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SUBJECT: SUMMARY OF 1990-2000 VOC EMISSIONS INVENTORY DATA, VERSION 01.12

I. OVERVIEW

This memorandum provides a general summary of the December 2001 version of the Department of Pesticide Regulation's (DPR's) volatile organic compound (VOC) emissions inventory. The VOC inventory is an estimate of potential reactive organic gas (ROG) emissions from agricultural and commercial structural pesticide applications in California (DPR, 1996). These emissions are of concern due to their potential to contribute to tropospheric ozone formation. The summary here includes time-series data of annual emissions in each of the five ozone non-attainment areas (NAA) and a more detailed overview of the most recent 2000 data, including principal active ingredients of VOC contributing products, and distribution of emissions by formulation classes in the various NAAs.

II. BACKGROUND

The calculation procedures for the 01.12 version of the inventory were significantly different than those used in the past. The purpose of revising the VOC calculation procedures was to improve the accuracy and consistency of the resultant potential VOC emission estimates. There were three general types of changes to input data or calculation procedure relative to past inventories:

A. Increase the types of pesticide applications considered as "agricultural pesticide use" in calculating potential VOC emissions from pesticide use report (PUR) data.

   This change was implemented to improve consistency between the term "agricultural use" as used in the VOC inventory and the legal definition of "agricultural use" in California's Food and Agriculture Code, section 11408. The effect of this change on statewide-calculated emissions was minor, corresponding to about a percent or less in most years.

B. Improve the screening process to remove adjuvant, technical, or inorganic products. Numerous products have been included in past inventory calculations that are not VOC sources. Conversely, other products that should have been included in the past were inadvertently omitted. The PUR data screening process has been improved to address these problems. The effect of this change on total 1990-2000 calculated emissions was moderate, corresponding to less than a few percent to up to 5-10 percent depending on year and geographic location.

C. Derive more realistic emission potentials (EP) for (a) several high-use pesticide products and (b) the default values assigned to newer products or others without EP data (Spurlock, 2002a).

For high-use products (e.g., metam-sodium products, sulfur products, sodium chlorate products), updated emission potentials were derived based on basic chemical principles and using confidential statements of formula supplied by pesticide registrants.

Default EPs for different formulation classes are now taken to be the median of EPs determined by thermogravimetric analysis (TGA; DPR, 1996) in the formulation class. This is as opposed to the historical practice of defining default EPs as equal to the highest TGA-based EP in each formulation class. The effect of the new EPs on inventory results is substantial, and the new inventory yields much more realistic estimates of pesticidal VOC emissions than previous inventories. In general, the current estimates based on more realistic EPs are 10-40 percent lower statewide than previous estimates.

These and other changes will be detailed in a report that summarizes and documents the VOC inventory calculation procedures (Spurlock, 2002b).

III. *VERSION 01.12* VOC INVENTORY RESULTS

A. NAA 1, Sacramento Metropolitan Area

Potential VOC emissions in the Sacramento Area ranged from about 1.6 million to 2.4 million pounds/year over the last decade, averaging approximately 5 percent of annual statewide emissions in any one year (Figure 1). The 2000 NAA 1 emissions amounted to 87% of 1990 NAA 1 baseline emissions, corresponding to a 13 percent reduction as compared to the 2005 target of a 20 percent reduction from the 1990 baseline. However, there is no consistent emission trend in NAA 1, and in 1996, 1997, and 1999 emissions actually exceeded the 1990 baseline. Approximately 93 percent of potential 2000 emissions in NAA 1 were attributable to agricultural uses with the remaining 7 percent from commercial structural pesticide applications.

The peak months for 2000 NAA 1 emissions were March-June (Figure 2), partially driven by the rice herbicide application period of April-June. Products containing the two herbicides, molinate and thiobencarb, accounted for 21 percent of the total potential 2000 emissions in

NAA 1, and 30 percent during the peak months of March-June (Table 1). However, similar to most other areas of California the largest contributors to VOC emissions were fumigants; together fumigant products containing metam-sodium, methyl bromide, and 1,3-dichloropropene contributed to approximately 29 percent of 2000 NAA 1 potential emissions. These three fumigants also accounted for 29 percent of emissions during the peak months of March-June.

Among formulation classes, emulsifiable concentrates (EC) and granular/flake formulations contributed more than 70 percent to total emissions in NAA 1 (Table 2). While a single metam-sodium product contributed almost a third of the EC emissions, the remainder of major pesticides in the EC group consisted of insecticides and preemergent herbicides. The granular/flake formulation represents the dominance of rice pesticides in NAA 1 agriculture; more than 85 percent of granular/flake emissions resulted from the granular formulations of the rice herbicides molinate and thiobencarb.

TABLE 1. Top ten “primary active ingredients” contributing to 2000 potential VOC emissions in NAA 1, the Sacramento Metropolitan Area, listed in order of total emissions of products containing the primary active ingredient. Here the primary active ingredient is defined as the pesticidal active ingredient present at the highest percentage in a product. This approach prevents “double-counting” of emissions from products containing two active ingredients.

Primary AI	**Total product emissions (lbs)**	**% Total product emissions**
METAM-SODIUM	323853	19.4%
MOLINATE	305780	18.3%
METHYL BROMIDE	110060	6.6%
CHLORPYRIFOS	79669	4.8%
TRIFLURALIN	61615	3.7%
PEBULATE	49367	3.0%
1,3-DICHLOROPROPENE	49009	2.9%
THIOBENCARB	48249	2.9%
OXYFLUORFEN	39643	2.4%
GLYPHOSATE, ISOPROPYLAMINE SALT	35363	2.1%

TABLE 2. 2000 potential emissions in NAA 1 by formulation category.

Formulation code	**Formulation**	**Total emissions (lbs)**	**% Total emissions**
A0	DUST/POWDER	20411	1.2%
B0	EMULSIFIABLE CONCENTRATE	802426	48.0%
C0	FLOWABLE CONCENTRATE	29367	1.8%
E0	GRANULAR/FLAKE	394220	23.6%
H0	OIL	5647	0.3%
J0	PELLET/TABLET/CAKE	11396	0.7%
L0	PRESSURIZED GAS	67562	4.0%
M0	PRESSURIZED LIQUID	81586	4.9%
N0	SOLUBLE POWDER	984	0.1%
O0	READY-TO-USE-SOLUTIONS	122824	7.4%
P0	WETTABLE POWDER	12602	0.8%
Q0	AQUEOUS SUSPENSION	2941	0.2%
R0	DRY FLOWABLE	9753	0.6%
S0	AQUEOUS CONCENTRATE	109063	6.5%

B. NAA 2, San Joaquin Valley

Potential VOC emissions in NAA 2 have shown a significant downward trend since 1995 (Figure 1, $p<0.01$), although in 2000 the estimated emission of 18.8 million lbs/year is still about 2 million pounds greater than the 2005 target goal of 16.6 million pounds. Approximately 98 percent of potential 2000 emissions in NAA 2 were attributable to agricultural uses with the remaining 2 percent from commercial structural pesticide applications.

Emissions in NAA 2 were distributed relatively evenly over the year as compared to other NAA's; emissions ranged from about 1.3 to 2 million lbs/month. The principal contributors to 2000 NAA 2 potential emissions were fumigants; more than 55 percent of potential emissions were attributable to products in which metam-sodium, methyl bromide, or 1,3-dichloropropene were the primary active ingredient (Table 3). ECs contributed almost 40 percent of 2000 potential emissions, along with substantial contributions from ready-to-use solutions (23%), pressurized gas (14%), and aqueous concentrate formulations (10%, Table 4).

During the peak ozone forming summer months of June-September (inclusive), NAA 2 potential emissions were similarly dominated by the fumigants metam-sodium, methyl bromide and 1,3-dichloropropene, although at 44 percent, the fumigant contribution was a somewhat lesser portion of NAA 2 emissions during this time than during the entire year.

The distribution of summertime emissions among formulation categories was similar to that seen during the entire year.

TABLE 3. Top ten "primary active ingredients" contributing to 2000 potential VOC emissions in NAA 2, the San Joaquin Valley, listed in order of total emissions of products containing the primary active ingredient. Here the primary active ingredient is defined as the pesticidal active ingredient present at the highest percentage in a product. This approach prevents "double-counting" of emissions from products containing two active ingredients.

Primary AI	**Total product emissions (lbs)**	**% Total product emissions**
METAM-SODIUM	4112959	21.9%
METHYL BROMIDE	3170410	16.9%
1,3-DICHLOROPROPENE	3167038	16.9%
CHLORPYRIFOS	1319593	7.0%
OXYFLUORFEN	615595	3.3%
TRIFLURALIN	560382	3.0%
PENDIMETHALIN	337728	1.8%
GLYPHOSATE, ISOPROPYLAMINE SALT	272143	1.4%
SULFUR	242958	1.3%
ACROLEIN	235236	1.3%

TABLE 4. 2000 Estimated potential emissions in NAA 2 by formulation category.

Formulation code	**Formulation**	**Total emissions (lbs)**	**% Total emissions**
A0	DUST/POWDER	208125	1.1%
B0	EMULSIFIABLE CONCENTRATE	7468715	39.8%
C0	FLOWABLE CONCENTRATE	324368	1.7%
E0	GRANULAR/FLAKE	296600	1.6%
H0	OIL	128530	0.7%
J0	PELLET/TABLET/CAKE	34616	0.2%
L0	PRESSURIZED GAS	2552775	13.6%
M0	PRESSURIZED LIQUID	1081115	5.8%
N0	SOLUBLE POWDER	14819	0.1%
O0	READY-TO-USE-SOLUTIONS	4382619	23.3%
P0	WETTABLE POWDER	276697	1.5%
Q0	AQUEOUS SUSPENSION	18051	0.1%
R0	DRY FLOWABLE	84485	0.4%
S0	AQUEOUS CONCENTRATE	1899503	10.1%

C. NAA 3, Southeast Desert

Potential VOC emissions in NAA 3 have increased significantly since the early 1990s, and 2000 potential emissions were the highest with emissions of approximately 180 percent of the baseline year 1990. Approximately 95 percent of potential 2000 emissions in NAA 3 were attributable to agricultural uses with the remaining 5 percent from commercial structural pesticide applications.

Similar to previous years in NAA 3, the 2000 potential emissions were highest during the winter and early spring months of December and January-March (Figure 2). The fall months of September and October contributed less than 2 percent each of the total year's NAA 3 emissions. Emissions in NAA 3 are driven almost entirely by fumigants; more than 86 percent of 2000 NAA 3 emissions are attributable to products in which metam-sodium, methyl bromide, or 1,3-dichloropropene was the primary active ingredient. The distribution among formulation categories is similarly dominated by those formulation categories to which the fumigant products belong. The principal formulation categories for metam-sodium products are "ready-to-use liquid" and EC, those for methyl bromide are "pressurized gas" or "pressurized liquid", and the most common telone products are classified as "ready-to-use liquid".

TABLE 5. Top ten "primary active ingredients" contributing to 2000 potential VOC emissions in NAA 3, the Southeast Desert, listed in order of total emissions of products containing the primary active ingredient. Here the primary active ingredient is defined as the pesticidal active ingredient present at the highest percentage in a product. This approach prevents "double-counting" of emissions from products containing two active ingredients.

Primary AI	**Total product emissions (lbs)**	**% Total product emissions**
METAM-SODIUM	782222	44.3%
METHYL BROMIDE	604571	34.3%
1,3-DICHLOROPROPENE	132347	7.5%
CHLORPYRIFOS	29094	1.6%
GIBBERELLINS	24237	1.4%
TRIFLURALIN	23874	1.4%
PERMETHRIN	22618	1.3%
DIAZINON	15232	0.9%
BENSULIDE	11073	0.6%
DIMETHOATE	9942	0.6%

TABLE 6. 2000 Estimated potential emissions in NAA 3 by formulation category.

Formulation code	**Formulation**	**Total emissions (lbs)**	**% Total emissions**
A0	DUST/POWDER	4002	0.2%
B0	EMULSIFIABLE CONCENTRATE	363881	20.6%
C0	FLOWABLE CONCENTRATE	12801	0.7%
E0	GRANULAR/FLAKE	17756	1.0%
H0	OIL	148	0.0%
J0	PELLET/TABLET/CAKE	1164	0.1%
L0	PRESSURIZED GAS	456111	25.8%
M0	PRESSURIZED LIQUID	137781	7.8%
N0	SOLUBLE POWDER	231	0.0%
O0	READY-TO-USE-SOLUTIONS	548752	31.1%
P0	WETTABLE POWDER	4809	0.3%
Q0	AQUEOUS SUSPENSION	3033	0.2%
R0	DRY FLOWABLE	601	0.0%
S0	AQUEOUS CONCENTRATE	214012	12.1%

D. NAA 4, Ventura

Potential VOC emissions in NAA 4, Ventura, have steadily increased since the mid-1990s. In 2000, potential emissions were more than 140 percent of the 1990 baseline level. More than 99 percent of potential 2000 emissions in NAA 4 were attributable to agricultural uses, with commercial structural pesticide applications accounting for only one-half of one percent of emissions.

The 2000 NAA 4 potential emissions were highest in the summer months, with the highest emissions occurring in August (Figure 2). Potential emissions in NAA 4 were dominated by fumigants; 89 percent of total emissions were attributable to products in which metam-sodium, methyl bromide, or 1,3-dichloropropene were the primary active ingredient (Table 7). As methyl bromide alone accounted for more than 82 percent of potential emissions, the corresponding formulation categories of methyl bromide products, pressurized gas and pressurized liquid, were the dominant formulations contributing to 2000 NAA 4 emissions.

TABLE 7. Top ten "primary active ingredients" contributing to 2000 potential VOC emissions in NAA 4, Ventura, listed in order of total emissions of products containing the primary active ingredient. Here the primary active ingredient is defined as the pesticidal active ingredient present at the highest percentage in a product. This approach prevents "double-counting" of emissions from products containing two active ingredients.

Primary AI	**Total product emissions (lbs)**	**% Total product emissions**
METHYL BROMIDE	2476563	82.2%
METAM-SODIUM	156693	5.2%
CHLORPYRIFOS	73493	2.4%
1,3-DICHLOROPROPENE	46299	1.5%
PETROLEUM OIL, UNCLASSIFIED	32457	1.1%
METALDEHYDE	27969	0.9%
OXAMYL	22229	0.7%
GLYPHOSATE, ISOPROPYLAMINE SALT	13462	0.4%
AVERMECTIN	11154	0.4%
GIBBERELLINS	9533	0.3%

TABLE 8. 2000 potential emissions in NAA 4 by formulation category.

Formulation code	**Formulation**	**Total emissions (lbs)**	**% Total emissions**
A0	DUST/POWDER	712	0.0%
B0	EMULSIFIABLE CONCENTRATE	315047	10.5%
C0	FLOWABLE CONCENTRATE	12983	0.4%
E0	GRANULAR/FLAKE	13788	0.5%
H0	OIL	17379	0.6%
J0	PELLET/TABLET/CAKE	20546	0.7%
L0	PRESSURIZED GAS	1471876	48.9%
M0	PRESSURIZED LIQUID	960371	31.9%
N0	SOLUBLE POWDER	758	0.0%
O0	READY-TO-USE-SOLUTIONS	121919	4.0%
P0	WETTABLE POWDER	7497	0.2%
Q0	AQUEOUS SUSPENSION	1704	0.1%
R0	DRY FLOWABLE	1882	0.1%
S0	AQUEOUS CONCENTRATE	64868	2.2%

E. NAA 5, South Coast

Potential VOC emissions in NAA 5 decreased dramatically early in the 1990s, and stayed at constant low level since 1993. In 2000, potential emissions were approximately 35 percent of the 1990 baseline level, far below the 2005 reduction goal (Figure 1). Approximately 55 percent of potential 2000 emissions in NAA 5 were attributable to agricultural uses, with commercial structural pesticide applications accounting for the remaining 45 percent of emissions.

The distribution of 2000 NAA 5 potential emissions were relatively even over the year, with the exception of August which was much higher than other months (Figure 2). While the fumigant contribution to 2000 NAA 5 emissions was less than in other NAAs, the fumigant products containing methyl bromide and metam-sodium as primary active ingredients still comprised 46 percent of the total emissions. The organophosphate and pyrethroid insecticides - chlorpyrifos, diazinon, permethrin, bifenthrin and cypermethrin - contributed more than 31 percent of total 2000 NAA 5 emissions. Numerous products containing these insecticides are formulated as ECs, and this is reflected in the formulation category breakdown in Table 10.

TABLE 9. Top ten “primary active ingredients” contributing to 2000 potential VOC emissions in NAA 5, the South Coast, listed in order of total emissions of products containing the primary active ingredient. Here the primary active ingredient is defined as the pesticidal active ingredient present at the highest percentage in a product. This approach prevents “double-counting” of emissions from products containing two active ingredients.

Primary AI	**Total product emissions (lbs)**	**% Total product emissions**
METHYL BROMIDE	932532	43.8%
CHLORPYRIFOS	251193	11.8%
DIAZINON	181689	8.5%
PERMETHRIN	115664	5.4%
BIFENTHRIN	62409	2.9%
CYPERMETHRIN	50741	2.4%
METAM-SODIUM	48553	2.3%
PROPOXUR	37767	1.8%
N-OCTYL BICYCLOHEPTENE DICARBOXIMIDE	30295	1.4%
DITHIOPYR	29997	1.4%

TABLE 10. 2000 potential emissions in NAA 5 by formulation category.

Formulation code	**Formulation**	**Total emissions (lbs)**	**% Total emissions**
A0	DUST/POWDER	6772	0.3%
B0	EMULSIFIABLE CONCENTRATE	754342	35.5%
C0	FLOWABLE CONCENTRATE	12060	0.6%
E0	GRANULAR/FLAKE	137153	6.4%
H0	OIL	2976	0.1%
J0	PELLET/TABLET/CAKE	11166	0.5%
L0	PRESSURIZED GAS	899458	42.3%
M0	PRESSURIZED LIQUID	199741	9.4%
N0	SOLUBLE POWDER	1492	0.1%
O0	READY-TO-USE-SOLUTIONS	37538	1.8%
P0	WETTABLE POWDER	10187	0.5%
Q0	AQUEOUS SUSPENSION	9221	0.4%
R0	DRY FLOWABLE	2381	0.1%
S0	AQUEOUS CONCENTRATE	41952	2.0%

IV. POTENTIAL VOC EMISSION ESTIMATES AND THEIR UNCERTAINTY

(i) About estimates of potential VOC emissions

The potential VOC emission from a given pesticide product application is calculated as follows.

potential VOC emission (pounds) = (pounds product applied) x EP

Consequently the accuracy of DPR's annual estimated potential VOC emission inventory depends on the accuracy of both emission potential (EP) data and the pesticide use report (PUR) data.

(ii) What are EPs?

The EP is that fraction of a pesticide product that is assumed to potentially contribute to atmospheric VOCs. Consequently the VOC emission estimates reported in this inventory are estimates of *potential emissions* – that is, they assume that the EPs calculated based on composition or TGA measurements represent the *maximum* actual product volatilization under field use conditions. In reality, numerous factors reduce a product's actual emission potential. As an example, post-application degradation/reaction of soil-injected fumigants decreases their actual emissions relative to emission estimates based on amount of product applied and the assumed 100 percent emission potential. There are numerous other factors and/or use practices that reduce a product's post-application volatilization. These include sorption to soil, absorption by plant leaves, soil incorporation, post-application water-in, etc. The emission estimates are best considered as maximum possible emissions from the reported pesticide applications.

(iii) EP: water/inorganic subtraction vs. TGA vs. default

The current "gold standard" for determining EPs is TGA. For products without TGA data, other estimation methods are employed (Spurlock, 2002a). These include the use of default EPs, or EPs calculated using water and/or inorganic subtraction methods, where the entire product is assumed volatile except for the water and/or inorganic fraction determined from the product's confidential statement-of-formula.

Requests to registrants for TGA EP data were made in the early- to mid-1990s. Since that time the annual number of submitted TGA EP data packages has steadily declined. Currently DPR receives only a few data packages each year. TGA data for many products used in the early 1990s are similarly sparse. Consequently, the fraction of emissions estimated from water and/or inorganic subtraction or defaults is increasing (Figure 3a), contributing uncertainty to recent year VOC emission estimates.

(iv) PUR data

California's mandatory PUR system was initiated in 1990. Certain characteristics of the PUR data raise questions concerning (a) accuracy of reported data in earlier years, and (b) reporting compliance in earlier years. Standardized statistically-based procedures have been developed to identify probable "outliers" in the PUR (Wilhoit, 1998), where outliers are data that are so extreme in value as to probably be in error. Here outliers are identified as those PUR records that exceed criterion 1a <u>or</u> criterion 2b <u>or</u> criterion 4d presented in Wilhoit, 1998. Based on these definitions, the mass fraction of total reported PUR use data [pounds product applied] comprised of outliers has been steadily decreasing since 1990, from about 6-8 percent to about 2-4 percent in recent years (Figure 3b). The VOC calculation procedure completely excludes probable outliers; consequently a larger fraction of reported pesticide applications were omitted from the VOC inventories for the early 1990s than from the inventories for more recent years. The net effect is that the VOC emission estimates for the early 1990s are probably biased downward because a greater portion of PUR data were excluded from inventory calculations in early years.

The total number of pesticide applications reported in the PUR has sharply increased from about 1.95 million in the early 1990s to about 2.6 million in recent years (Figure 3c). Similarly, total pounds of reported pesticide products applied (excluding outliers) also increased steadily during the early 90's (Figure 3d). These data suggest that reporting compliance was lower in the early years of the PUR program. If so, the net effect of lower reporting compliance would also be to reduce reported pesticide use (and estimated VOC emissions) in the earlier years.

V. SUMMARY

The current inventory indicates that the Southeast Desert (NAA 3) and Ventura NAAs (NAA 4) will probably experience the greatest difficulty in meeting the 2005 VOC emission goal of 80 percent of estimated 1990 VOC emission levels. Both of these NAAs have demonstrated a trend of increasing estimated emissions since the mid-1990s. In 2000, over 85 percent of estimated VOC emissions in both of these NAAs were attributable to agricultural applications of products containing the fumigants methyl bromide, metam-sodium, or 1,3 dichloropropene.

Data for the Sacramento Metropolitan Area (NAA 1) has been highly variable in recent years. While year 2000 estimated VOC emissions approached the 2005 reduction goal, emissions in other recent years have been much higher. Consequently, there is no clearly established trend in VOC emissions in NAA 1. In contrast, there has been a significant downward trend in San Joaquin (NAA 2) estimated emissions since 1995; the 2005 goal will be easily met before 2005 *if the current trend in NAA 2 estimated emission continues*. Estimated emissions in the South Coast (NAA 5), are already well below the 2005 reduction goal as they have been for several years.

The potential VOC emission estimates discussed in this memorandum are based on EPs that describe the maximum emissions from a pesticide application; actual emissions may be less than the estimates reported in this inventory. There is significant uncertainty in estimated potential VOC emissions arising from uncertainty in the underlying PUR data, especially in earlier years. In addition, some data suggest that there may have been low reporting compliance in the early years of the PUR. Both effects would serve to bias calculated emissions for the early 1990s downward.

VI. REFERENCES

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Spurlock, F. 2002a. Memorandum to J. Sanders. Methodology for determining VOC emission potentials of pesticide products. January 7, 2002. Environmental Monitoring Branch, Dept. Pesticide Regulation.

Spurlock, F. 2002b. IN PREPARATION. Procedures for estimating potential VOC emissions from California pesticide use data. EH report. Environmental Monitoring Branch, Dept. Pesticide Regulation.

Attachments

cc: Randy Segawa, Senior Environmental Research Scientist (w/Attachments)
Mark Pepple, Senior Environmental Research Scientist (w/Attachments)

bcc: Spurlock Surname File

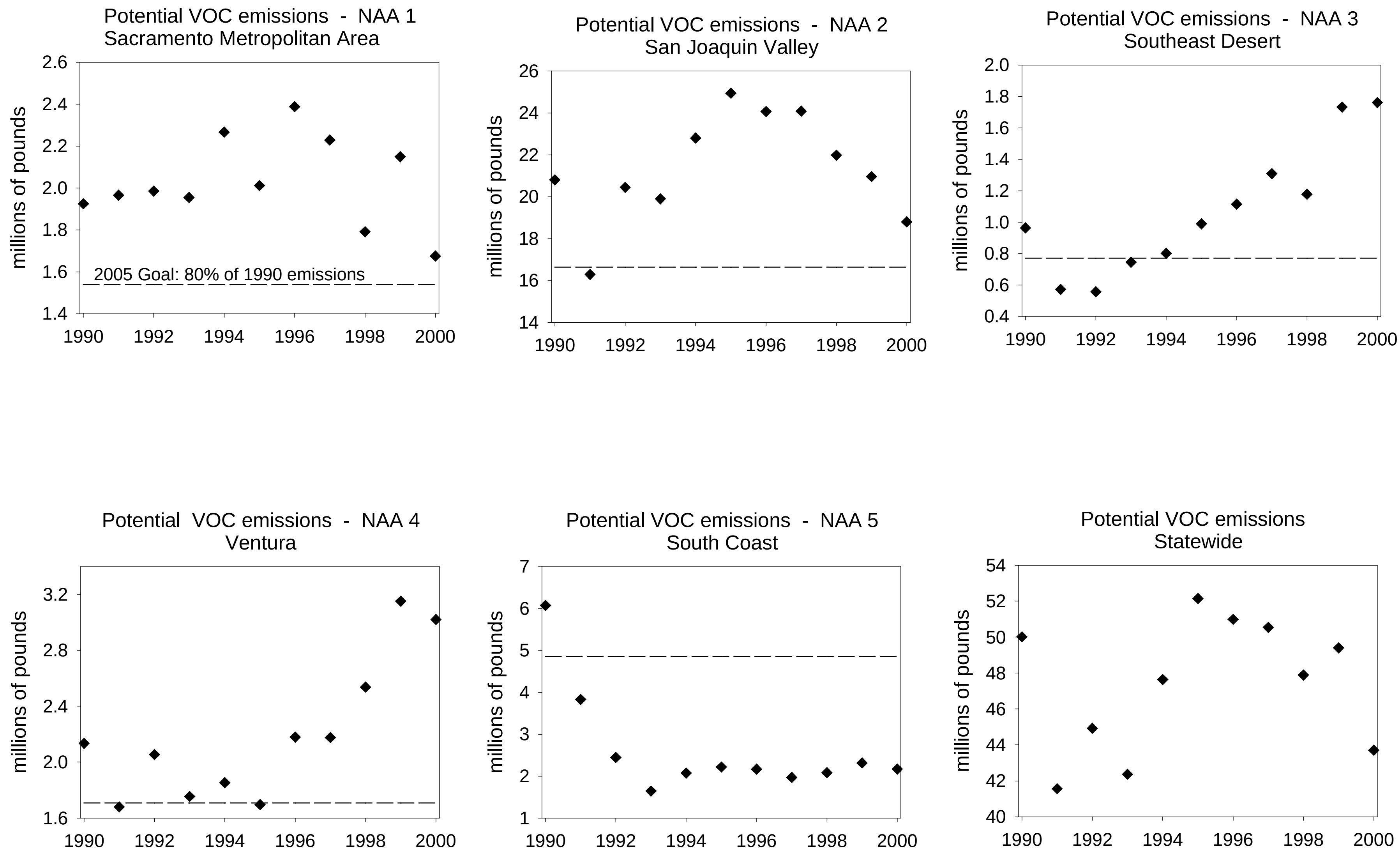

Figure 1. 1990 - 2000 estimated potential VOC emissions from agricultural and commercial structural pesticide use by non-attainment area (NAA)

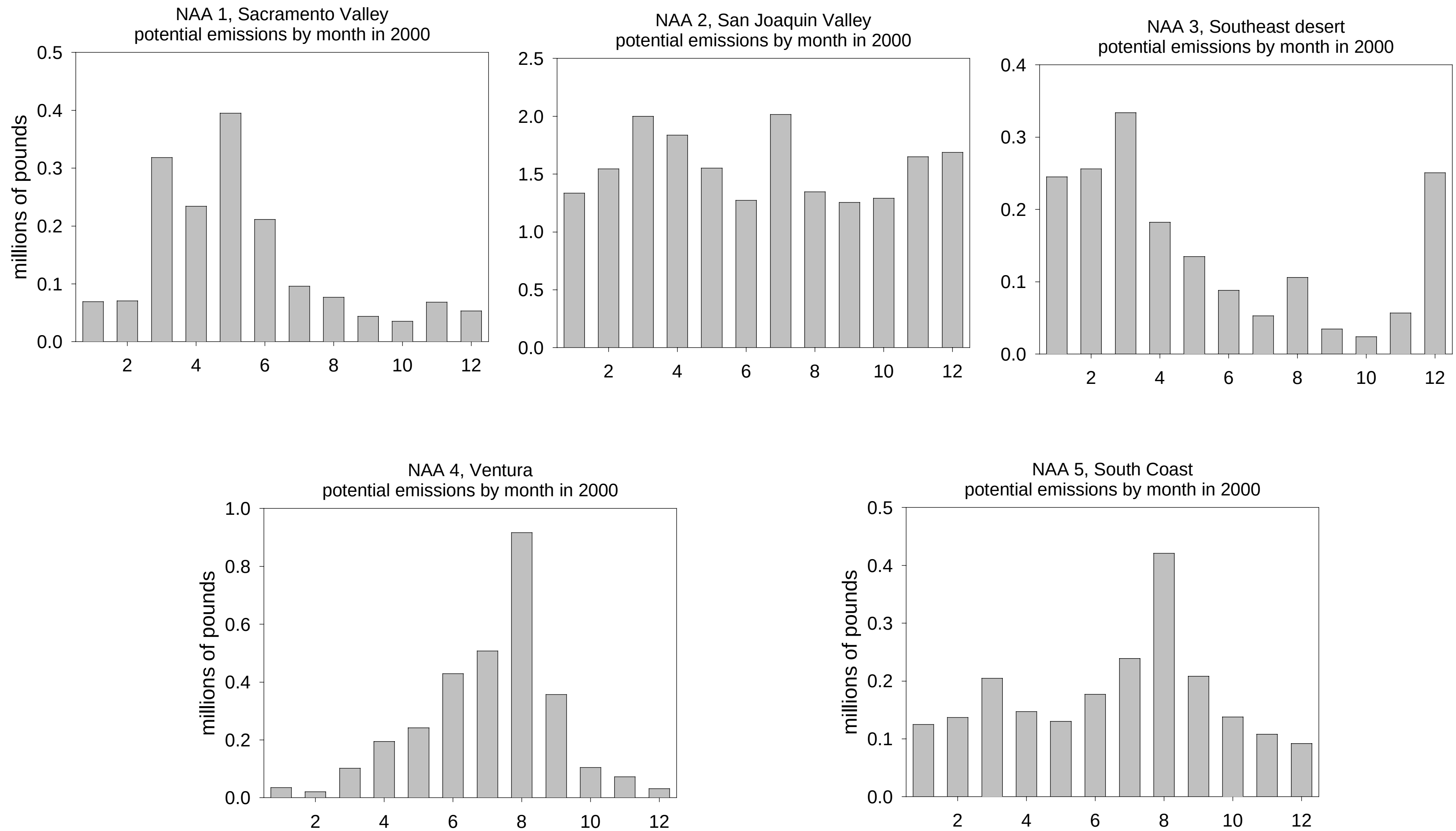

Figure 2. 2000 estimated VOC emissions by month in individual NAAs.

note y-axis scale differences among graphs

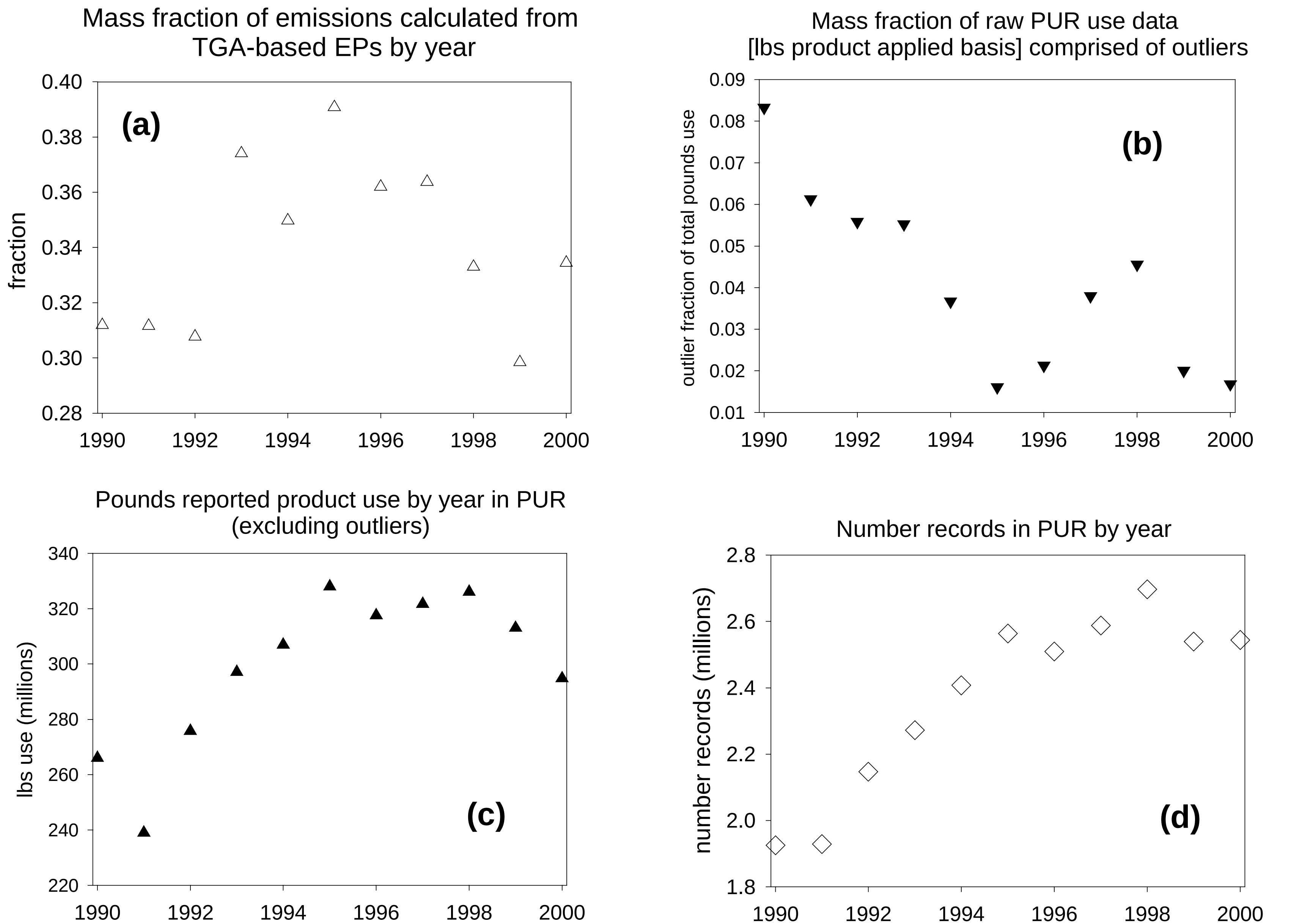

Figure 3. Factors that may affect relative accuracy of VOC inventory among years