

**Title:** Characterization of VOCs and particulates from swine finishing facilities and relationship of these compounds to human health, **NPB #02-148**

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**Abstract:** Emissions from animal feeding operations have been a prominent topic in Iowa for a number of years, particularly during 2002. The issue gained momentum at the federal level during that year as well, resulting in the release of a National Research Council report "Air Emissions from Animal Feeding Operations: Current Knowledge, Future Needs" in December 2002. To address the issue a study was initiated to determine 1) if phase of production influenced air composition and odor at the facility, 2) if operations of the same type and size differ in concentrations of the compounds of interest, 3) how concentrations of compounds change as they move downwind of a given site, and 4) how climatic factors influence the concentrations downwind. Air samples were collected or analyzed onsite twice weekly at each site for a 10-week period between May 2002 and August 2002 from two breeding and gestation facilities utilizing aerated earthen storage structures and from five deep-pit finishing facilities. Sites within a given production phase (sow or finishing facility) were matched for size of operation. Samples were collected immediately outside of a building or on the berm of the manure storage structure and at points downwind of the location (approximately 50, 100, and 200 m). Air was analyzed for hydrogen sulfide content (onsite), odor (collected samples in 10-L Tedlar bags), and composition (volatile fatty acids, phenols, indoles, alkanes in collected samples). During sampling, temperature, windspeed and direction, humidity and solar cover were recorded. Results indicate that differences between the sow sites and the finishing sites were limited to differences in butyric acid concentration, only. Site within a specific phase of production contributed to differences in concentrations of particulates, odor, hydrogen sulfide, butyric and valeric acids and all of the phenolic compounds quantified. The data suggest that the type of swine system had little effect on the concentrations of most of the monitored compounds as well as odor. However, the management practices of the site itself contributes to differences in analyte concentrations to a much greater extent than production phase differences (breeding and gestation versus finishing). Equations to develop downwind concentrations of all measured compounds were developed. The equations take into account temperature, humidity, and solar cover and are based on the concentrations at the source (ie., building or berm) that were observed in this study. Based on the data collected and the conditions represented in the current study, estimated hydrogen sulfide concentrations would be below 15 ppb at some point beyond 50 m downwind of the source. This estimate is using the statistically-adjusted average source concentration observed for all sites and all sampling events. Variation in source concentration did occur, however so on any given sampling day at any given site it is possible that 15 ppb would have been measured within 50 m of the source. Therefore, caution must be executed to avoid over-interpreting the results from this study.

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**Introduction:** Attention towards gaseous emissions from poultry and livestock facilities continues to be a prevalent issue for the industry. These emissions, once considered a nuisance concern, are now scrutinized as a public health threat. Odors have been identified by those affected as a contributor to deterioration of health, causing nausea, dizziness, eye, nose, and throat irritation, shortness of breath, stress, and drowsiness (Schiffman et al., 1998). Research indicates that while some of these effects may be perceived (Knasko, 1993) many of the responses are real physiological reactions to exposure to odorants (Donham, 1996). While the physiological effect of odor may be in question, there is no doubt that odors stemming from livestock production can be a nuisance. Many state governments have sought regulatory solutions to odor concerns. Most regulations employ either a single chemical parameter for odor indication (i.e., Minnesota has a hydrogen sulfide standard) or an olfactory assessment (i.e., Colorado uses a Scentometer, a handheld instrument used for onsite observation, manufactured by Barneby and Chaney Corp.). At the federal level, ambient air quality standards will likely be revised in the near future to address particulates and ammonia, in particular, as primary (health-based) pollutants. Some states are considering health-based standards for other pollutants as well. In February 2002, the University of Iowa and Iowa State University released a report on human health impacts from CAFOs. The EPA commissioned the National Academy of Science to develop a similar report at the national level that was released in December 2002 (National Research Council, 2002). The challenge in considering the development of standards for odor and other gases is the paucity of data that exists documenting not only 'typical' concentrations in the community and at the property line, but the health effects of long- and short-term exposures of these concentrations.

Limited work has been conducted that thoroughly characterizes the composition of air collected at or near animal production facilities (Zahn et al., 1997; Gralapp et al., 2001). Even more restricted is work that goes on to quantify a substantial number of identified compounds from these samples at multiple locations within and beyond the facility borders. Thorough investigation of the compounds that result in these differences will provide a better understanding of the compounds that must be controlled to avoid nuisance conditions.

### **Objectives**

As regulatory continues to be discussed at state and federal levels, adequate characterization of gaseous emissions (concentrations and components) is needed. Additionally, methods to evaluate emissions, for compliance purposes, are necessary. The specific objectives of the proposed research were to:

- *Characterize particulate and gaseous concentrations in and around both sow and finishing swine production facilities, and*
- *Study the downwind movement of the emissions.*

### **Procedures**

Air samples were collected from finishing and gestation operations twice weekly for a 10-wk period, between May 6, 2002 and August 5, 2002. Not all operations were initiated concurrently in order to accommodate pig flows. Two commercial breeding and gestation operations (S) with lagoon systems (L) and five commercial deep-pit finishing operations (F) served as collection sites. All sites were located in the state of Iowa.

Air samples for gas chromatography-mass spectrometry (GC-MS) analysis, using an Agilent 6890 gas chromatograph coupled to an Agilent 5973 mass spectrometer, were collected from each site visit via adsorption onto solid phase microextraction (SPME) fibers (Supelco, Inc.; Bellefonte, PA). The SPME fibers were allowed to adsorb air samples for a 10-min period. Following collection, SPME fibers were transported in a cooler back to Iowa State University for analysis. Gas chromatography allowed for quantification of specific chemical compounds identified in the collected samples. All compounds that could be quantified using the standard procedures of the laboratory are listed in Table 1. The list represents compounds routinely identified in similar samples collected over the past four years by the laboratory. In addition, the data report generated from each sample was scanned to determine if additional compounds were routinely identified in an effort to expand the number of compounds quantified.

**Table 1.** Chemical compounds quantified in collected samples using gas chromatography-mass spectrometry.

Acetic acid	Decane	Carbon disulfide
Propanoic acid	Undecane	Dimethyl disulfide
<i>isobutyric</i>	Dodecane	
Butanoic	Pentane	
3-methylbutanoic	Nonane	
Pentanoic	Tridecane	Ethanethiol
	Tetradecane	Propanethiol
Phenol		Butanethiol
3-methylphenol	Indole	
4-methylphenol	2-methylindole	1-decene
2-ethylphenol	3-methylindole	Nonanal
3-ethylphenol	4-methylindole	Gamma-Butyrolactone
4-ethylphenol		
2,6- <i>bis</i> (1,1-dimethylethyl)phenol		

Samples were collected in 10-L Tedlar bags for transport to the olfactometry laboratory at Iowa State University. Human assessment of the samples was conducted using an Ascent olfactometer (St. Croix Sensory, Stillwater, MN) and eight trained panelists. Panelists determine the odor dilution threshold for each sample; a higher value indicating a stronger odor. Further description of the procedures used by that laboratory are described elsewhere (Gralapp et al., 2001). A Jerome meter was used to collect onsite measures of hydrogen sulfide concentrations. This instrument has a detection range of 0.003 – 50ppm with a relative standard deviation of 5%. Particulates greater than 4 micron (PM<sub>4</sub>) were collected throughout the sampling period at two finishing sites using an air sampling pump and particulate filters (SKC, Inc.). A 20-min sampling time was used with a flow rate of 3 L/min. Change in mass of the particulate filters represents mass of particulates collected over the sampling period.

Samples were collected from a source (0 m from a building) and from points downwind of the source (approximately 50, 100, and 200m). Actual distance from the building was recorded for each collection point. At the two breeding and gestation sites, samples were collected at the lagoon berm and downwind points of the lagoon as well. Wind direction was identified and sampling points determined accordingly on each day of sampling such that all measurements were always collected downwind of the source. On each sampling day solar cover was characterized (i.e. sunny, partly sunny, cloudy, raining) and temperature, humidity, and wind speed data were recorded at each site.

All data were analyzed by procedures of the SAS statistical package. A general linear model was used to determine the fixed effects of production phase, site within production phase, and source (lagoon or building) on measured variables. Distance from the source served as a continuous independent variable as both a linear and a quadratic term. Wind direction, wind speed, and solar cover served as covariates in models assessing outdoor measures. Stepwise linear regression was used to develop an odor prediction equation from analytes quantified by GC-MS.

**Results:** Chemical constituents, particulates, and odor concentration of sampled air

Least square means of all measurements taken immediately outside of the building monitored for each study site, pooled across project years, are depicted in table 2. Numerical differences in table 2 that may appear to be different, reflect large variation in measurements that occurred from one sampling day to the next for any given site. Because, samples were always collected in a downwind direction of the building source, wind direction was not a significant term. Windspeed only affected particulate, phenol, decane, and undecane concentrations ( $P > 0.05$ ). Degree of solar cover did have an effect on concentrations of hydrogen sulfide and decane as well as odor dilution threshold with greater concentrations on sunny days compared to overcast or rainy days in all cases. Probability levels of production phase and site within production phase effects on concentrations of measured chemical constituents are presented in table 3. Production phase effects were

observed for butyrate concentration, only suggesting that the type of swine system had little effect on the concentrations of most of the monitored compounds as well as odor. Farm site within a given production phase effects were observed for a number of measurements indicating that management within a given operation contributes to differences in analyte concentrations to a much greater extent than production phase differences (breeding and gestation versus finishing) between locations. (table 3).

Fewer compounds were observed in samples collected from the lagoon berms of the S sites and analyzed by GC-MS than in samples collected from the buildings of both the S and F sites (table 4 compared to table 2). The presence of fewer compounds at detectable levels may have been the result of biological processing of manure that took place in the lagoons. The lagoons did have aerators on them which likely contributed to undetectable concentrations of the analytes of interest; most of which are the result of anaerobic processes. It is notable that concentrations of some compounds were greater at the lagoon berm than at the building (table 2 compared to table 4) and hydrogen sulfide concentrations, in particular, appear reversed between the two sites. However, the implications of this finding may be unimportant when one considers that proposed regulatory rules in Iowa consider separation distance measures, making the source of emission within a farm indiscernible. The value in knowing what sources contribute to the emissions is in developing suitable controls for the offending portions of an operation.

**Table 2.** Least squares means of measures collected from locations immediately outside of each study site building (0 m).

Production phase		Finish	Finish	Finish	Finish	Finish	Sow	Sow
	Site	A	B	C	D	E	A	B
Measure	Units							
Particulates	mg/L	1.63	3.16					
Odor Dilution	ou1	275	548	392	226	277	326	241
H2S	ppb	73	407	6	58	125	133	83
Acetate	ppm	83.593	88.765	73.767	69.908	79.902	30.290	84.346
Propionate	ppm	3.700	5.763	0.000	0.000	0.527	0.000	0.273
Butyrate	ppm	3.775	2.658	0.000	0.966	0.623	0.374	0.201
Valerate	ppm	0.776	0.169	0.000	0.037	0.000	0.024	0.000
Phenol	ppm	0.647	1.343	0.321	0.331	0.397	0.205	0.329
4-Methylphenol	ppm	0.296	0.859	0.000	0.082	0.076	0.095	0.190
4-Ethylphenol	ppm	0.000	0.042	0.000	0.000	0.000	0.013	0.003
3-Methylindole	ppm	0.004	0.016	0.000	0.000	0.000	0.010	0.001
g-Butyrolactone	ppm	8.182	28.453	213.584	14.881	10.486	144.371	7.033
Nonanal	ppm	17.292	3.072	0.546	7.814	6.534	6.045	5.931
Decane	ppm	2.952	0.718	1.552	0.876	2.334	0.369	0.763
Undecane	ppm	4.885	2.187	7.851	2.912	2.976	2.345	3.122
Dodecane	ppm	3.409	2.063	15.814	3.358	1.437	3.497	3.816
Tridecane	ppm	4.036	8.032	2.403	2.716	0.850	4.601	1.506
Tetradecane	ppm	6.918	2.010	0.923	2.109	1.914	4.866	1.657
1-Decene	ppm	0.000	0.085	0.000	0.010	0.000	0.000	0.000

1ou – odor units, representing the number of dilutions with odor free air needed for the odor sample to be barely detected by 50% of a human panel

**Table 3.** Probability levels for production phase and site within phase (farm) effects of concentrations of chemical constituents, odor, and particulates immediately outside of the building at each study site.

Measure	Production phase	Farm
Particulates		0.0272
Odor dilution threshold	0.2820	0.0007
H2S	0.3972	<0.0001
Acetate	0.3102	0.7326
Propionate	0.3216	0.3679
Butyrate	0.0393	0.0115
Valerate	0.0855	<0.0001
Phenol	0.0912	0.0226
4-Methylphenol	0.1512	<0.0001
4-Ethylphenol	0.9258	0.0017
3-Methylindole	0.6625	0.0052
<i>g</i> -Butyrolactone	0.6063	0.0598
Nonanal	0.4018	0.8950
Decane	0.7386	0.0742
Undecane	0.5344	0.8918
Dodecane	0.4832	0.0990
Tridecane	0.8529	0.7091
Tetradecane	0.7634	0.1942
1-Decene	0.4280	0.6393

**Table 4.** Least squares means of measures collected from the berm of the downwind side of lagoons at the breeding and gestation sites (0 m) and probability value (P-value) of farm effect.

Measure	Units	Sow A	Sow B	P-value
H2S	ppb	69	441	0.0136
Acetate	ppm	33.937	33.863	0.9980
Phenol	ppm	0.088	0.086	0.9840
<i>g</i> -Butyrolactone	ppm	11.040	83.225	0.1373
Nonanal	ppm	3.977	6.694	0.5037
Decane	ppm	0.469	0.401	0.9131
Undecane	ppm	3.265	1.431	0.5848
Dodecane	ppm	3.302	2.354	0.7929
Tridecane	ppm	0.529	1.379	0.4611
Tetradecane	ppm	0.464	1.316	0.3736

**Table 5.** Coefficients for developing equations to estimate analyte concentration downwind, based on measures collected at the source. Absence of a value indicates non-significance for that term.

Measure	Units	Intercept	Linear coefficient	Quadratic coefficient	Temperature, °F	Humidity, %
Odor Dilution Threshold	ou <sup>1</sup>	656	-1.10635		-3.9987182	0.1668352
H <sub>2</sub> S	ppb	112	-2.56388	0.0098889	-0.13099100	0.8510286
Acetate	ppm	419.995	-0.34967		-3.91366930	-1.1873090
Propionate	ppm	3.95739	-0.02261	0.000091813	-0.03631482	0.001714060
Butyrate	ppm	1.44123	-0.01836	0.000071810	-0.00496169	-0.001982327
Valerate	ppm	0.25623	-0.00211	0.0000081551	-0.00164762	-0.000620717
Phenol	ppm	2.95560	-0.00486	0.000021498	-0.02859648	-0.007071304
4-Methylphenol	ppm	0.25721	-0.00341	0.0000136681	-0.00133425	0.0006476934
4-Ethylphenol	ppm	0.01571	-0.00013	0.0000005094	-0.00014149	0.0000369593
3-Methylindole	ppm	0.00580	-0.00006	0.0000002597	-0.00004620	0.0000242921
g-Butyrolactone	ppm	753.28542	0.69379		-7.62285320	-2.3482686
Nonanal	ppm	64.72466	-0.00319		-0.56247386	-0.26884622
Decane	ppm	25.782843	-0.00454		-0.26780021	-0.07883275
Undecane	ppm	70.71587	0.00631		-0.71663478	-0.21643365
Dodecane	ppm	70.68739	-0.00931		-0.71365341	-0.19735578
Tridecane	ppm	34.93069	-0.05780	0.00040804	-0.36890001	-0.06064195
Tetradecane	ppm	18.45792	-0.03679	0.0020897	-0.18169013	-0.03169910
1-Decene	ppm	0.21667	-0.00101	0.0000076030	-0.00234459	-0.000339696

1ou – odor units, representing the number of dilutions with odor free air needed for the odor sample to be barely detected by 50% of a human panel

*Estimating downwind concentrations of chemical constituents and odor concentration*

Measurements collected at distances downwind from a building location at each site were analyzed as continuous independent variables to determine degradation curves for each measure as it diluted with distance (linear, quadratic). Coefficients for significant terms of each measurement (linear and quadratic or linear, only) as well as coefficients for temperature and humidity are depicted in table 5. Because specie effects were observed for only a limited number of measures, species was not included in the model. Table 6 depicts constants specific to solar cover on a particular day. Using coefficients in tables 5 and 6, concentrations downwind of a facility on a cloudy day are estimated as follows:

$$\text{H}_2\text{S, ppb} = 149.62229 - 2.49845 (\text{distance from building, m}) + 0.0094062 (\text{distance from building, m})^2 - 0.13099100(\text{temperature, } ^\circ\text{F}) + 0.8510286(\text{humidity, \%}) + 9.5641211$$

**Table 6.** Solar cover coefficients for developing equations to estimate analyte concentration downwind, based on measures collected at the source and relative to sunny conditions. Sunny conditions require no coefficient.

Measure	Units	Cloudy	Partly cloudy	Partly sunny	Raining
Odor Dilution Threshold	ou <sup>1</sup>	-147.8426667	-58.4368556	-31.3171719	-127.9837593
H <sub>2</sub> S	ppb	9.5641211	0.9288899	-31.0552121	9.4790280
Acetate	ppm	3.1410.35	25.6754980	15.9926059	7.5135130
Propionate	ppm	-0.166340335	-0.347160675	-0.455046168	-0.395996499
Butyrate	ppm	0.070129332	0.034749690	-0.217640917	-0.111723934
Valerate	ppm	0.0398011601	0.0404410482	-0.0148931833	-0.0198846724
Phenol	ppm	-0.003533083	0.248595232	0.161112042	-0.186619163
4-Methylphenol	ppm	-0.0536428954	-0.0146380448	-0.0471651559	-0.0646054647
4-Ethylphenol	ppm	-0.0040857710	0.0013844295	-0.0017474233	-0.0047029781
3-Methylindole	ppm	-0.0017822801	0.0005572329	-0.0011704097	-0.0024720606
<i>g</i> -Butyrolactone	ppm	-24.5127710	29.2337830	60.7123596	-43.2201329
Nonanal	ppm	2.22263851	2.46485896	3.50172505	-1.24992735
Decane	ppm	0.26718729	0.68106850	2.11667484	-0.85587882
Undecane	ppm	-0.95817085	1.36655607	0.91900923	-4.36131449
Dodecane	ppm	-2.59421022	1.07701245	-1.17807461	-5.17303754
Tridecane	ppm	-2.54372247	0.86408234	2.68491136	-3.53627914
Tetradecane	ppm	-1.18511211	2.17011458	0.46069125	-2.04675920
1-Decene	ppm	-0.0162866660	0.0463084686	0.0174017808	-0.0210519951

Using the coefficients in tables 5 and 6, estimates of H<sub>2</sub>S concentration (ppb) were developed for a variety of scenarios (table 7). Note that at the source, on an 75° F day with 80% humidity and predominantly clouds, the concentration is 217 ppb at the building. This value represents the least squares mean of the data collected during this project for similar conditions. Actual values deviating from this would have a profound effect on the estimated downwind concentration as well.

**Table 7.** Calculated downwind H<sub>2</sub>S concentrations for various climatic and distance scenarios based on coefficients in tables 5 and 6.

Distance, m	Temperature, °F	Humidity, %	Solar cover	Calculated H <sub>2</sub> S, ppb
0	75	80	Cloudy	217
50	75	80	Cloudy	95
50	75	80	Sunny	85
50	75	40	Sunny	51
100	75	80	Cloudy	No detect
100	45	80	Cloudy	No detect
100	45	40	Cloudy	No detect

*Development of an odor prediction equation from chemical constituents*

Using the analytes quantified by GC-MS, an odor prediction equation was developed using stepwise regression procedures. Each analyte was included in the prediction equation as a quartic term. Non-significant terms were reduced to a cubic term, then to a quadratic term. All variables remained in the final equation as at least linear terms. The equation accounted for 32% of the variation in response observed (R<sup>2</sup> = 0.32). Removal of outliers from the data set resulted in no improvement in predictive capability. Simple correlations between odor measurements and individual analytes measured by GC-MS and H<sub>2</sub>S demonstrated that H<sub>2</sub>S was best correlated (r = 0.34) followed by 4-methylphenol (r = 0.33), 4-ethylphenol (r = 0.24), butyric acid (r =

0.23), propionate and 3-methylindole ( $r = 0.17$ , each). All other analytes had correlation coefficients  $< 0.16$ . All analyte concentrations are expressed as parts per million (ppm).

$$\begin{aligned} \text{Odor dilution threshold} = & 244 - 3.0(\text{acetate}) + 0.003(\text{acetate})^2 - 37.5(\text{propionate}) + 0.072(\text{propionate})^2 + \\ & 203.6(\text{butyrate}) - 49.83(\text{butyrate})^2 + 2.22(\text{butyrate})^3 - 622.97(\text{valerate}) + 339.92(\text{valerate})^2 + 312.10(\text{phenol}) \\ & - 162.09(\text{phenol})^2 + 17.18(\text{phenol})^3 + 77.86(4\text{-methylphenol}) + 39.69(4\text{-methylphenol})^2 + 7962.34(4\text{-} \\ & \text{ethylphenol}) - 126949.7(4\text{-ethylphenol})^2 + 265438(4\text{-ethylphenol})^3 + 73401(3\text{-methylindole}) - 2678775(3\text{-} \\ & \text{methylindole})^2 + 24789329(3\text{-methylindole})^3 + 0.41(\text{gamma-butyrolactone}) - 21.49(\text{decane}) - 0.31(\text{decane})^2 \\ & + 1.81(\text{nonanal}) - 0.13(\text{nonanal})^2 - 8.67(\text{undecane}) - 0.67(\text{undecane})^2 + 27.36(\text{dodecane}) - 1.69(\text{dodecane})^2 \\ & + 0.02(\text{dodecane})^3 + 171.1(\text{nonane}) - 21.43(\text{nonane})^2 - 0.16(\text{nonane})^3 + 733.76(1\text{-decene}) - 267.8(1\text{-} \\ & \text{decene})^2 - 50.38(1\text{-decene})^3 - 0.57(\text{tridecane}) - 0.22(\text{tridecane})^2 + 0.33(\text{tetradecane}) + 0.017(\text{tetradecane})^2 \\ & - 0.01(\text{tetradecane})^3 \end{aligned}$$

Development of an odor prediction equation that considers the interactive effects of the compounds analyzed by GC-MS is also planned for the peer-reviewed manuscript that will result from this work.

### Summary

Results show that management of a swine operation has greater influence on concentration of emitted gases than does type of production at the unit. For example, when comparing deep-pit finishing facilities to breeding and gestation operations with outside manure storage average odor dilution threshold values were similar for the two types of systems. However, within the five finishing sites studied, odor dilution values, represented as a statistical mean across the entire season's sampling, varied considerably. Large standard deviations in measurements from one sampling day to the next, make it difficult to predict concentrations on any given day. However, prediction equations were developed to estimate downwind concentrations based on the climatic conditions of the day and distance from the source. The current data does not, however, consider the initial source concentration. Rather, the equations are based on the actual concentrations observed in the study. The data has been contributed to a much larger data set with the purpose of validating an emissions prediction model. The model has been developed with the intended use as a siting tool or to evaluate the impact of mitigation techniques on pollutant concentrations observed at residences downwind of a livestock operation and does consider the source concentration as a factor in determining downwind concentrations.

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