Dairy Permitting Advisory Group

Dairy Emissions Factors for Volatile Organic Compounds

Recommendation to the San Joaquin Valley Air Pollution Control Officer Regarding VOC Emissions From Dairies. Final Report - May 6, 2005

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I. Introduction

The purpose of this report is to advise the Air Pollution Control Officer of the San Joaquin Valley Air Pollution Control District (District) on a method for estimating atmospheric emissions of Volatile Organic Compounds (VOCs) from dairies. The Dairy Permitting Advisory Group (DPAG) is tasked with recommending an "emissions factor" or factors to be used for calculating overall emissions of VOCs [1] from dairies.

DPAG is a committee of community members and experts from a variety of fields. As part of a litigation settlement[2], the DPAG was tasked as follows:

> *"…develop a means for determining the volume of VOC emissions from individual dairies, including the development of a Dairy Emission Factor or Dairy Emission Factors for the dairy facility as a whole and, to the extent possible, for individual units that may be part of individual dairy operations, such as manure lagoons. The DPAG will issue a written report recommending a Dairy Emission Factor no later than April 15, 2005, such report will include a discussion of the scientific information considered and evaluated by the DPAG."*

Over the last three months, DPAG has reviewed the latest information available related to measuring and estimating emissions of VOCs from dairies. Information has been provided in written form and through spoken testimony before DPAG. Taken as a whole, this information has greatly increased our current understanding of VOC emissions from dairies.

DPAG wishes to thank the researchers who have worked hard to complete their work to meet the needs of regulators, the dairy industry and valley residents, and who have repeatedly taken time out of their schedules to meet with DPAG, discuss findings and answer questions. Without these dedicated researchers this report would not have been possible.

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[1] See Appendix A for formal definition.

[2] Western United Dairymen, Alliance of Western Milk Producers v. San Joaquin Valley Air Pollution Control District, settled in the Fresno Superior Court September 2004.

Quality of the Data:

This report is released on May 6, 2005, the revised date from April 15, 2005, as mutually agreed to by the parties, because of the legal obligation previously mentioned, not because DPAG now has all the necessary data to do the best job possible. All parties involved agree that additional information is necessary before fully understanding dairy VOC emissions, and that the emissions factors that are recommended herein will necessarily evolve as more data become available.

There are also limitations to the comprehensiveness of the sampling and analytical tools available and used by the researchers. In addition, many VOCs that are known to be emitted from dairy operations, such as oxygenated hydrocarbons and volatile fatty acids, require specialized techniques and protocols. The data for these emissions are therefore also limited. Conclusions reached in this report on the quantities of these harder-to-measure VOCs are therefore open to debate.

Finally, this report relies, in part, on data that have not been previously published, and have not been rigorously peer reviewed.

Transfer of Results:

Different types of sampling and analytical methods were used to gather the data contained in this report. The measurements took place at two operating dairies in the San Joaquin Valley (one in Kings County and one in Merced) and at a purpose-built chamber at University of California, Davis. Some field-based tests measured surface flux from various processes at a dairy. Some tests measured ambient concentrations then used those measured concentrations to model emission rates from combinations of sources. The UC Davis tests measured emissions using environmental chambers. Different sampling and analytical methodologies were employed by these researchers. These differences in test procedures and methodologies lead to data that are not always directly comparable between studies.

In addition, some researchers have reported efforts to capture maximum emissions from given processes, while others have reported no such efforts. The effect of management practices on VOC emissions is largely unknown. The studies were performed in the field at two dairies, both flush dairies with freestalls, located in California's San Joaquin Valley. These dairies were considered typical of current Valley dairies.

Some DPAG members concluded that these California studies did not measure all potential VOC compounds and sources, and therefore reviewed additional documents provided by other research efforts. However, other DPAG members concluded that data from California studies, which were specifically designed to measure VOCs from California dairies do provide the best basis for an emission factor.

We have attempted to compare and combine the results of all of these studies, as appropriate, while compiling the best possible recommended Dairy Emission Factors. However, these results may not be transferable to other climates or other dairy designs, nor are we attempting to describe how dairy management practices might influence these results.

Given these limitations, it is important that any Dairy Emission Factors developed by the DPAG not be considered as the final step in developing VOC emissions estimation techniques for dairies. There are many ongoing and anticipated efforts to design and perform appropriate research and to provide more and improved data related to VOC emissions from dairies, and we expect and encourage interested parties to continue to work together to enhance the available data and to incorporate that data into ever-improving dairy emissions estimating techniques. To that end, we have provided some research design recommendations at the end of this report that we believe can enhance the effectiveness and usefulness of these future efforts.

II. Description of Approaches to Analyzing Available Data

DPAG was able to achieve consensus on two basic approaches to analyzing the available data for an emission factor. The first, "process-based" approach builds an emission factor primarily on the work of two research teams, one led by Dr. Frank Mitloehner of the University of California, Davis, and the other led by Dr. Chuck Schmidt, an independent researcher. Dr. Mitloehner's work focused on measuring emissions directly from cows, their fresh waste and feed in environmental chambers. Data are preliminary and are described in a letter from Dr. Mitloehner to the District. In the case of Dr. Schmidt, the research is described in a paper by Schmidt et al, delivered at the 14th USEPA Annual Emissions Inventory Conference in April 2005. Schmidt's work focused on using flux chambers to measure emissions from surfaces at representative parts of dairies, including dairy lagoons, corrals, manure piles and other surfaces. This process-based approach also depended on additional data as will be described in Section III.

The second "whole-dairy" approach builds an emission factor based primarily on the work of a research team led by Dr. Charles Krauter of California State University at Fresno. The Krauter team took air samples upwind and downwind of dairies in Merced and Kings County and used an atmospheric model and sample analysis technique to measure VOC emissions. Additional data were also relied upon in this whole-dairy approach as described in Section IV.

While DPAG was able to reach a consensus that these general approaches are valid, DPAG was not able to reach consensus on each constituent measured and reported. There was substantial disagreement, to list just two examples, in the interpretation of scientific data on volatile fatty acids (VFAs) emissions and on amine emissions. In the interest of making a useful recommendation to the APCO, DPAG is reporting the areas of agreement and disagreement in detail as three separate viewpoints, resulting in three differing total dairy emission factors.

III. Process-Based Approach

After substantial discussion, the DPAG agreed that the following table represents an adequate summary of the process-based approach to calculating the major constituents of VOC. Where there is disagreement, alternate methods for calculating an emission factor are reported for each constituent, with notes provided to explain the case for each method. (*Please note that DPAG members agreed to the convention of rounding off emission factors to increments of 1/10 of 1 pound per cow per year*).

Process or Constituent	**Emissions (lb/hd-yr) Viewpoint 1**	**Emissions (lb/hd-yr) Viewpoint 2**	**Emissions (lb/hd-yr) Viewpoint 3**
1. Emissions from Cows and Feed in Environmental Chamber	2.7	3.4	4.3
2. Amines from dairy processes	0.2	0.2	11.0
3. VOCs (except VFAs and Amines) from miscellaneous dairy processes	1.2	1.2	1.2
4. VOCs (except VFAs and Amines) from lagoons and storage ponds	1.0	1.0	1.0
5. Volatile Fatty Acids (VFAs) (From dairy processes)	0.5	7.5	17.0
6. Phenols (From dairy processes)	0	0	2.6
7. Land Application	NA	NA	1.0
8. Feed storage, settling basins, composting, & manure disturbance	Included Above or Insignificant	NA	0.1
Total	**5.6**	**13.3**	**38.2**

NA: Not Available

General Comments for Viewpoint 1

Where there is disagreement, alternate methods for calculating an emission factor are reported for each constituent, with notes provided to explain the case for each method. **Proponents of Viewpoint 1 do not recommend that the APCO calculate a "compromise" between alternate methods recommended here**. Rather we recommend a careful review focused on the areas of disagreement. Different factors represented here often generally represent substantial disagreement in the approach and interpretation of scientific data. We believe selecting a numerical value in the mid-range is not an appropriate method for resolving this disagreement. We strongly recommend that before adopting values, the APCO first examine the differences carefully and adopt the most valid, science-based approach. The researchers utilized EPA approved methodologies and provided sampling protocols and field plans that were designed to capture specific compounds by the SJAPCD and CARB.

General Comments for Viewpoint 2

Emissions factors for Viewpoint 2 were presented to DPAG as "Proposed Compromise VOC Emissions Factors, 5/1/2005"

General Comments for Viewpoint 3

It is the view of some members of the DPAG that the data on which we are supposed to base a VOC emission factor recommendation are insufficient and may result in an artificially low factor. We believe that the research outlined above may underestimate VOC emissions from California dairies for the following reasons:

- Several significant VOC compounds known to be emitted from dairy processes were not measured, were measured using inadequate methodology or unreliable analysis, or considered by the researcher to be too preliminary to release for DPAG's use.

- Several emissions sources related to dairies were not included in this research. Additionally it should be noted, that the California field research was limited to two dairies. Procedures that occurred prior to testing may have affected results, for example: some tests were conducted after recent corral scraping, which is generally done very few times per year.

- The lack of data on three of the most important components of VOCs from dairies is troubling. Published papers cite VFAs, amines and aromatics including phenols as important VOC constituents of emissions from dairies and animal waste in general. (Miller & Varel). These three constituents together with alcohols and esters are commonly reported odor compounds.

- Other gaps in the data from the four California studies relate to the fact that they may not have covered all processes related to dairies. Land application of manure, feed storage and solid separation are just a few of the processes that are not represented, but are expected to contribute significantly to VOC emissions. For example, surface application of manure increases VOC emissions, forming wide odor plumes that are notably high in wet conditions.

Given these considerations, it is apparent that emission factors derived from the four recent California research projects do not adequately represent total VOC emissions from dairies. We have added supplemental data from studies published in peer review journals to fill in the remaining information gaps, which appear to be quite large.

Constituent/Process 1 – Emissions from cows and feed in environmental chamber

Case for Viewpoint 1

Dr. Mitloehner collected data and analyzed samples using two different technologies. The first was Proton Transfer Reaction Mass Spectrometer (PTRMS). This technology provides real-time quantification of numerous VOC compounds with detection limits as low as a few parts per trillion. These compounds include alkenes, alcohols, aldehydes, aromatics, ketones, nitriles, sulphides and others (Europhysics News (2004) Vol. 35 No. 6). Some DPAG members have contended that additional compounds (1,2,4 trimethylbenzene and 1,3,5 trimethyl benzene) should be added to a base number of 2.7 to "correct" for these missing compounds. However, these compounds were also included in the TO-15 standard used by Dr. Mitloehner, also referred to in his letter. Barring statements to the contrary by Dr. Mitloehner, there is no evidence to support that these compounds should be added to the emission factor. We recommend that the APCO confirm through consultation with Dr. Mitloehner that his emission factor included a review of both TO-15 data and PTRMS.

Case for Viewpoint 2

Enteric and feed VOCs and VOCs from fresh excreta were measured by PTRMS, based on Dr. Mitloehner's 4/13/05 presentation and 4/17/05 letter to DPAG. Dr. Mitloehner estimates from PTRMS measurements that 3 lb/hd-yr VOC are emitted from the cow and fresh excreta in environmental chambers. This number was adjusted downward by 10%, to 2.7 lb/hd-yr, to represent only the enteric emissions, since previous measurements had shown that emissions from fresh excreta represented approximately 10% of emissions in the test chamber.

Although these tests provide the best available data for enteric and feed emissions, this still may be a significant underestimate of these compounds since many VOC compounds that may be present in enteric emissions such as ethanol were not detected by the PTRMS system. Dr. Goldstein, who performed the PTRMS measurements, indicated to DPAG that PTRMS would not effectively measure amines, or volatile fatty acids or phenols, which are difficult to collect through sampling tubes.

An additional 0.7 lb/hd-yr VOC was added to the 2.7 lb/hd-yr VOC based on data from TO-15 measurements of cows in environmental chambers presented by Dr. Mitloehner on 1/26/2005 (See calculations below). Aromatic compounds that are not counted by PTRMS were added. Other compounds measured in tests that may be misidentified and are not expected in cow emissions (e.g., chlorinated solvents, fluorocarbons, etc.) were excluded.

This may be a significant underestimate since many VOC compounds that may be present in enteric emissions are not properly detected and identified by Method TO-15 and PTRMS (amines, VFAs, phenols).

Finally, although Mitloehner's preliminary data have indicated significant VFAs in enteric/fresh manure emissions, he has stated that the VFA concentration measurements are so uncertain that they should not be used in determining VOC emissions from dairies. Therefore, no attempt is made to account for enteric VFA emissions in this proposal.

Calculation: From Dr. Mitloehner's results presented to DPAG on 1/26/2005 for cow and excreta.

1,2,4 trimethylbenzene 0.104
1,3,5 trimethylbenzene 0.236
0.340 kg/hd-yr

90% of 0.340 is 0.306 kg/hd-yr (an estimated 90% of emissions is from cow)

2.2 lb/Kg * 0.306 kg/hd-yr = 0.7 lb/hd-yr

Case for Viewpoint 3

See Case for Viewpoint 2 - Emissions from cows and feed in environmental chamber above. For viewpoint 3, total VFAs and Phenols are calculated elsewhere.

Constituent/Process 2 – Amines from Dairy Processes

Case for Viewpoint 1

Studies completed to date do not support anything other than a low emission factor. The work of Dr. Chuck Schmidt provides a quantification of ethyl amine which is likely to be a slight overestimate of this compound but contains no other amine compounds. Preliminary data reported by Dr. Mitloehner (letter dated 4/17/05) may contain other amines, such as Trimethyl Amine, however, without speciated data it is impossible at this time to determine with any accuracy how complete this information would be. An overestimation margin inherent in Dr. Mitloehner's report of the preliminary data may be sufficient to capture any amines not captured by the PTRMS and TO-15 methods he used; however, there is no way to verify this at this time. In any case amines captured by Dr. Mitloehner would be representative of only a portion of the processes on a dairy, e.g. emissions directly from cows, their fresh waste, and from feed.

An emission factor for amines based on the work of Rabaud cannot be used. We note that this study did not calculate any emission factors for amines but reported only concentrations. Nothing in the Rabaud work suggests a ratio of VFAs and amines can be assumed. For VFAs, standards for 11 gases were used and only two gases, acetic acid and valeric, resulted in any detects. There were only four detects (out of 48 samples) for acetic acid and one detect for valeric acid (out of 48 samples). Amine sampling also did not produce consistent results. 1,4-butanediamine appeared in only one (of 48) samples, 2-methyl-1-propanamine was detected in two samples, and diethylamine also appears in only two samples. Only

butylamine was found consistently (20 samples). Given these results, there is no basis to support even at this site a consistent emission rate for either compound or a ratio between the two.

As Dr. Meyer reported to DPAG (letter, 5/4/05), differences in the basic biological processes that create VFAs and amines vary widely depending on feedstuffs, and therefore cannot be compared from study to study.

Given this lack of data for quantification, we recommend that the Schmidt data provide the only reliable measurement.

Case for Viewpoint 2

Non-enteric amine emissions from miscellaneous dairy processes were directly measured using flux chambers and are based on data submitted to DPAG from C.E. Schmidt through CARB. This may be a significant underestimate since many common processes, including solids separation, composting, and land application, were not tested by Dr. Schmidt, and perhaps as important, no amines other than ethylamine were reported. Several other amines are believed to exist in dairy emissions

Case for Viewpoint 3

Total amine emissions were calculated based on an average of data from Schmidt, Ngwabie 2005, and Rabaud 2003.

The Schmidt emission factor of 0.18 lb amines/head-year is based on non-enteric amine emissions from miscellaneous dairy processes directly measured using flux chambers. Based on data submitted to DPAG from C.E. Schmidt through CARB, this may be a significant underestimate since no amines other than ethylamine were reported, though other amines are known to exist in dairy emissions, as evidenced by the estimates below.

Rabaud, et al, Atmospheric Environment 37 (2003) 933–940 reports the following VFAs and amines sampled from a northern California dairy:

	Min. ug/m3	Max ug/m3
Acetic Acid	37.1	414.9
Valeric Acid		87.92
Total VFAs	37.1	502.82
1,4 butanediamine		70.42
butylamine	81.64	561.43
2-Methyl-1-propananamine	104.21	110.88
Diethylamine	190.7	231.34
Total Amines	376.55	974.07

Total Amines are 2.5 times higher than VFAs. Using the factor for VFAs above, total amines = 32.7 lb/head-year

Ngwabie 2005 reports emissions from a cowshed housing 120 cows in Germany. Trimethylamine was the only amine reported, with a concentration reported as 14ppb as compared to ammonia reported at 4ppm. Using the ratio of amines to ammonia, amines were estimated at 0.03 lb/head-year. This is a significant underestimate since only one amine was reported.

The total average amines = (0.18 + 32.7 + 0.03)/3 = **11.0 lb/head-year**

Constituent/Process 3 - VOCs (except VFAs and Amines) from miscellaneous dairy processes

Consensus among all three viewpoints. Based on data submitted to DPAG from C.E. Schmidt through CARB.

Constituent/Process 4 - VOCs (except VFAs and Amines) from storage ponds and lagoons

Consensus among all three viewpoints. Based on an average of Merced County and Kings County measurements by Dr. Krauter and Dr. Schmidt using Method TO-15.

Constituent/Process 5 – Volatile Fatty Acids from dairy processes

Case for Viewpoint 1

Volatile fatty acids (VFAs), also known as volatile organic acids, were measured from processes by Dr. Chuck Schmidt. For determining VFA emissions at local San Joaquin dairies, the Schmidt et al. study is the most appropriate, as it was the only study reviewed by DPAG that measured VFAs under conditions at a California dairy.

Dr. Schmidt measured VFA emissions at multiple process locations (feedlanes, corrals, lagoons, etc.) using an EPA-approved test method (USEPA validated chamber method used for the flux measurement), all measurements were made in situ, an EPA-approved analytical method (Gas chromatography/High performance liquid chromatography/Ultraviolet Visible Absorption Spectrophotometer). Testing was conducted at all significant dairy area sources (corrals, flushed lanes, lagoons, etc.) with the U.S. EPA-validated chamber. All samples collected and analyzed resulted in non-detects, with detection limits 60 ppbv to 260 ppbv.

Two emission estimates were developed: 1) assuming that acetic acid was at the detection limit for all processes and that it represented 37% of the total VFAs; and 2) assuming a more realistic estimate of actual acetic acid emissions of 1/3rd the detection limit based on CARB guidance on how to proceed concerning measurements below the detection limit when no measurements were reported above the detection limit. The ratio of acetic acid to total VFAs was taken from Koziel, et al. This is conservative, based on McGinn et al., which reports that acetic acid was 54 to 67% of total VFAs. It is also consistent with the dairy characterization study, Rabaud et al.

Using a worst-case assumption of VFA emissions at the detection limit for all processes, the VFA emissions are 1.42 lbs/head-year. Since all measurements at all processes were below the non-detect limits, a more reasonable assumption using the CARB method is that VFA emissions are 0.47 lbs/head-year. In the table above, this is rounded to 0.5. This estimate includes emissions from flushed lanes (pre- and post-flushed); feedlane pile; freestall bed; turnouts; heifer pens; milk parlor; separator solids, storage pile solids; bedding pile solids; primary lagoon (see also "VFA Emission Rate Technical Assessment" prepared by Dr. Julia Lester).

While the work done on California dairies is the most representative of California dairies, work by Koziel et al on Texas feedlots corroborates the Schmidt research. Koziel calculated an emission factor of .53 lbs/steer-year in a situation with higher stocking density. Because beef steers in general are fed diets higher in starch than dairy cattle, more starch is excreted and deposited in a smaller area than on a California dairy. This suggests that emission rates per meter squared would be higher on a beef cattle feedlot than on a dairy. Nutritional studies suggest that starch excretions can be as much as five to eight times higher per unit in beef manure compared to dairy manure (Avila et al, Espindola et al, Joy et al).

Some DPAG members have suggested VFAs be calculated based on the work of McGinn et al; this line of thinking suggests that McGinn can be used to establish a ratio of ammonia to VFAs, and consequently, VFAs can be calculated based on the California dairy emission factor for ammonia. However, the McGinn work is a study of beef feedlots in Canada, which by nature do not represent California dairy conditions. Also, McGinn was conducted in entirely different climatic conditions than the California studies. Furthermore, there is no evidence to suggest that a downwind sample-based ratio is reflective of conditions on the beef feedlots themselves or that the ratio of ammonia to VFAs remains constant between the feedlots and the downwind sampling station three miles away. As McGinn notes, "much of the emitted ammonia was deposited to soil immediately downwind, enough to supply all of the nitrogen needed for crop growth." Hobbs cannot be used as a basis for calculating VFA emissions. There is no scientific basis for applying the emission factors from Hobbs to California dairies (presentation of Dr. Julia Lester to DPAG, April 13, 2005; also comments of Dr. Deanne Meyer during meetings). This is also discussed in the phenols section (viewpoint 1) of this report. Finally, it makes more sense to utilize direct surface measurements of VFAs from a California dairy (Schmidt) than to extrapolate a relationship between ammonia and VFAs from Canadian beef feedlots and attempt to apply that to California conditions.

Case for Viewpoint 2

Non-enteric Volatile Fatty Acid (VFA) emissions were estimated assuming approximately 60-80% of manure being flushed to the separators and lagoons (from California RWQCB Fact Sheet #4 for Nutrient and Irrigation Water Management Plans). Used 60% in this calculation, to conservatively estimate emissions on the low side in order to propose an emissions factor. Emission factor from Koziel, et al (1.17 lb/hd-yr, discussed in DPAG meeting, 4/13/05, after adjusting for dairy cow food intake), was used for the assumed 40% dry manure conditions in corrals, since these tests were performed in summertime drylots in Texas. Emissions rate from

Hobbs, et al (11.9 lb/hd-yr, from the Hobbs report, 11.4 million dairy cattle produce 61.6 kilotons of VOCs per year), were used for the assumed 60% of manure that goes to wet processes including flush lanes, separation ponds, and lagoons, since these tests were performed using manure slurries. Neither of these tests would have measured enteric VFAs, so enteric VFAs remain uncounted in this emissions factor.

Dr. Schmidt's results were not included in viewpoint 2 based on indications from other researchers that the method used by Schmidt, which included a flux chamber and a long sampling line, would not yield reliable results for VFAs, which tend to stick to sampling apparatuses.

Case for Viewpoint 3

The only researcher to provide VFA data based on valid and reliable sampling techniques withheld the data. VFAs are known to come from undigested starch. Feed digestion produces the vast majority of VOCs in the form of VFAs, alcohols, aldehydes & ketones (Rabaud, 2003). Aged manure is believed to release twice the amount of VFAs as fresh manure (Miller & Varel).

Volatile Fatty Acid (VFA) emissions were estimated taking an average of VFA factors derived from the following studies: Hobbs, et al (11.9 lb/hd-yr, from the Hobbs report, 11.4 million dairy cattle produce 61.6 kilotons of VOCs per year), McGinn 2003 and Ngwabie 2005. The last two factors were developed by taking an average VFA number and using the ratio between VFAs and ammonia multiplied by a California ammonia factor per the following data and calculations:

Average VFA-to-ammonia ratio (expressed as percentage) applied to current ammonia EF of 74 lbs-NH_3/hd-yr (used in state-wide emissions inventory applications):

McGinn,

Average VFAs (6000 head feedlot) = 23.3 μg/m^3

Average Ammonia (6000 head feedlot) = 130 μg/m^3

18% (VFA concentration/Ammonia Conc.) x 74 lb NH3/hd-year = 13.3 lb VFA/head-year

Ngwabie,

Reported median Ethanol/Formic Acid3 = 1.4 ppm

Reported median ammonia = 4.0 ppm

35% (VFA conc. / Ammonia Conc.) x 74 lb NH3/hd-year = 25.9 lb VFA/head-year

For comparison purposes:

Average ammonia emissions per Idaho Conservation League and Independent Dairy Environmental Action League (IDEAL)[4] report: (122 + 71 + 39)/3 = **77.3 lbs-NH3/hd-yr**

[3] This was reported as "ethanol/formic acid" likely because the molecular weights of both compounds are the same: 46, indicating that the signal could represent either compound or a mixture of the two. We assumed conservatively (worst case) that formic acid, a VFA, accounted for the whole concentration of 1.4 ppm.

[4] Idaho's dairy industry formed the Independent Dairy Environmental Action League (IDEAL) as a legal defense and environmental research fund. IDEAL's core members are 734 Idaho dairies that ship milk and belong to the Idaho Dairymen's Association (IDA). Also involved are several state and government agencies, Idaho Power, the University of Idaho and the Idaho Conservation League.

Average Schmidt ammonia emissions from 2004 ARB study and 1995 EPA study = (136 + 18)/2 = **77 lbs-NH3/hd-yr**

VFA emission factor = (11.9 + 13.3 + 25.9)/3 = **17.0 lb/head-year**

Constituent 6 – Phenols

Case for Viewpoint 1

At no point during the DPAG process has any information been presented from California dairy studies suggesting that phenols are a significant constituent of dairy VOC emissions. There have been indications in the literature that phenols are a constituent in odors, but quantifications are not provided in the literature. Some DPAG members have suggested that an emission factor for phenols based on Hobbs et al be used. However, Hobbs bases his emission factors for British dairies on the assumption that emission rates for volatile fatty acids and phenols may be calculated based on an assumed relationship between ammonia emissions from manure slurry and emission rates of fatty acids and phenols. The paper establishes no basis for this assumption and in fact provides evidence to the contrary; as Hobbs states in the paper: "we did not find a significant correlation between NMVOC and ammonia emissions for dairy cattle slurry, as the former were too low." Hobbs also provides no information or calculations to support his assertions related to an emission factor, such as the volume of manure treated compared to manure production from each cow. As such, the emission factors for VFAs and phenols within the paper cannot be supported or verified.

Even if the Hobbs calculations were verifiable or his research had supported the ammonia-VOC ratio contention, the conditions under which the research was conducted are not representative of conditions on a California dairy. Only a small fraction of the manure solids on a typical dairy are stored in a slurry condition. The remainder are stored in conditions that are either highly dilute (lagoons/retention ponds) or much dryer solid form (separated solids, manure in corrals).

There are additional data from California studies that directly contradicts the Hobbs emission factors for VFAs and phenols. First, Schmidt et al, reported non-detects of VFAs from a California dairy. Even if it is assumed that actual emissions of acetic acid, the primary VFA, in the Schmidt study is at detection limits, Hobbs predicts an emission rate more than an order of magnitude higher. Similar errors were noted when the Hobbs data were compared to California studies of the poultry industry (Summers letter, April 13, 2005). The data provided by Dr. Mitloehner also demonstrate clearly that the predicted emission rate by Hobbs is in error specifically for phenols. Hobbs predicts a per-cow emission factor of more than 5 pounds per year of phenols. Mitloehner, using the PTRMS methodology, which measures phenols and many other compounds, reported a total emission rate for all compounds of 3.0 including fresh excreta, gaseous emissions directly from the cow, and emissions of feed. The largest constituent was methanol at 1.35 lbs/hd-year, and the second largest constituent acetone/propanol at a combined 0.35 lbs/hd-year. Mitloehner reports that "all remaining detected VOC gases were detected at very low concentrations" and did not total more than 1.5 pounds combined. Simply

put, this strongly suggests that phenols from cows and their fresh waste do not exceed 0.35 lbs and are probably much smaller if detectable and quantifiable at all. Secondly, this clearly demonstrates that the Hobbs emission factors cannot be accurate and in fact represent an error of at least one order of magnitude or more. Since these errors are borne out in several studies and for several constituents, Hobbs simply cannot be considered to represent an accurate emission factor.

There is additional evidence to support the contention that, at least in statistical terms, phenol emissions are close to zero. Powers (2003) reported concentrations of VFAs and phenols from dairies, poultry and swine facilities. Powers reported phenol concentrations to be between 400 and 660 times less than VFA concentrations. It is important to note that the Powers study was not designed to calculate emission rates and as such, these concentrations cannot be used to estimate per head emissions or other emission rates. However, given these ratios of VFA emissions as reported by both Schmidt and Koziel, total phenol emissions can be expected to be in the neighborhood of ~0.001 pounds/head-year.

In the absence of an emission factor specifically for phenols, an interim estimate of zero represents the most responsible recommendation for several reasons. First, given the significant evidence above that the number is well below 0.35 lbs/hd-year and in fact, considering the Powers study, may be one to two orders of magnitude below the VFA emission factors calculated from the Schmidt study, an emission factor of zero represents a much smaller potential error than any other recommendation. Second, there is a strong likelihood the emissions reported by Mitloehner in fact include phenols from fresh waste, feed and enteric emissions, limiting other potential emissions to a few processes. Estimating any other amount of emissions in the absence of measured data from California dairies could not be said to be representative of California dairies. We believe that DPAG has not been presented with any information to date that can credibly be used to calculate emission factors of phenols from any type of livestock in any environment. We see no evidence suggesting that emissions of phenols from dairies are measurable in any statistically significant manner.

Case for Viewpoint 2

Non-enteric phenols were not measured by any test. Total non-enteric VOC emissions are undercounted additionally because there has been no known measurement of phenols, and no estimate of phenol emissions is being attempted for this proposal. It is important to note that phenols are expected to exist in dairy emissions, and that some literature indicates that phenol emissions are among the more significant of VOC emissions from dairies**.**

Case for Viewpoint 3

Non-enteric Phenols were not measured by any of the four California studies, yet they are expected to exist in dairy emissions. Some literature indicates that phenol emissions are among the more significant of VOC emissions from dairies. The recommended phenol factor is based on an average of two studies outside of California that report phenol emissions from dairy manure: Hobbs and McGinn.

Air district staff previously developed a factor of 5.2 lb/head-year for phenols from the Hobbs study. Although several DPAG members have noted discomfort with this number, we recommend using the factor averaged with a phenol estimate from a second study (below) because of the lack of data available and the need to characterize this important emissions component.

The McGinn study reported a phenol concentration (0.003 - 0.434 ug/m3) that was roughly 0.02% of ammonia levels leading to an emission factor of 0.02 lb phenol/head-year. Averaged with the factor above, we recommend a phenol factor of **2.6 lb/head-year.**

Constituent 7, 8 - Land application, feed storage, settling basins, composting, & manure disturbance

Case for Viewpoint 1
Application of manure to croplands should not be included within a dairy emission factor; it is not part of a dairy as defined by the settlement agreement because it is not a confined animal facility within the California Health and Safety Code. Regardless, no information is available to estimate these emissions. In the absence of such information, an emission factor cannot be supported. Because application of manure and manure water is carried out only sporadically, emissions from application to crops are probably insignificant on a year-round basis.

Case for Viewpoint 2
Emissions measurements from these processes are not available.

Case for Viewpoint 3
McGinn (2003) reports incredibly high levels of VFAs as measured over 2-3 days after manure was applied to fields. The total average VFA concentration, 278 ug/m3, was over five times higher than the maximum VFA level (48 $\mu g/m^3$) measured from the overall feedlot operations. It is possible that VFAs from field application of manure from feedlots may be higher than that for dairies – or it may be lower. However, the measurements indicate that land application of manure can generate significant VFA emissions. Because land application is sporadic, we expect VFA emissions to be roughly an order of magnitude lower than that from daily operations. Therefore, a factor of **1 lb/head-year** is appropriate.

Settling basins, composting, manure disturbance (scraping, piling, pile moving, etc.) and feed storage are dairy practices that are not specifically represented in the testing reviewed by the DPAG. An argument can be made that Krauter's testing is whole-dairy testing, and so use of his results would include these sources to the extent they existed at the dairies tested. However, no settling basins or composting operations are known to exist at the sites of Krauter's testing. In addition, the expected emissions of VOCs that will occur when piles of manure are disturbed and moved will only be expected to be measured when the disturbance is ongoing. Further information is necessary to determine whether such activities were occurring at the time of the testing. However, since we know that these emissions are not

zero, we recommend a very conservative emission factor of **0.1 lb/head-year** as a placeholder until more information is available.

IV. Whole-dairy Approach

After substantial discussion, the DPAG agreed that the following table represents an adequate summary of the whole-dairy approach to calculating the major constituents of VOC. Where there is disagreement, alternate methods for calculating an emission factor are reported for each constituent, with notes provided to explain the case for each method.

Constituent	**Emissions (lb/hd-yr) Viewpoint 1**	**Emissions (lb/hd-yr) Viewpoint 2**	**Emissions (lb/hd-yr) Viewpoint 3**
1. Carbonyl Emissions from Corrals (Cassel)	0.5	1.0	1.0
2. Carbonyl Emissions from Lagoon (Cassel)	0.0	0.1	0.1
3. TNMHC Emissions from corrals (Krauter)	3.7	5.3	5.3
4. Methanol Emissions (Mitloehner)	1.4	1.4	1.4
5. Ethylamine Emissions (Schmidt)	0.2	0.2	0.2
6. Volatile Fatty Acids (VFAs)	0.5 (Schmidt)	7.0 (Miller et al)	17.0 (Avg. of several)
7. Other Oxygenates	0	0.1 (Rabaud)	0
8. Other Amines	0	0.3 (Miller et al)	11.0 (Schmidt, Ngwabie, Rabaud)
9. Phenols	0	Included in the amine calculation	2.6 (Hobbs et al, McGinn et al)
10. Land application and Other Sources	0	0	1.1
Total	**6.3**	**15.4**	**39.7**

Specific Differences in Viewpoints for Whole Dairy Approach

Constituents 1, 2, and 3 – Carbonyl Emissions from Corrals and Lagoons (Cassel) and Total Non-methane Hydrocarbon Measurements from Corrals and Lagoons (Krauter)

Case for Viewpoint 1

Viewpoint 1 was based on average of values reported by each researcher. Cassel reported five sampling events at the same Merced County dairy; three in Fall 2003 and two in Summer 2004. In pounds/hd-year, Cassel reported emissions of 0.47, 0.33, 0.16, 0.4, and 1.16. The median of these sampling events is 0.4 and the mean is 0.504, rounded to 0.5 in the table above. Some DPAG members have suggested that in data sets with multiple points, the highest data point should be used as representative of the data set. However, no evidence has been presented to support this contention. It is evident that for any given data set, the mean and median are more representative of the data set than the highest or lowest data point. Purposely choosing the highest data point in multiple data sets only compounds this error.

Similarly to above, Cassel reported five sampling events in Fall 2003 and Summer 2004 at a Merced County dairy. Results were 0.0, 0.02, 0.07, 0.02, and 0.08 lbs/hd-year. The mean of these data is 0.038 and the median is 0.045. In either case, the number is rounded to zero using the convention of rounding to the nearest 1/10 lb.

Using upwind/downwind sampling to capture concentrations, U.S. EPA method TO-15 for analysis and a U.S. EPA-approved model for calculating emission rates from concentrations, Krauter reported data from a total of five sampling events, three of these at a dairy in Kings County and two from a dairy in Merced County. Emission factors reported from the Kings sampling events were 1.5, 2.9 and 2.5 lbs/hd-year, with a mean of 2.3. The Merced events were 5.25 and 5.1, with a mean of 5.175. The mean of these means was 3.74, rounded to 3.7 in the table above.

For similar reasons to those stated above, the means used for calculating an emission factor are more representative of the data set than the use of the highest data point as suggested by some DPAG members. In this case, failure to use a mean also results in completely discounting results from the Kings County sampling events. Again, no data have been presented to DPAG to support the contention that selecting the highest data point is more representative of the data set than selecting a mean or median.

Case for Viewpoints 2 and 3

With the small number of samples collected (3-5 readings), the lower numbers are viewed as possible partial detection events for upwind-downwind sampling approaches and the higher values are used to provide a reasonable estimate of the emissions. The small number of samples does not allow determination of whether this value is the maximum or typical value that would be found from more extensive sampling. For Constituent 3, TNMHC Emissions from corrals (Krauter), the result is based on TO-15 identified compounds, only. This mass does not include

unidentified or tentatively identified additional compounds that may be determined by other analysis or measurement techniques.

Constituents 4 and 5 – Methanol and Ethylamine Emissions

Consensus among all three viewpoints as discussed in the process based approach.

Constituent 6 – VFA

Viewpoints 1 and 3 are as described in **Section III – Process-Based Approach**. The Viewpoint 2 emission factor is based on a comparison of data from Koziel and Miller et al, and included a determination of the potential for liquid VFAs to be emitted into the atmosphere. Volatile fatty acids (VFA) from cattle slurry were measured by Varel et al. This measurement cannot be used directly because much of the VFA contained in a liquid slurry may either not evaporate or be consumed by chemical processes. Therefore, the information from slurry measurements must be correlated to observed measurements of VFA in air. A study by Koziel in Texas provides these data for cattle. The observed ambient VFA and liquid VFA concentrations must also be converted to dairy equivalent emissions. The relationship and conversion are completed by use of additional total alcohol data provided by Varel that can be linked to the VFA measurements in the same report, and Schmidt data on dairy alcohol emissions. The end result of complex calculations identified that roughly ten percent of the slurry VFA at emitted into the air and the quantification of total VFA for fresh and day old manure was calculated as seven pounds per head. Literature on observed downwind VFA concentrations suggests that this estimate could be low and other approaches for estimating this component may be considered.

Constituent 7 – Other Oxygenates

Case for Viewpoint 1

No evidence has been presented to DPAG to suggest that a significant component of oxygenated VOCs were not accounted for. By far the largest oxygenated VOC found in any of the studies was methanol (Mitloehner) and it has been included above). The next largest oxygenated VOC, acetone/propanol, was measured both by Krauter and Mitloehner (although as an exempt compound it is not included in the totals). It should also be noted that acetic acid, an oxygenated VOC and the most prevalent VFA, is included in the table above (in the VFA section) and several other common VFAs include oxygen atoms. Besides acetone and ethanol, the TO-15 method used by Krauter also captures other common oxygenated VOCs, such as ethyl acetate and isopropyl alcohol.

In sum, no sufficient basis has been given for adding additional oxygenated compounds and it is a misstatement to suggest that important oxygenated VOCs expected in cattle emissions are not included in the table above as described here.

Case for Viewpoint 2
A study, performed at the UC Davis dairy, measured atmospheric downwind concentrations of 3-hexanol as well as ethanol. The proportion of other alcohols documented in this study and documented in literature is small in comparison to measured ethanol. Data from the UC Davis study were used with Schmidt calculations of ethanol per head to provide an approximate value of other oxygenates. The consideration of uncertainty for missing oxygenates not measured in other studies should be approximately of this calculated magnitude. The measurement from UC Davis does not preclude that minor amounts of additional alcohols may be detected in different locations and circumstances.

Constituent 8 – Other Amines

Case for Viewpoint 1
Viewpoint 1 is as described in **Section III – Process-Based Approach**.

Case for Viewpoint 2
Amines and Phenols were measured together by Varel et al, on cattle slurry. The ratio between observed amines and phenols and total alcohol measured with the same method was used with Schmidt data on alcohol per head to quantify potential phenol and amine emissions. Literature on observed downwind phenol concentrations suggests that this estimate may be low; however those reports lack sufficient detail to allow per head quantification.

Case for Viewpoint 3
Viewpoint 3 is as described in **Section III – Process-Based Approach**.

Constituent 9 – Phenols and Constituent 10 - Emissions from Land Application and Other Processes

Case for Viewpoint 1 and 3
Viewpoints 1 and 3 are as described in **Section III – Process-Based Approach**.

Case for Viewpoint 2
Viewpoint 2 quantification of phenols is included in the preceding Viewpoint 2 amine calculation.

V. Summary and Conclusions

Both the process-based approach and whole-dairy approach tended to generate similar emissions estimates. DPAG members agree that the process-based approach of section III received significantly more attention and vetting. The largest areas of uncertainty or disagreement for most DPAG members centered on three specific chemical groups – VFAs, amines and phenols. However, DPAG sees the apparent agreement between Section III and Section IV as providing some validation to either approach, so long as the areas of serious disagreement can be resolved in a manner that is based on good science.

DPAG reiterates that any conclusions drawn from this report should be revisited in the near future as additional data become available.

VI. Future research recommendations

DPAG members recommended consideration of the following research topics in the future:

- Standardized methods and a target suite of compounds are required to guide future research work regarding dairy emissions. Specific research should be undertaken to determine which gases should be included from amines, oxygenated VOCs, VFAs, phenols and other potentially important compounds identified in this report, as well as to develop improved sampling, analytical and quantification methods.
- Significant work is needed to better understand the role of emissions from feed versus direct emissions from cows and their waste. Direct emissions from feed appear to be important in overall VOC emissions and evidence was also presented to DPAG suggesting that diet may have an impact on emissions from cows and manure decomposition.
- Additional data are needed on different process emissions and effects of management practices on emissions.

Some DPAG members believe that the ozone-forming potential of livestock-related VOCs must be more thoroughly evaluated. The role of livestock operations in ozone formation needs to be better understood. Other DPAG members disagree with this position.

Appendix A
Definition of VOC, from District Rule1020

3.53 Volatile Organic Compound (VOC): any compound containing at least one (1) atom of carbon except for the following exempt compounds:

- methane,
- ethane,
- carbon monoxide,
- carbon dioxide,
- carbonic acid,
- metallic carbides or carbonates,
- ammonium carbonate,
- acetone,
- methyl acetate,
- methylene chloride,
- methyl chloroform (1,1,1-trichloroethane),
- perchloroethylene (tetrachloroethylene),
- CFC-113 (1,1,2-trichloro-1,2,2-trifluoroethane),
- CFC-11 (trichlorofluoromethane),
- CFC-12 (dichlorodifluoromethane),
- HCFC-22 (chlorodifluoromethane),
- HFC-23 (trifluoromethane),
- CFC-114 (1,2-dichloro 1,1,2,2-tetrafluoroethane),
- CFC-115 (chloropentafluoroethane),
- HCFC-123 (1,1,1-trifluoro 2,2-dichloroethane),
- HCFC-124 (2-chloro-1,1,1,2-tetrafluoroethane),
- HFC-125 (pentafluoroethane),
- HFC-134 (1,1,2,2-tetrafluoroethane),
- HFC-134a (1,1,1,2-tetrafluoroethane),
- HCFC-141b (1,1-dichloro 1-fluoroethane),
- HCFC-142b (1-chloro-1,1 difluoroethane),
- HFC-143a (1,1,1-trifluoroethane),
- HFC-152a (1,1-difluoroethane),
- HCFC-225ca (3,3-dichloro-1,1,1,2,2-pentafluoropropane),
- HCFC-225cb (1,3-dichloro-1,1,2,2,3-pentafluoropropane),
- HFC-43-10mee (1,1,1,2,3,4,4,5,5,5-decafluoropentane),
- HFC-32 (difluoromethane),
- HFC-161 (ethylfluoride),
- HFC-236fa (1,1,1,3,3,3-hexafluoropropane),
- HFC-245ca (1,1,2,2,3-pentafluoropropane),
- HFC-245ea (1,1,2,3,3-pentafluoropropane),
- HFC-245eb (1,1,1,2,3-pentafluoropropane),
- HFC-245fa (1,1,1,3,3-pentafluoropropane),
- HFC-236ea (1,1,1,2,3,3-hexafluoropropane),
- HFC-365mfc (1,1,1,3,3-pentafluorobutane),
- HCFC-31 (chlorofluoromethane),
- HCFC-151a (1-chloro-1-fluoroethane),
- HCFC-123a (1,2-dichloro-1,1,2-trifluoroethane),
- $C_4F_9OCH_3$ (1,1,1,2,2,3,3,4,4-nonafluoro-4-methoxy-butane),
- $(CF_3)_2CFCF_2OCH_3$ (2-(difluoromethoxymethyl)-1,1,1,2,3,3,3-heptafluoropropane),
- $C_4F_9OC_2H_5$ (1-ethoxy-1,1,2,2,3,3,4,4,4-nonafluorobutane),
- $(CF_3)_2CFCF_2OC_2H_5$ (2-(ethoxydifluoromethyl)-1,1,1,2,3,3,3-heptafluoropropane),
- cyclic, branched, or linear completely methylated siloxane compounds,*
- parachlorobenzotrifluoride, and
- the following four classes of perfluorocarbon (PFC) compounds:*

1) cyclic, branched, or linear, completely fluorinated alkanes,
2) cyclic, branched, or linear, completely fluorinated ethers with no unsaturations,
3) cyclic, branched, or linear, completely fluorinated tertiary amines with no unsaturations, and
4) sulfur containing perfluorocarbons with no unsaturations and with sulfur bonds only to carbon and fluorine.

*perfluorocarbon and methylated siloxane compounds shall be assumed to be absent from a product or process unless a manufacturer or facility operator identifies the specific individual compounds (from broad classes of these compounds) and the amounts present in the product or process, and identifies a validated test method which can be used to quantify the specific compounds.

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