa Barbara	BLO	0.80	0	7484	11256	27100	29323	32848	36528	36731
Ventura	SHA	0.51	0	2089	9137	12761	27899	35841	37300	40658
Ventura	ABD	0.18	0	4549	10672	27009	36628	41553	44901	44928
Ventura	UWC	0.82	0	3814	10091	23899	40930	49955	52006	57849
Ventura	PVW	0.56	0	0	9549	19280	29076	46804	48215	51166

In Figure 11, the mean of weekly average air concentrations was compared to average of weekly use at various spatial scales ranging from 3x3 to 13x13 mile^2 for each site in Monterey/Santa Cruz and Kern Counties. A high concentration usually corresponded to a high use, but there were a few exceptions which did not follow this trend. For example, the concentrations at the CRS site of Kern County were higher than those measured at some sites of Monterey/Santa Cruz Counties, but the use surrounding the CRS site was not proportionally high in large areas.

3.3 Correlation and Regression Models

R^2 and EMS values of regression models over various spatial and temporal scales are shown for the ARB 2001 data(Table 7a) and for the AMBI 2001 data(Table 7b). The threshold value for a significant regression decreases with the increase of sample size. All regressions using ARB 2001 monitoring data are significant at 95% significant level (Table 7a). The best regression was over the 3x3 area, and correlation gradually decreased when the use area increased. In terms of residuals (EMS), regressions over the 4-week period were better than those over the one week periods, and regressions over the 8-week period were the best (Figures 12 and 13). This trend was consistent with the regression models using the ARB 2000 data. However, 2000 regression effects were better than those using the year 2001 data.

Table 7a: R^2 between average air concentration (ppb) and average methyl bromide usage (lb/week) over various areas and periods using ARB 2001 monitoring data

Area	Time period					
	1 week (n * = 93)		4 weeks (n = 22)		8 weeks (n = 11)	
	R^2 **	EMS ***	R^2	EMS	R^2	EMS
area 1x1	0.147	8.04	0.394	2.39	0.593	1.67
area 3x3	0.288	6.71	0.322	2.68	0.742	1.05
area 5x5	0.197	7.57	0.325	2.67	0.644	1.45
area 7x7	0.178	7.75	0.326	2.67	0.590	1.68
area 9x9	0.158	7.93	0.305	2.75	0.558	1.81
area 11x11	0.163	7.89	0.298	2.77	0.542	1.87
area 13x13	0.164	7.87	0.294	2.79	0.521	1.96
area 15x15	0.146	8.05	0.265	2.90	0.488	2.09
Significant R^2 values						
$R^2_{0.10}$	0.018		0.081		0.176	
$R^2_{0.05}$	0.030		0.130		0.271	

* n is the number of samples for the regression.

** R^2 is often referred as the coefficient of determination, representing the decimal fraction of variation of air concentration that is explained by the regression model.

*** EMS is the average squared residuals (errors) not explained by the model.

Using AMBI 2001 monitoring data, the best regressions are for one week period(Table 7b). Almost no correlation is observed over the eight-week period except for the 1x1 area (Figures 14 and 15). The trend of AMBI regression models over the temporal scale contrasts with the corresponding trend of ARB 2000 and 2001regression models, where longer periods result in higher R^2values.

Table 7b: R^2 between average air concentration (ppb) and average methyl bromide usage (lb/week) over various areas and periods using AMBI 2001 monitoring data

Area	**Time period**					
	1 week (n = 63)		**4 weeks (n = 16)**		**8 weeks (n = 8)**	
	R^2	EMS	R^2	EMS	R^2	EMS
area 1x1	0.657	0.331	0.474	0.140	0.653	0.071
area 3x3	0.454	0.527	0.020	0.260	0.127	0.178
area 5x5	0.453	0.527	0.056	0.250	0.210	0.161
area 7x7	0.385	0.593	0.109	0.236	0.025	0.198
area 9x9	0.297	0.678	0.150	0.225	0.005	0.202
area 11x11	0.236	0.737	0.242	0.201	0.022	0.199
area 13x13	0.227	0.746	0.158	0.223	0.054	0.192
area 15x15	0.227	0.746	0.158	0.224	0.027	0.198
Significant R^2 values						
$R^2_{0.10}$	0.026		0.114		0.257	
$R^2_{0.05}$	0.043		0.181		0.386	

3.4 Comparison of the 8-week 2001 models to the 8-week 2000 models

Analyses in following paragraphs are based on the ARB 8-week average data. Figures 16 and 17 demonstrate the regression models derived separately from ARB 2000 and 2001 monitoring data. The data points and regression lines of the year 2000 are represented in blue color and open triangles, and those of the year 2001 are in red color and solid triangles.

In general, the 8-week regression models based on ARB 2000 data had higher R^2 values than the 2001 models over all use areas from 1x1 up to 15x15. The best regression of year 2000 was obtained in the 7x7 area with an R^2 value of 0.946, while the best 8-week regression model using ARB 2001 data was from the 3x3 use area with an R^2 value of 0.742. Except the 1x1 use area, all 2001 regression lines fell within the 95% confidence intervals of the 2000 regression lines. The regression models over the 1x1 area were dominated by a few points with high X and Y values (Figure 16). Therefore, regression models derived from ARB 2000 and 2001 monitoring data are not considered as statistically different.

Without exception, the 95% confidence intervals of regression coefficients of 2001 models overlapped to some degree with the counterparts of the 2000 models(Table 8). The intercepts of all regression lines were not significantly different from zero except on the 1x1 area. A positive air concentration was likely to be observed even if there were no applications over the 1x1 area. The 95% confidence intervals of slopes of all regression lines did not contain zero, indicating a positive relationship between air concentration and use over all areas.

Table 8: Comparison of parameter values of ARB 2001 and 2000 regression models

Area	a						b					
	2000			2001			2000			2001		
	Est *	CI_1 **	CI_2 ***	Est	CI_1	CI_2	Est	CI_1	CI_2	Est	CI_1	CI_2
1x1	0.83	0.25	1.42	1.24	0.32	2.17	0.00305	0.00228	0.00383	0.00161	0.00061	0.00262
3x3	0.51	-0.05	1.07	0.21	-0.75	1.17	0.00050	0.00039	0.00061	0.00065	0.00036	0.00094
5x5	0.26	-0.35	0.87	0.30	-0.84	1.43	0.00026	0.00020	0.00033	0.00023	0.00010	0.00036
7x7	0.12	-0.37	0.61	0.48	-0.69	1.64	0.00014	0.00012	0.00017	0.00011	0.00004	0.00018
9x9	0.04	-0.75	0.82	0.40	-0.86	1.67	0.00010	0.00007	0.00013	0.00008	0.00003	0.00013
11x11	0.12	-1.01	1.25	0.33	-1.00	1.66	0.00007	0.00004	0.00011	0.00006	0.00002	0.00010
13x13	0.09	-1.16	1.35	0.28	-1.11	1.68	0.00006	0.00003	0.00010	0.00006	0.00002	0.00009
15x15	0.12	-1.19	1.44	0.31	-1.15	1.76	0.00006	0.00003	0.00009	0.00005	0.00001	0.00009

* estimated parameter value (Est)

** lower threshold of the 95% confidence Interval of estimated parameter value (CI_1)

*** upper threshold of the 95% confidence Interval of estimated parameter value (CI_2)

3.5 Effects of Use Adjustments

As there were no substantial differences between regression models obtained separately from the ARB 2000 and 2001 monitoring data, the ARB data was consolidated and regressions were conducted based on the pooled date(Figures 18 and 19). The dependent variable was the mean of weekly average air concentrations over the 8-week period, and the independent variable was the mean of weekly methyl bromide use over that period. The best regression based on the pooled ARB data was on the 3x3 area, with the R^2 value of 0.837.

Regressions were also conducted based on the adjusted use by various factors, such as time, distance and wind direction(Table 9). All regressions remained to be significant at α=0.01 level. Time adjustments on use did not improve regression analysis of the ARB data. Distance adjustments as described in section 2.3.3 improved the regression. The best regression with the distance adjustment was obtained over the 5x5 area (R^2 = 0.904). It appeared that the distance adjustment worked better over the large area than over the small area. For example, with the distance adjustment, the R^2 value improved from 0.837 to 0.896 over the 3x3 use area, and from 0.602 to 0.805 over the 15x15 area. Wind direction adjustments failed to improve the regression in all areas. Adjustments by combined factors improved the regression over areas larger than 5x5, but not for areas smaller than 5x5.

Table 9: R^2 and EMS of regression models using pooled data of ARB 2000 and 2001 monitoring. The independent variable was the average of weekly methyl bromide use over 8-week period, unadjusted or adjusted

Area	Factors of Adjustment									
	None		Time		Distance		Wind Direction		All Factors	
	R^2	EMS	R^2	EMS	R^2	EMS	R^2	EMS	R^2	EMS
1x1	0.697	1.34	0.567	1.92	0.697	1.34	0.697	1.34	0.567	1.92
3x3	0.837	0.72	0.766	1.04	0.896	0.46	0.789	0.93	0.715	1.26
5x5	0.803	0.87	0.779	0.98	0.904	0.42	0.795	0.91	0.825	0.78
7x7	0.797	0.90	0.797	0.90	0.881	0.53	0.755	1.09	0.858	0.63
9x9	0.737	1.16	0.739	1.16	0.855	0.64	0.707	1.30	0.848	0.67
11x11	0.665	1.48	0.663	1.49	0.825	0.78	0.589	1.82	0.802	0.88
13x13	0.633	1.63	0.627	1.65	0.817	0.81	0.512	2.16	0.777	0.99
15x15	0.602	1.76	0.601	1.77	0.805	0.86	0.476	2.32	0.761	1.06

The number of samples: n = 22.
The critical R^2 value: $R^2_{0.01} = 0.242$.

4. Discussion

It was not accidental that the distance adjustment achieved the best result among the adjustments by individual factors. A remote application from the monitoring site imposed less impact on the monitoring results, and this effect wouldn't be changed by the time frame, be it one week, one month or two months. In general, the improvement by distance adjustment was better over large use areas than over small areas. There are at least two reasons. First, in this analysis the monitoring sites and pesticide applications were assumed to be at the center of sections. In reality they might be anywhere in a section. The error of distance between a sectional use location and a monitoring site location resulted from this assumption was relatively big in small use areas, and gradually diminished with larger use areas. Second, more use amount was discriminated by the distance factor at an increasing intensity when the use area increased. For example, from a 3x3 area to a 5x5 area, only 16 sections were added and f_d s: 1/2 . From a 13x13 area to a 15x15 area, however, 56 sections were added and f_d s: 1/6 . Therefore, the distance adjustment was more pronounced over large use areas.

The time adjustment, on the other hand, only makes sense in a short term, such as over a one-week period where applications on different days would have different chances of being captured by monitoring equipment. Taking the entire monitoring period, however, every single applications tend to be equally important to the average air concentration over the whole period, except those that occurred in the last several days of the period. Moreover, the application date in PUR reports did not reflect the exact date of application. The date in the PUR represented the last day of an application which may have taken place over several days. To that extent, the time adjustment on use is not reliable. Finally, the coefficients for time adjustment (Table 4) were based on the average emission fractions of four application methods and might be off from the true curve of soil emission fraction.

The wind direction adjustment would seem like a reasonable procedure. Nevertheless, like the time adjustment, it depended on knowing the exact time of an application. Multiple applications might be reported on a single application date in the PUR report, leading the time window ambiguous for the wind direction adjustment. Another reason was that wind directions at one weather station were used for several monitoring sites in the area, which might cause problems in a terrain that was not homogeneous and flat.

The use adjustment by time, distance and wind direction was a semi-empirical approach to explain the monitored air concentrations. It improved the regression in some cases, but failed in others. The procedure of use adjustments was complicated and sometimes the data required for these use adjustments was not available. Distance adjustment was easier. It used a simple algorithm to calculate distance and its coefficient, and it only depended on the PUR data. When it was successful, the model provided a more accurate prediction of the ambient air concentration from use data.

Although using adjusted use amount improved the regression model in some cases, it won't offer any help for regulatory requirements. For regulatory requirements, we still need to rely on regression models with the unadjusted use as the independent variable.

2001 regression models are not as good as 2000 models when using unadjusted use as the independent variable. The possible reasons that contribute to the poor correlation between use and air concentrations include:

(1) variation of weather conditions
(2) irregular concentration week and use week
(3) errors in pesticide use reports, such as repeated records and incorrect units
(4) intrinsic limitation of PUR reports, such as poor temporal and spatial resolutions of pesticide applications
(5) incomplete and inaccurate PUR reports
(6) errors in air concentration measurements

Factor (2) was unique to the year 2001 data.

5. Summary

The ARB 2001 air monitoring data of methyl bromide are in agreement with the empirical relationships established using the ARB 2000 monitoring data. Regression models derived separately from 2000 data and 2001 data of ARB air monitoring were not substantially different. The AMBI air monitoring data did not support a linear relationship between the use and air concentration over an eight-week period. Improvements were achieved for some regression models with use adjusted for distance or the combined factors of time, distance and wind direction. The spatial and temporal resolutions of PUR reports and the availability of wind direction data put some constraints on use adjustments by individual factors, such as distance, time and wind direction.

Acknowledgment

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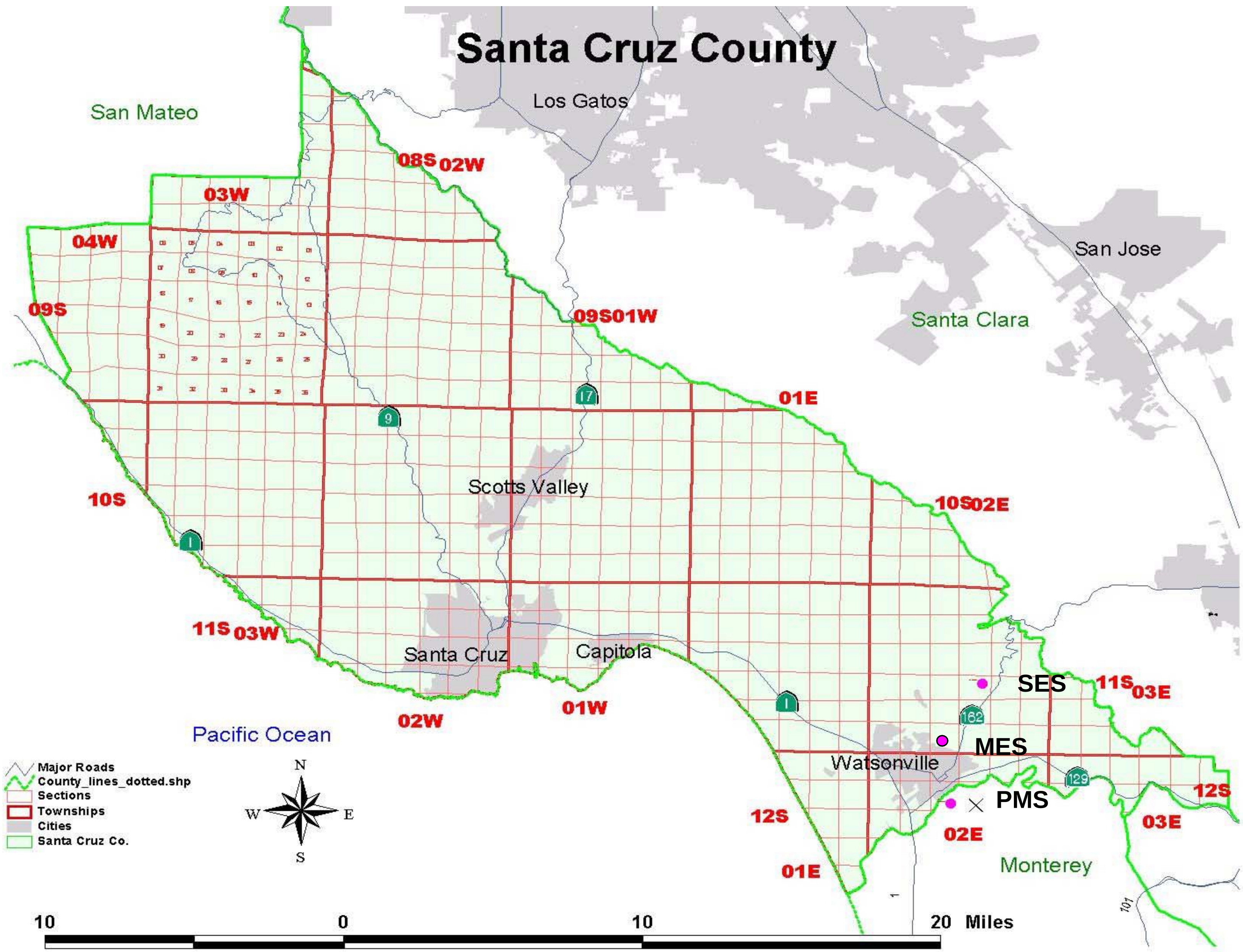


Figure 1 ARB 2001 monitoring sites in Santa Cruz County

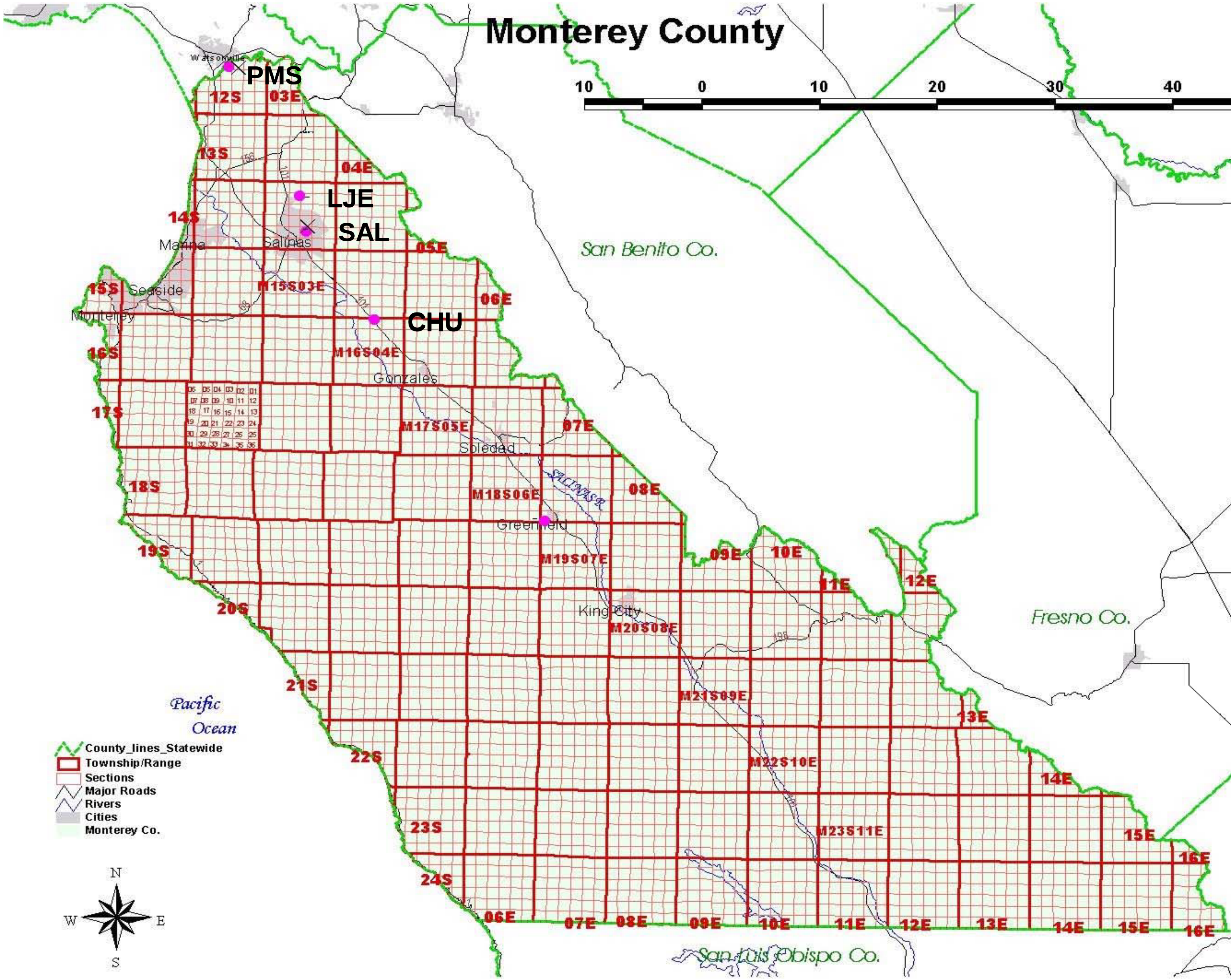


Figure 2 ARB 2001 monitoring sites in Monterey County

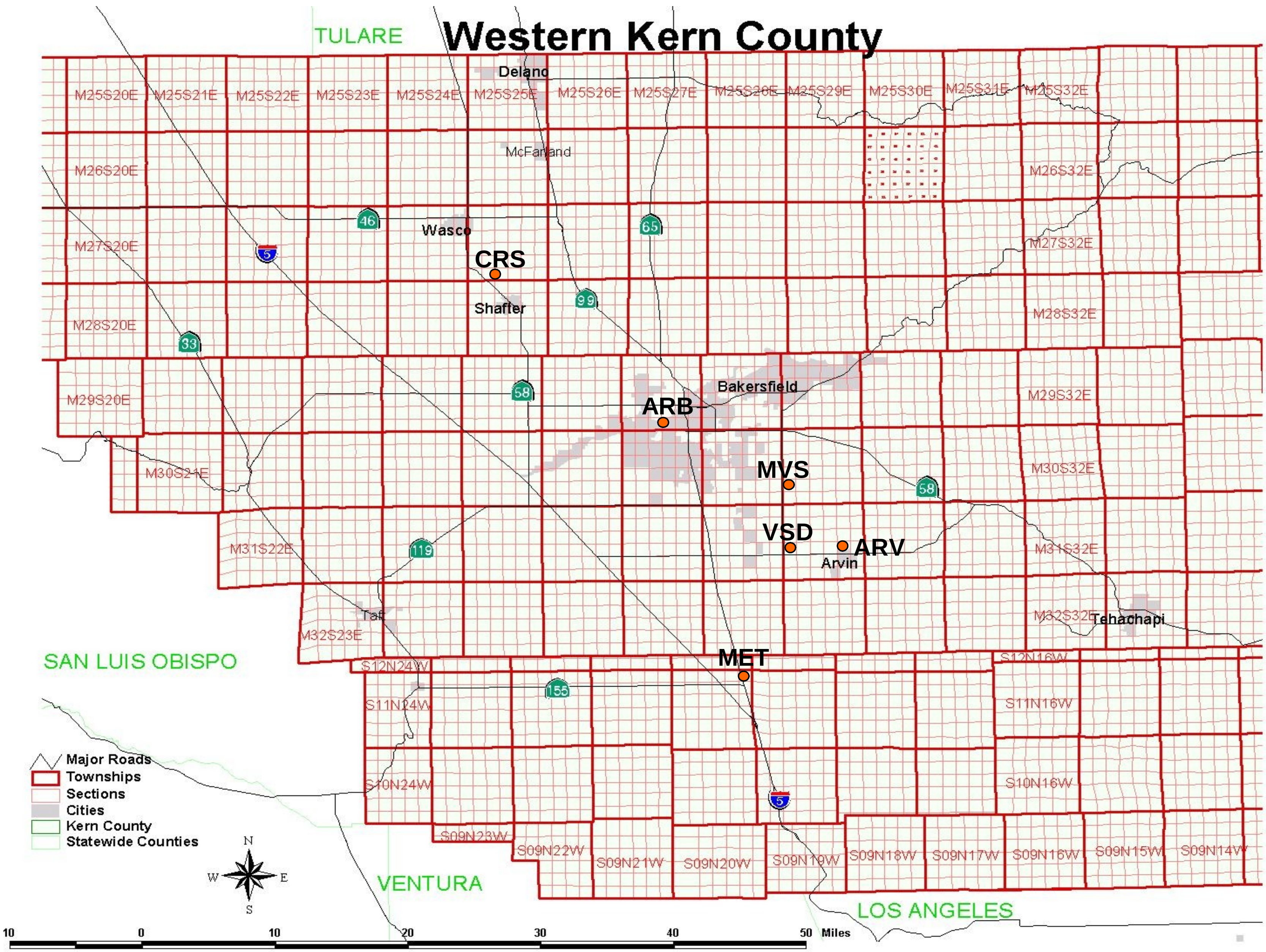


Figure 3 ARB 2001 monitoring sites in Kern County

Santa Maria

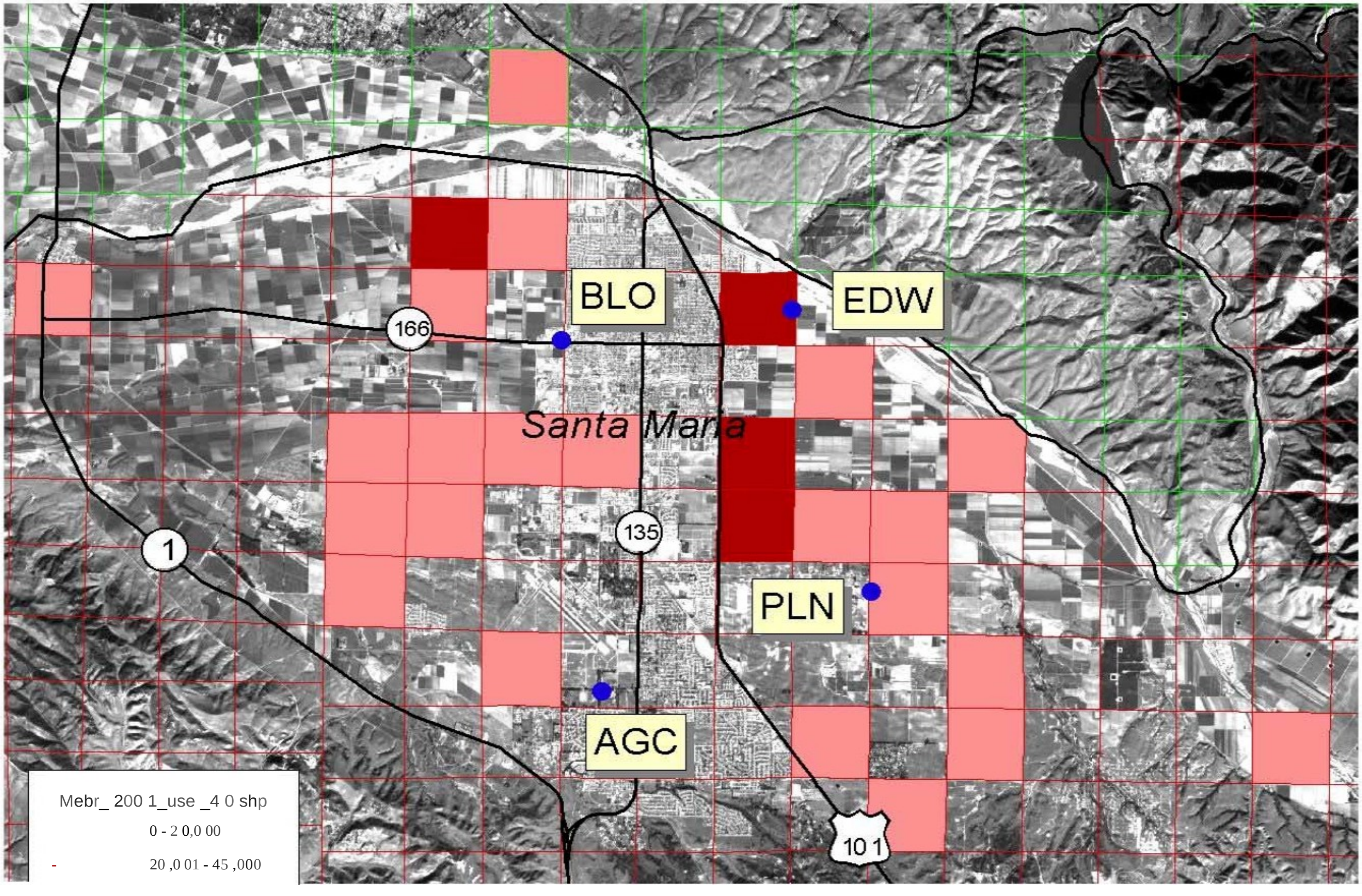
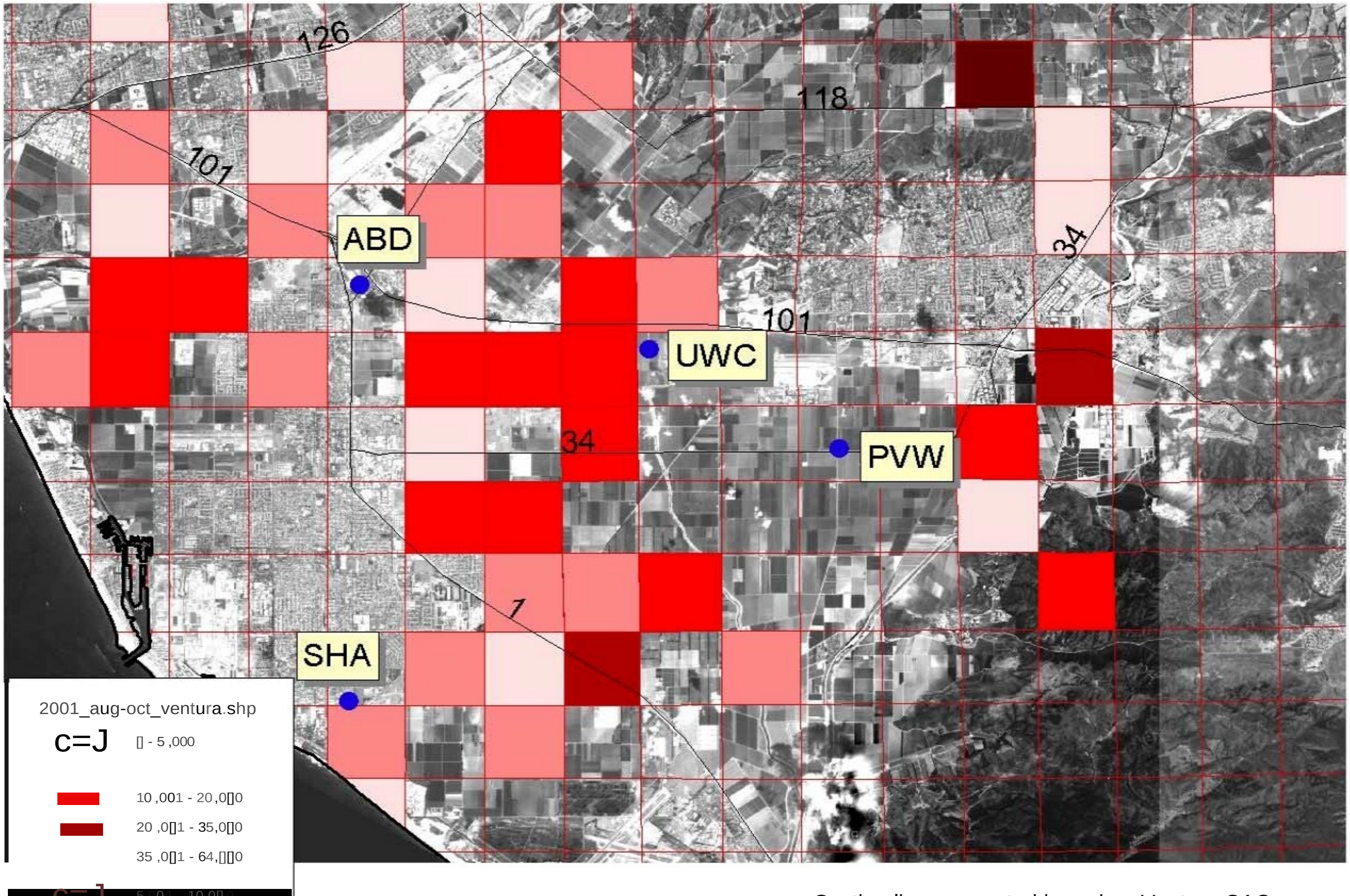


Figure 4 AMBI 2001 monitoring sites and methyl bromide use distributions in Santa Barbara County

Ventura County



Section lines corrected based on Ventura CAC map.

Figure 5 AMBI 2001 monitoring sites and methyl bromide use distributions in Ventura County

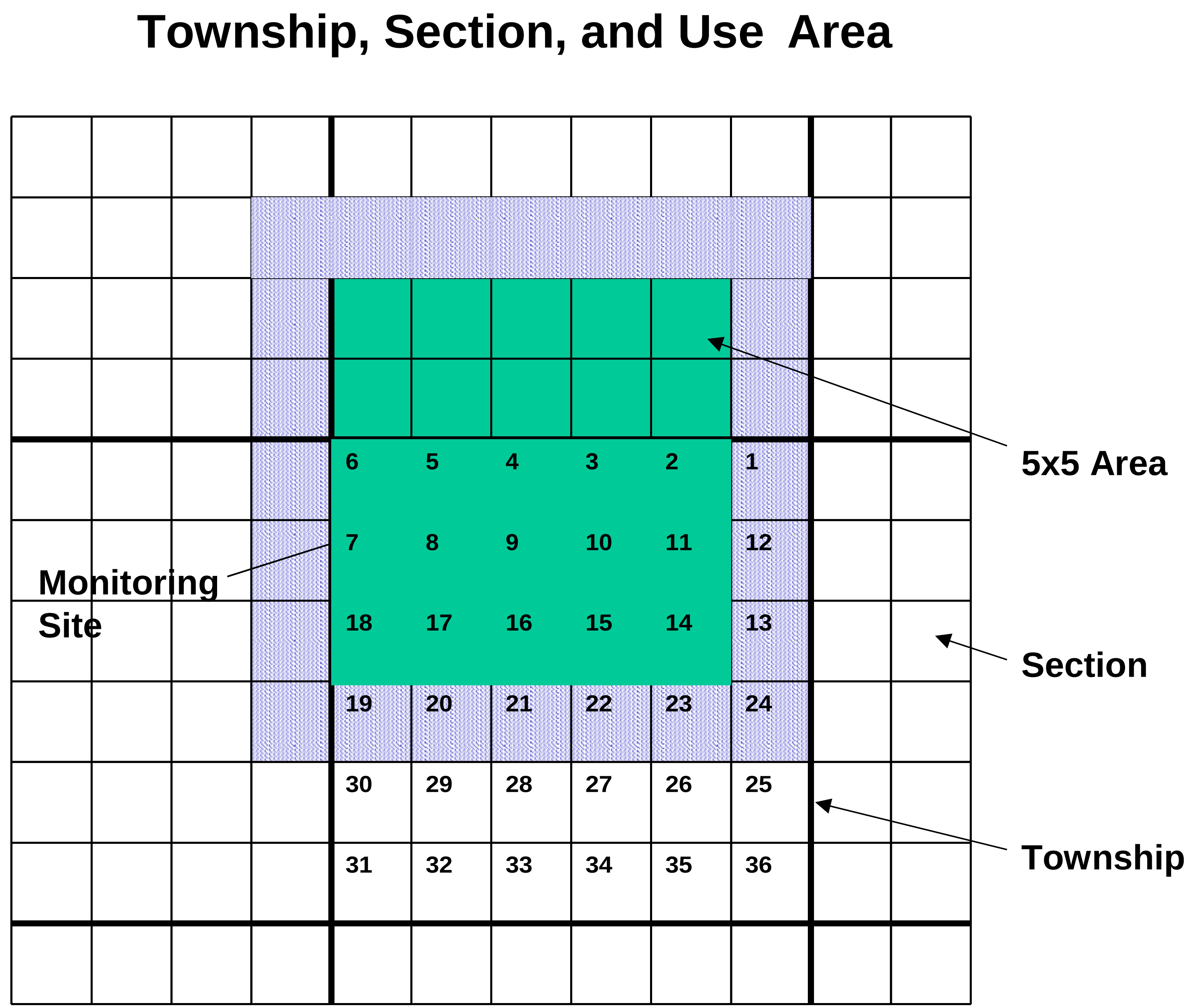
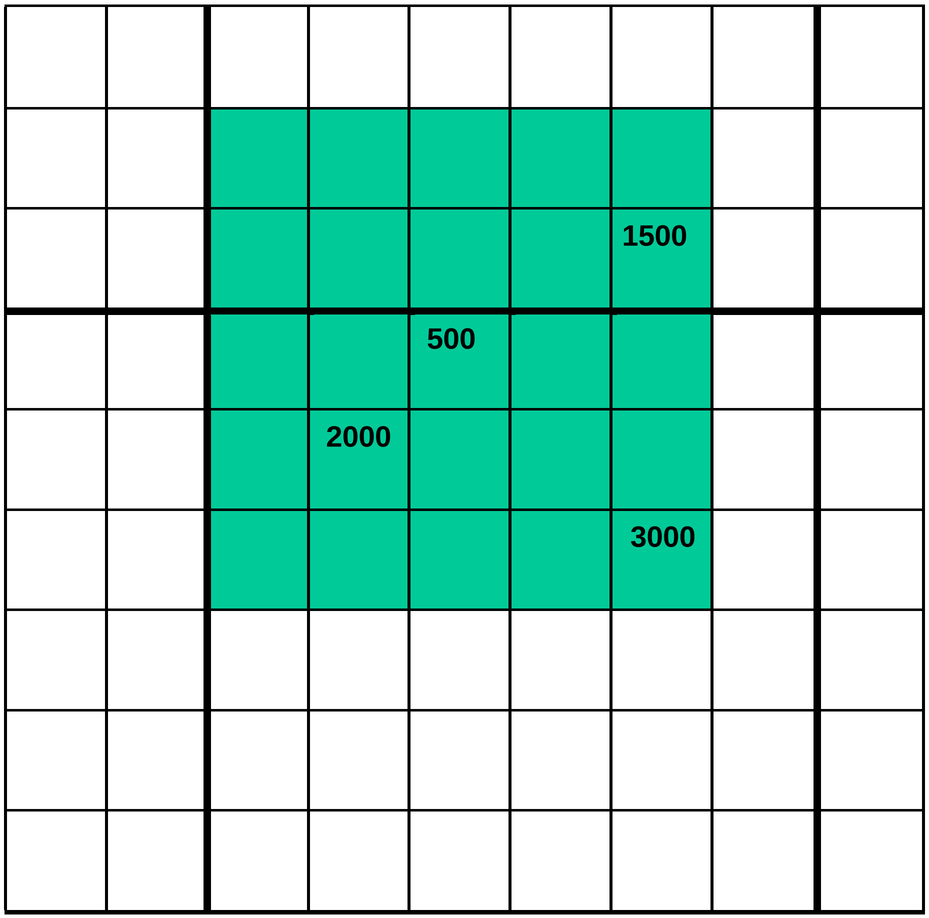


Figure 6 Illustration of township, section and the 5x5 use area

Unadjusted Use



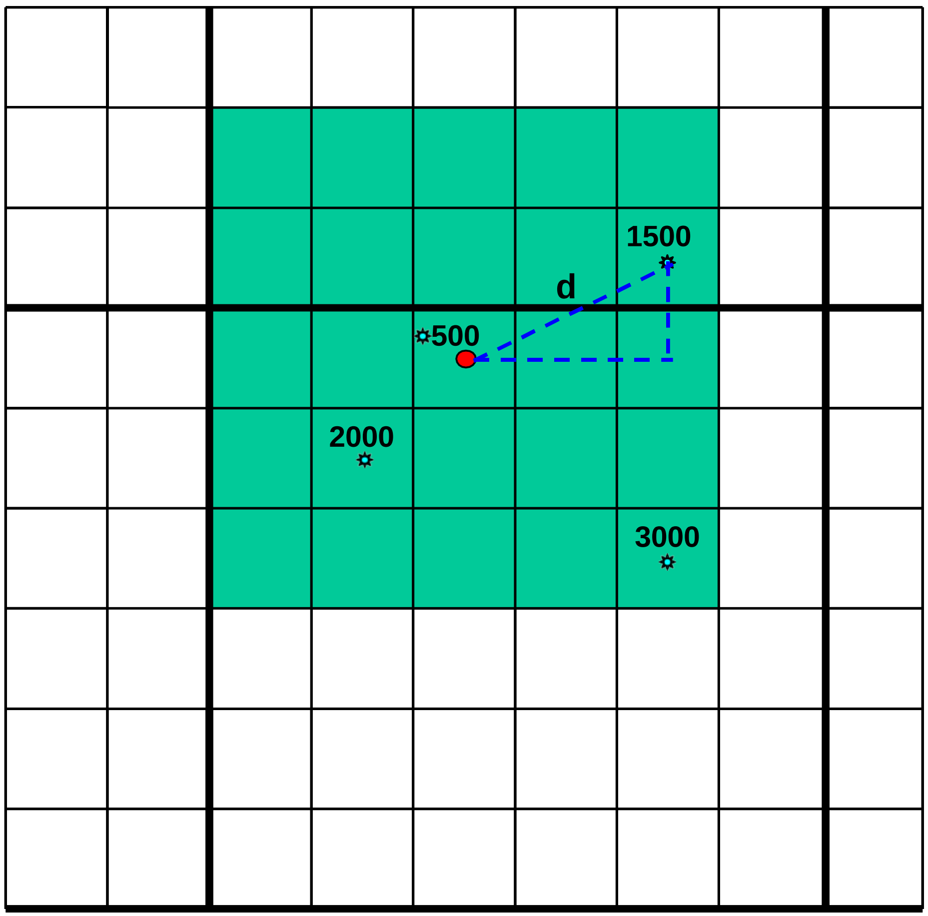
Unadjusted Use

= 500 + 1500 + 2000 + 3000

= 7000

Figure 7 Example: calculation of unadjusted use in the 5x5 area. The total use is the sum of use pounds in each section.

Distance Adjusted Use



$f_d = 1/d$

d: distance

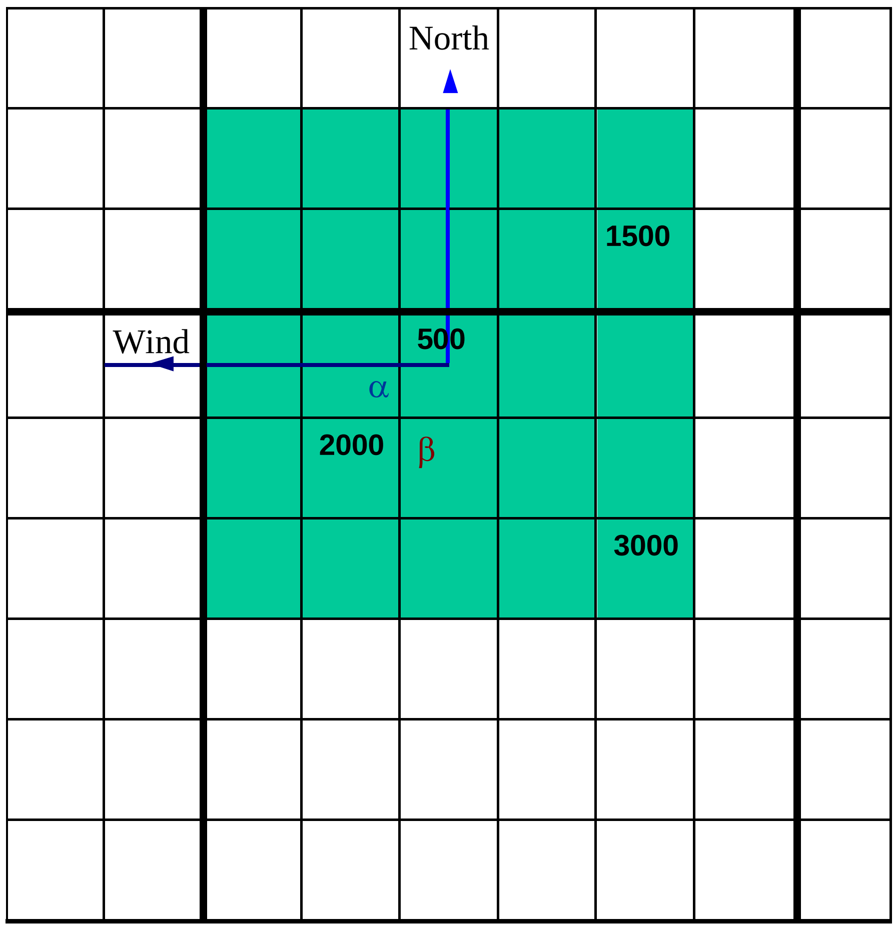
Distance Adjusted Use

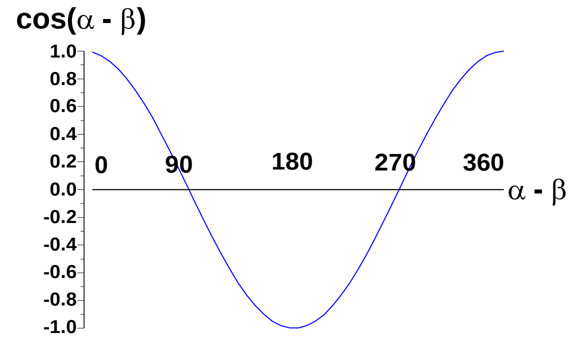
$= 500/0.5 + 1500/\sqrt{5} + 2000/\sqrt{2} + 3000/\sqrt{8}$

$= 4146$

Figure 8 Example: calculation of distance adjusted use in the 5x5 area. Distance (d) is calculated from the center of the monitoring section to the center of an application section, and the distance coefficient f_d is defined as 1/d. d = 0.5 if the application is in the monitoring section

Wind Direction Adjusted Use





$f_w = \max\{0, \cos(\alpha - \beta)\}$

α : wind direction

β: orientation of use section

Wind Direction Adjusted Use

= 500x0.5 + 1500x0 + 3000x0

+ 2000xcos(270°-225°)

= 1664

Figure 9 Example: calculation of wind direction adjusted use in the 5x5 area. α and β represent wind direction and orientation of a use section respectively, and the wind direction coefficient f_w is defined as max{ 0, cos(α - β)}. In the diagram wind direction is from west, and only upwind applications contribute to the adjusted use. In case an application is in the monitoring section, $f_w = 1$.

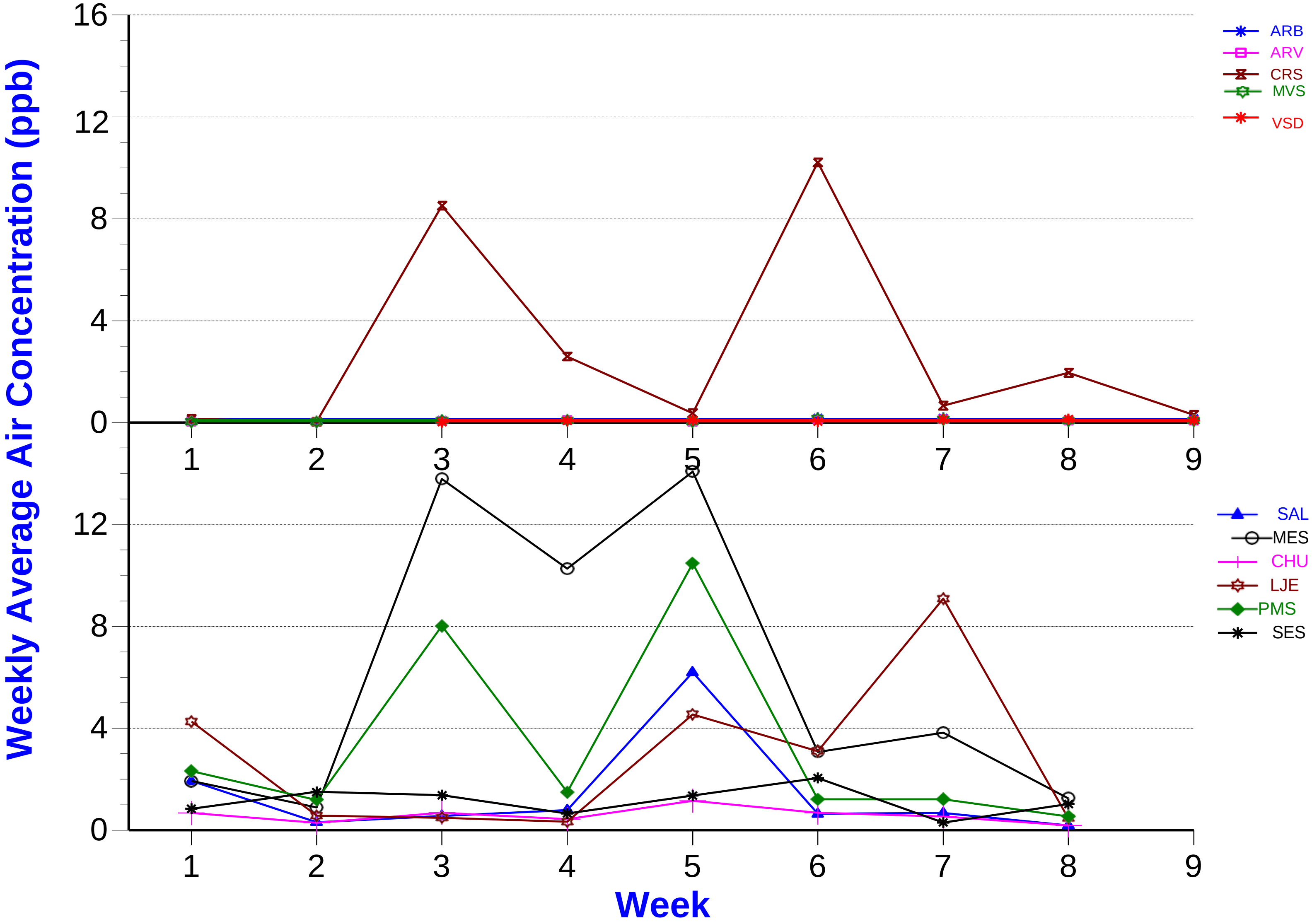


Figure 10 Variations of weekly average air concentrations at each of the ARB 2001 monitoring sites

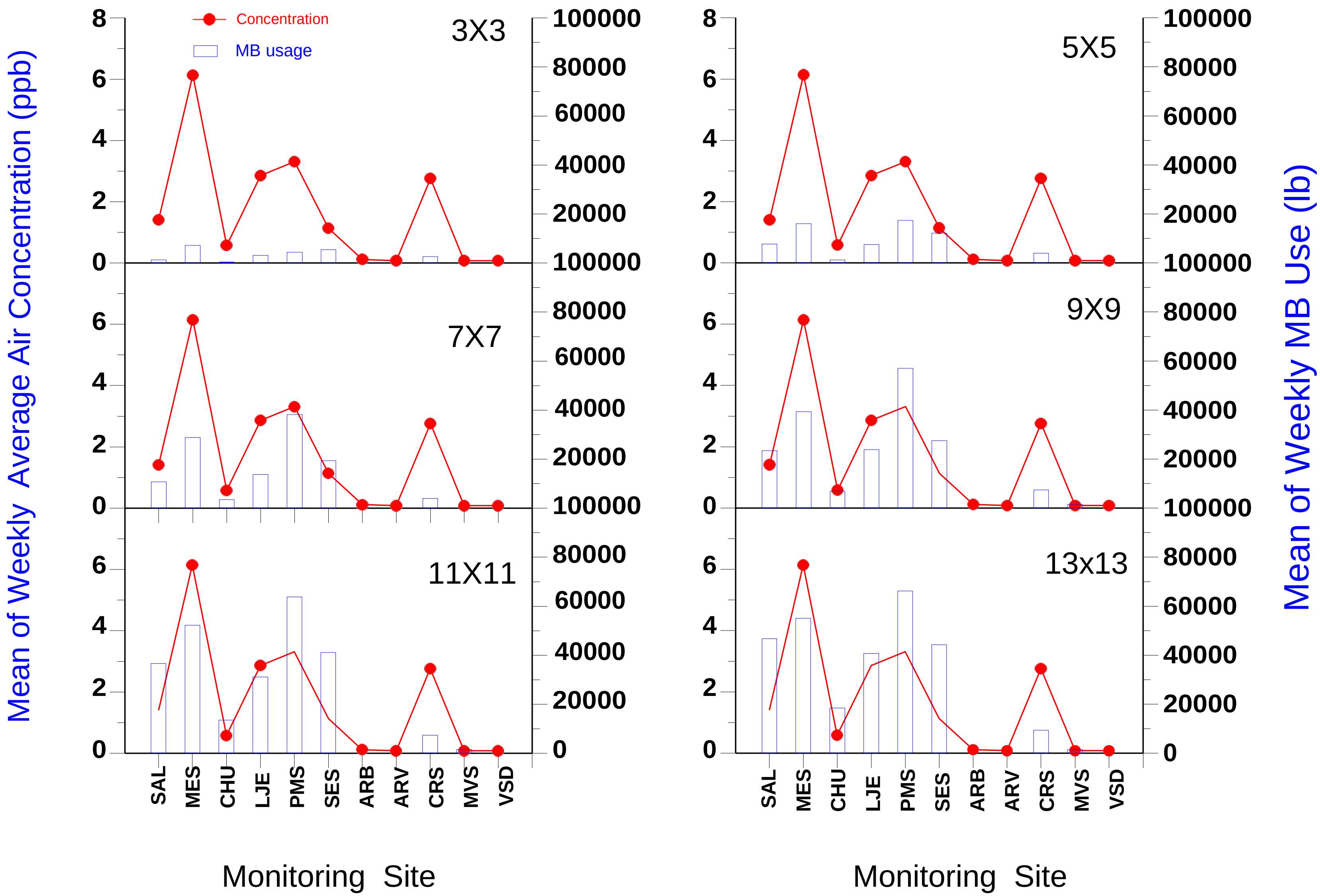


Figure 11 Weekly air concentration vs weekly methyl bromide use in various areas at all ARB 2001 monitoring sites

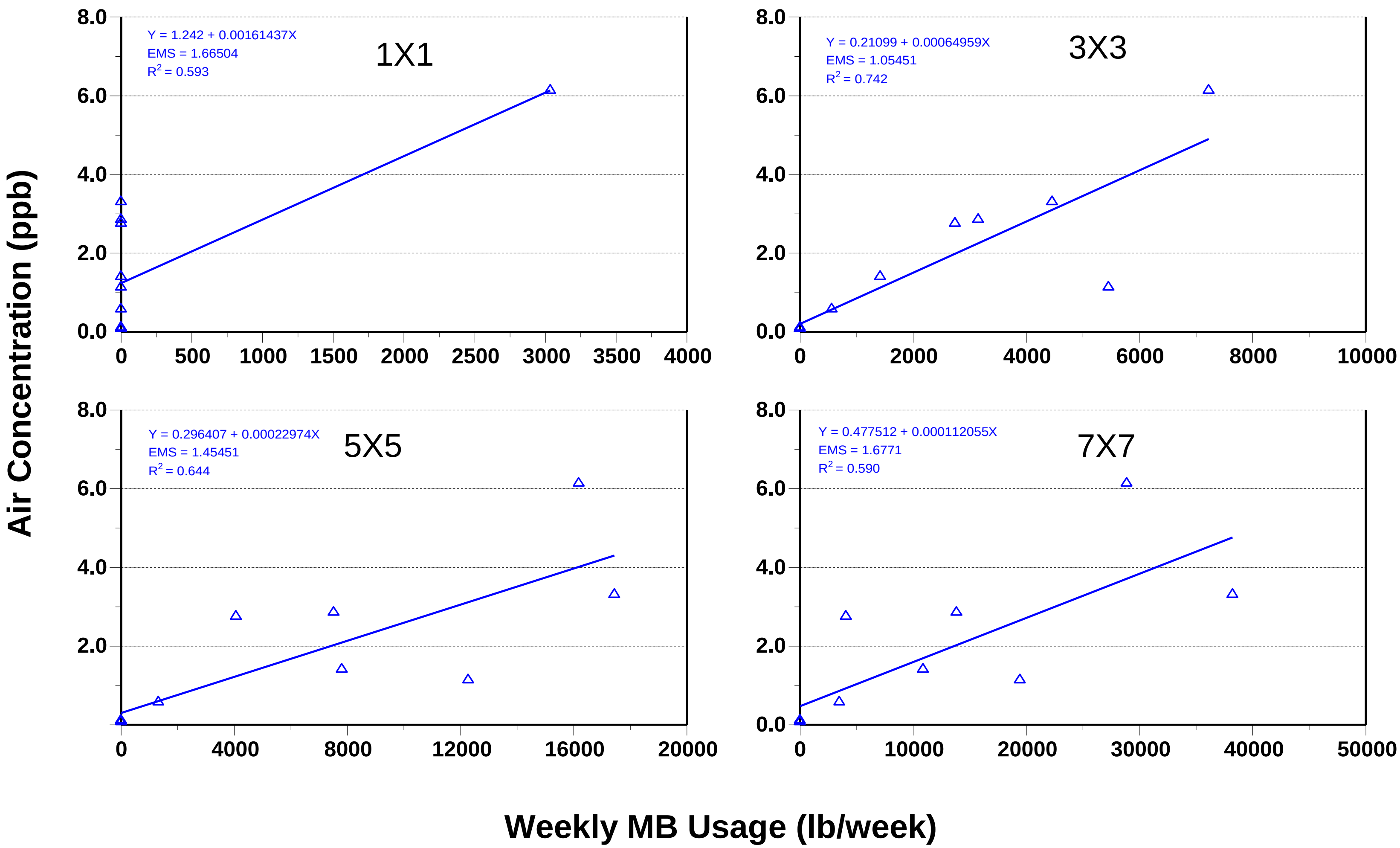


Figure 12 Regression between the mean of weekly average air concentrations and the mean of weekly uses over 8 week period based on the ARB 2001 monitoring data

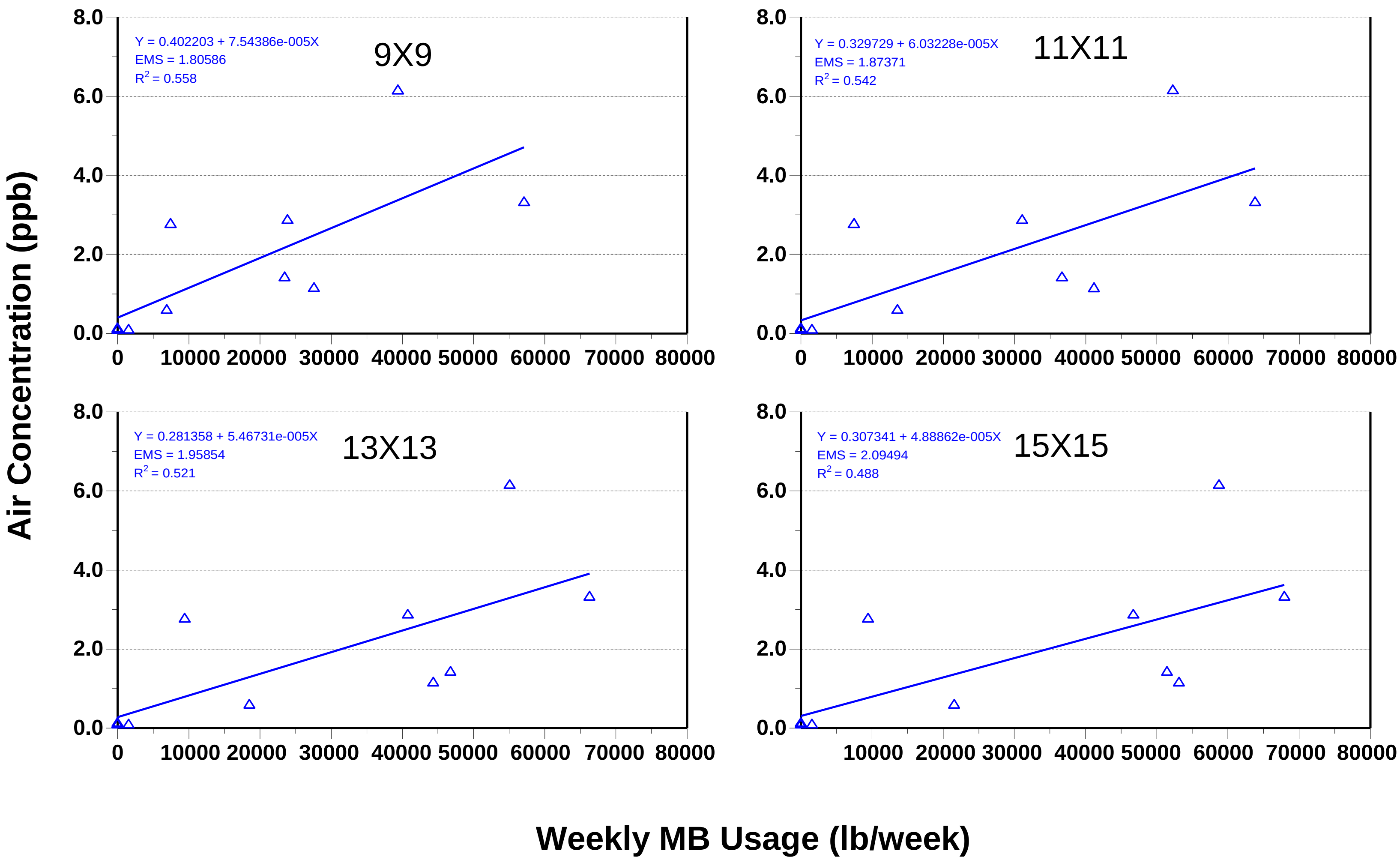


Figure 13 Regression between the mean of weekly average air concentrations and the mean of weekly uses over 8 week period based on the ARB 2001 monitoring data

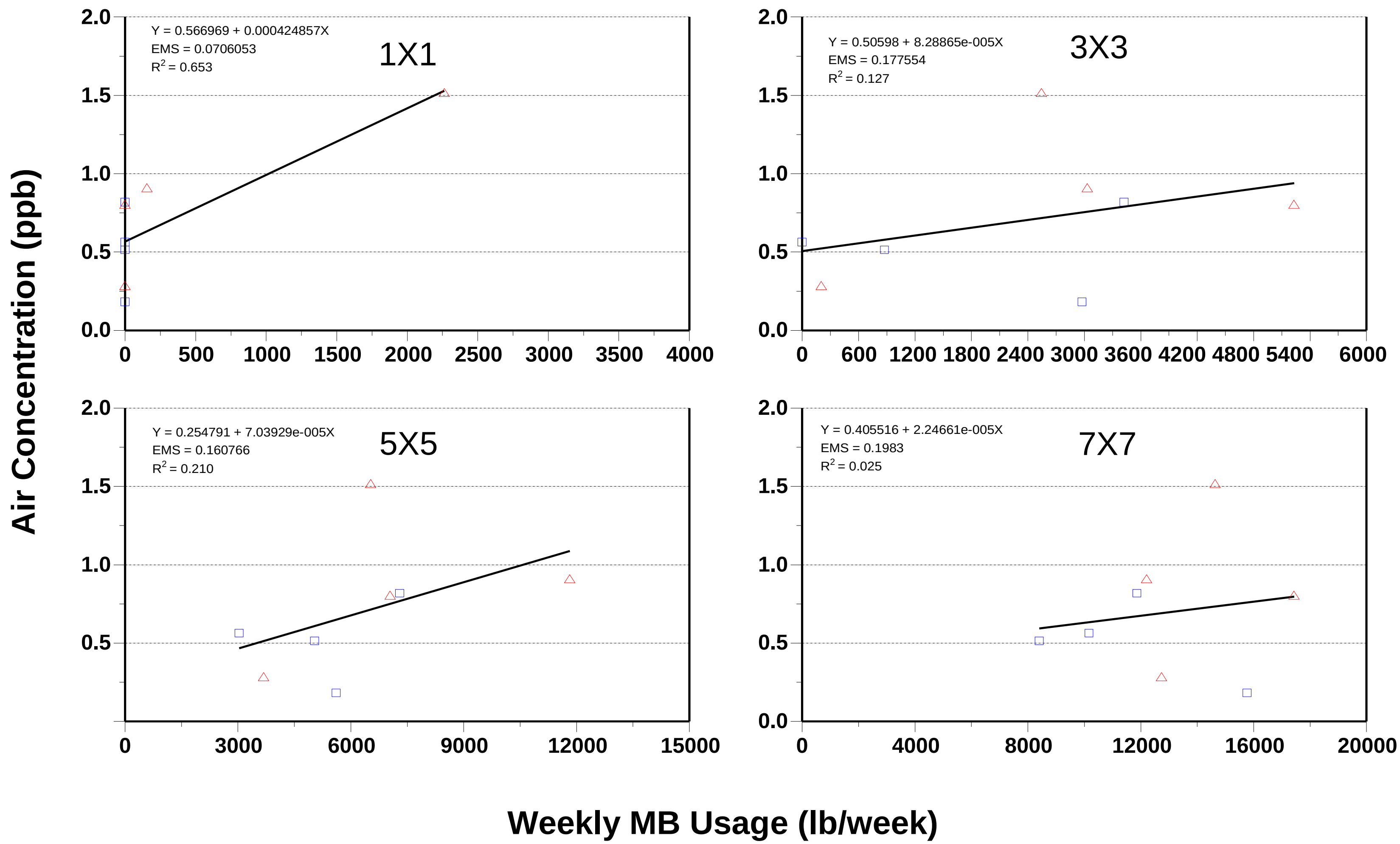


Figure 14 Regression between the mean of weekly average air concentrations and the mean of weekly uses over 8 week period based on the AMBI 2001 monitoring data. Triangles and squares represent monitoring data in Santa Maria of Santa Barbara County and Oxnard/Camarillo of Ventura County respectively.

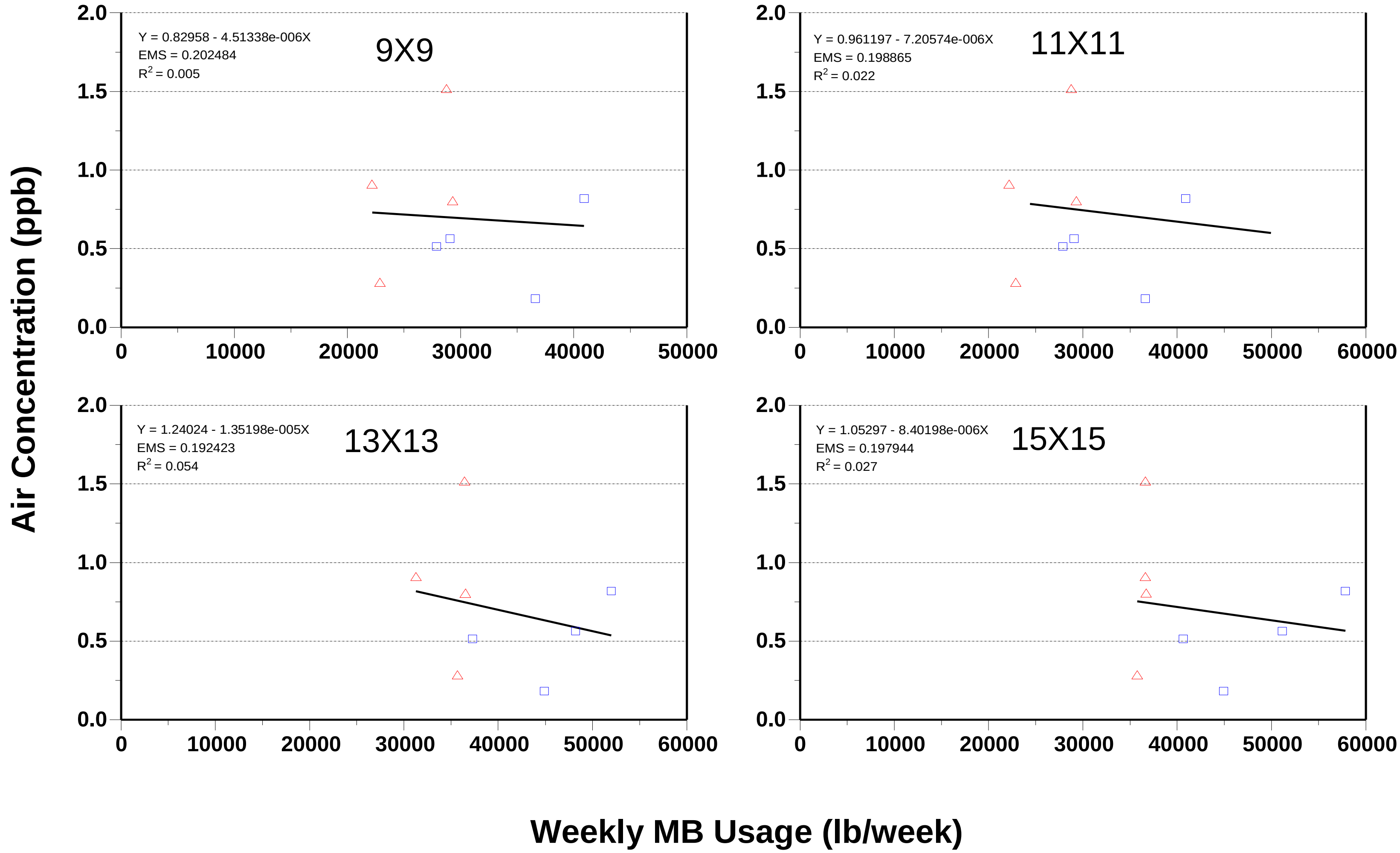


Figure 15 Regression between the mean of weekly average air concentrations and the mean of weekly uses over 8 week period based on the AMBI 2001 monitoring data. Triangles and squares represent monitoring data in Santa Maria of Santa Barbara County and Oxnard/Camarillo of Ventura County respectively.

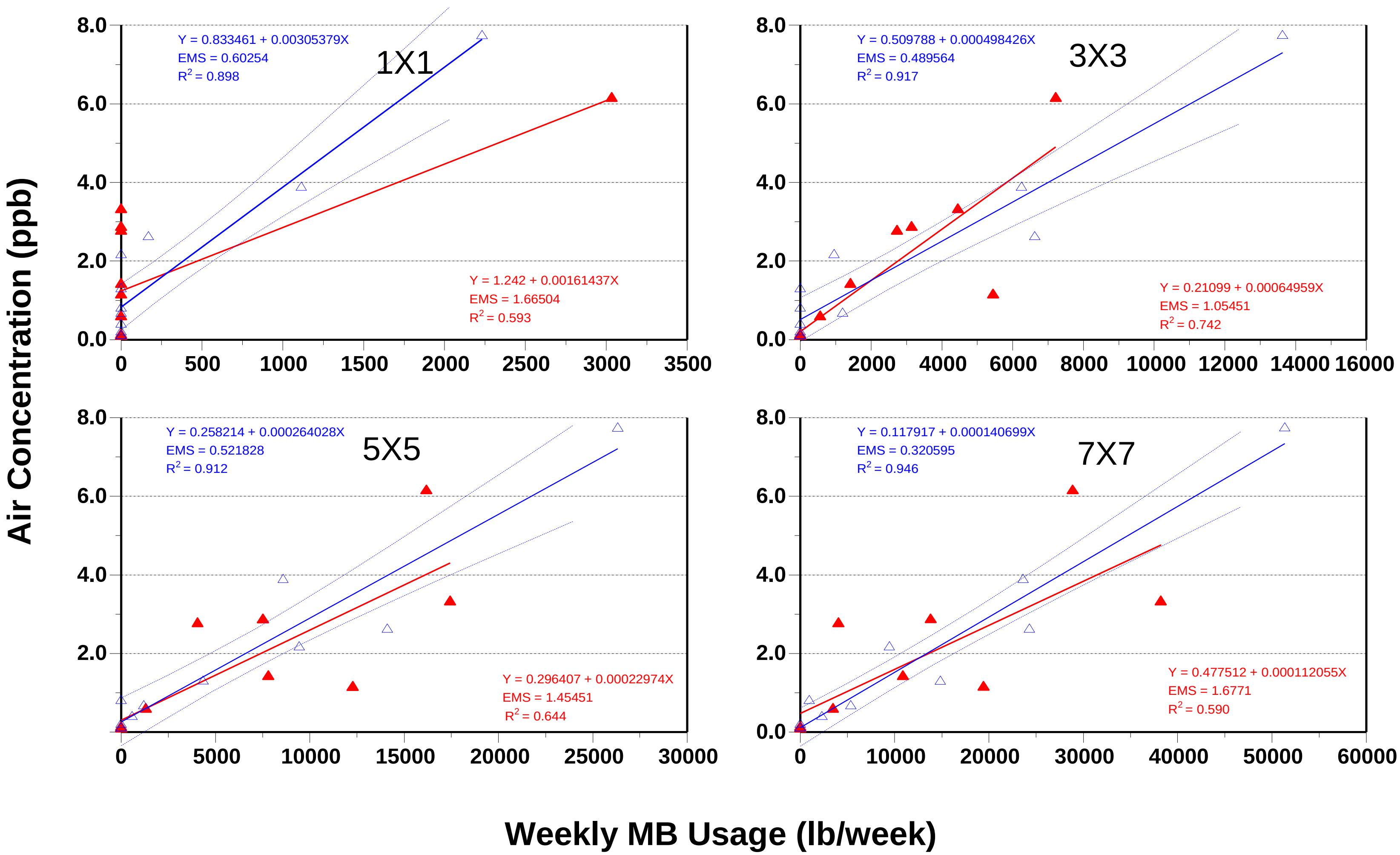


Figure 16 Comparison between ARB 2000 regression models and ARB 2001 regression models based on 8 week average data. Open and solid triangles represent 2000 and 2001 monitoring data respectively. Dash lines define the 95% confidence intervals of the ARB 2000 regression models.

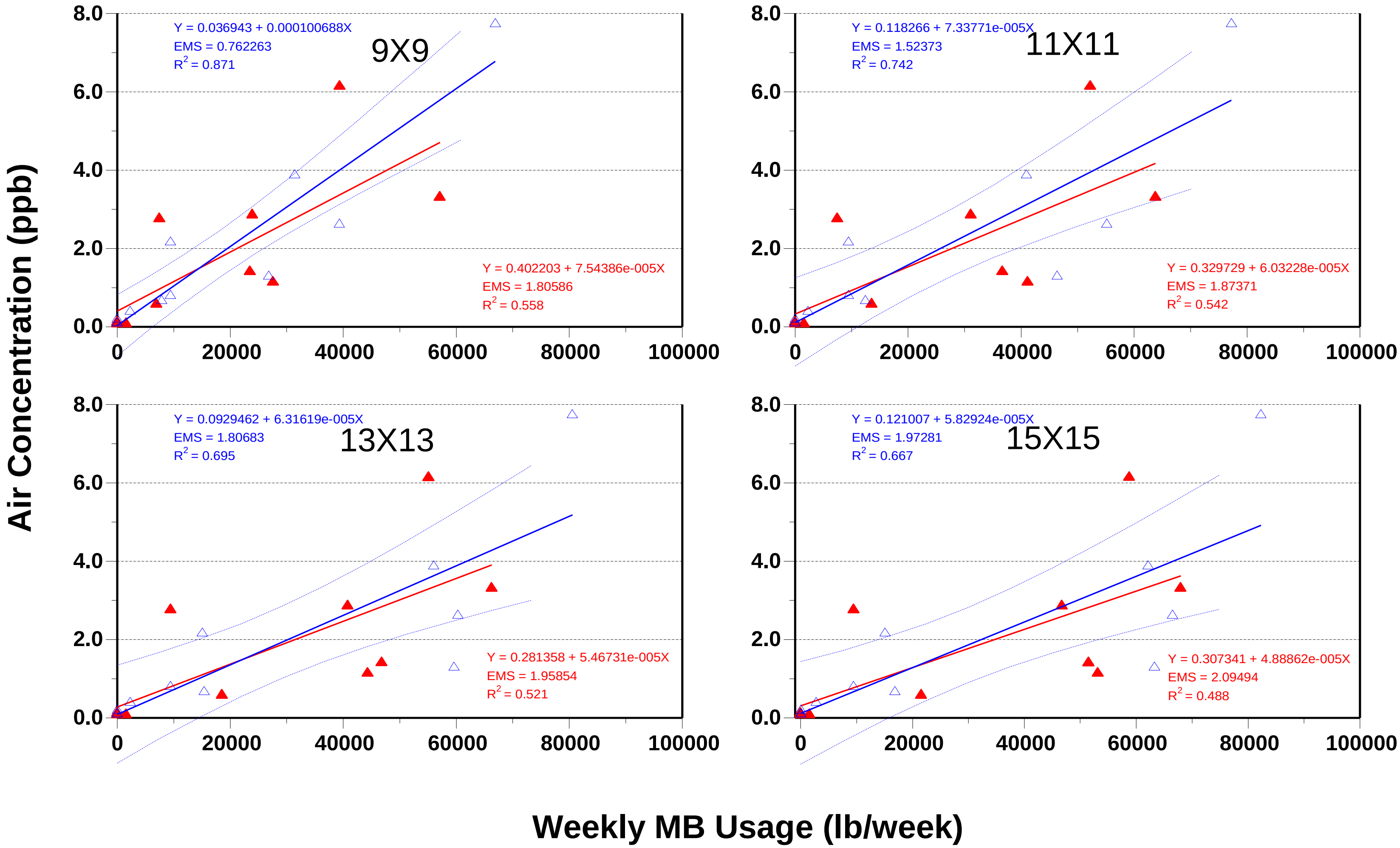


Figure 17 Comparison between ARB 2000 regression models and ARB 2001 regression models based on 8 week average data. Open and solid triangles represent 2000 and 2001 monitoring data respectively. Dash lines define the 95% confidence intervals of the ARB 2000 regression models.

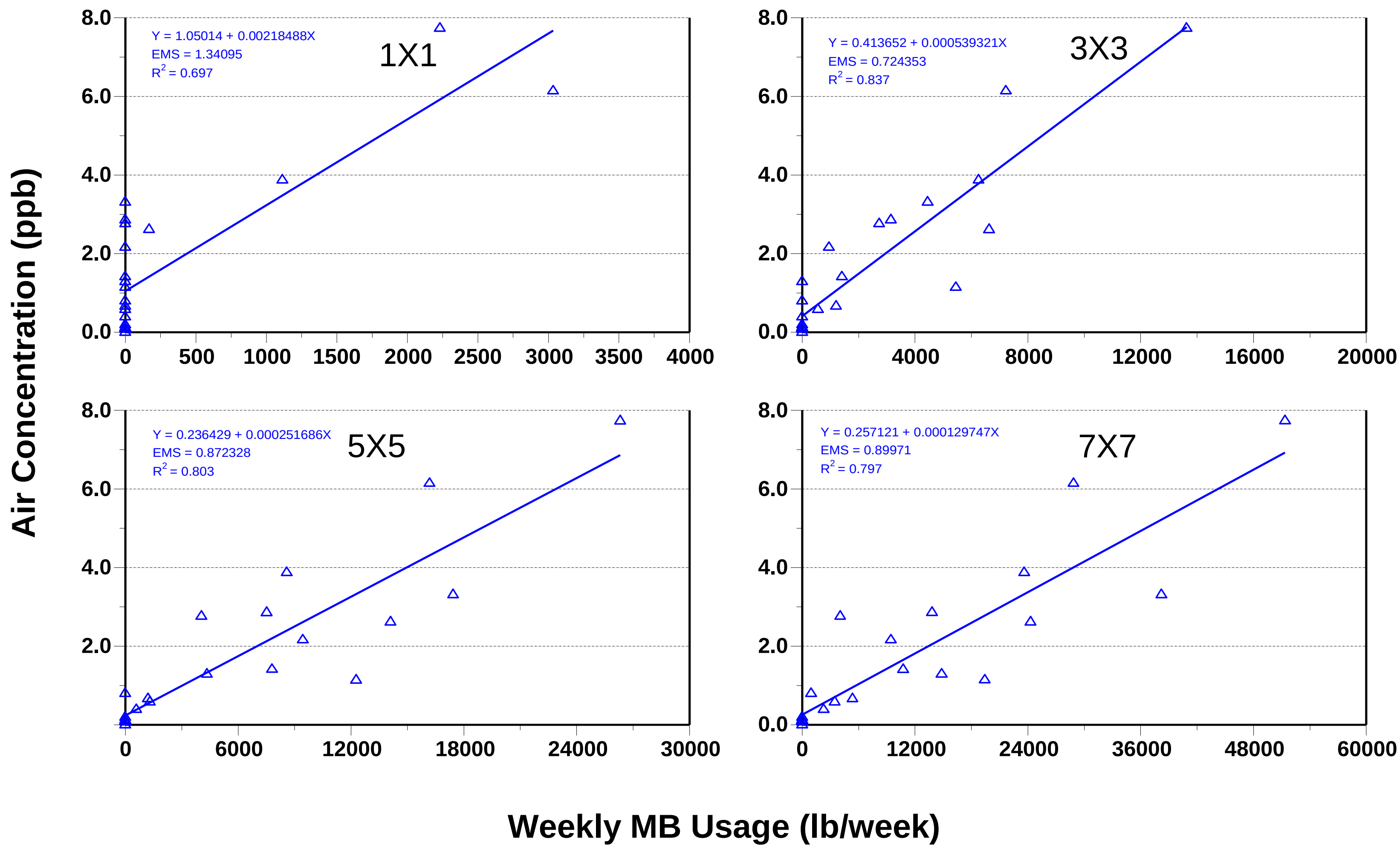


Figure 18 Regression models based on the pooled data of ARB 2000 and 2001 monitoring. The air concentration(Y) and the use(X) are means of weekly averages over the 8-week period.

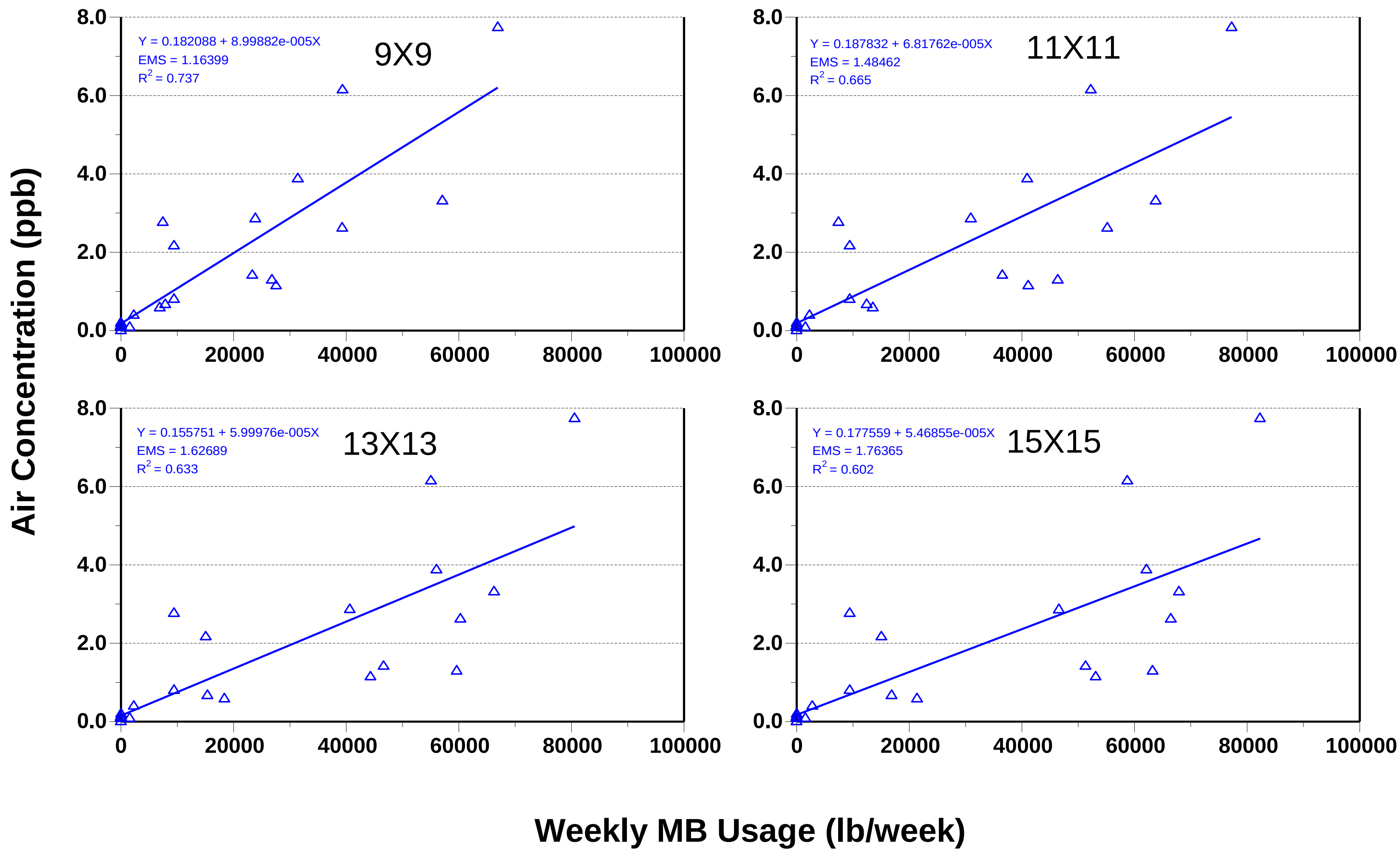


Figure 19 Regression models based on the pooled data of ARB 2000 and 2001 monitoring. The air concentration(Y) and the use(X) are means of weekly averages over the 8-week period.