Response to reviewers' comments

Reviewer #1:

Greetings, I have enjoyed reading this manuscript, and comparing it to previous publications in the field. Although the aircraft data were regional, the team has been successful discerning the CAFO emissions/VOCs from other sources. And although the road-passage measurements were only conducted once per site, good weather conditions were found (or selected!) and the results convincing. It is nice to see general agreement from the two approaches. There are several places in the ms where I would suggest careful re-editing to correct minor mistakes, grammar, and/or clarify. Some examples follow:

Reply: We would like to thank the reviewer for his/her work and nice comments. Please find the response to individual comments below.

line 28 'contributions to in the odor'

Reply: Corrected.

line 94 should clarify that measurements were only once per CAFO

Reply: Changed as the reviewer's suggestion. Please note that we performed downwind measurements twice for the dairy farm #2, beef feed yard #2 and the chicken house. The duplicated measurements usually agreed well. The last sentence of the paragraph is changed to: *We added a new VOC instrument to the payload, and performed mobile measurements in winter time (February, 2016) for the six CAFOs by sampling at their downwind flanks 1-2 times for each facility. Duplicated measurements at the same facilities agreed well.*

line 97 is a run-on sentence

Reply: Corrected. The sentence is changed to "*Here, we provide a brief description of the instrument (see details in Yuan et al., 2016).*"

line 128 provide formulas for chemicals at first mention

Reply: Chemical formulas are provided in the revised manuscript.

line 128 'around' is imprecise; 'along the downwind flanks' might be better

Reply: We decide to keep using “around”, as measurements were conducted for the upwind flanks of the facility as well.

line 137 since there was no access to the facilities, ’waste may be largely’ and near here it should be stated that ’waste cleaning time/practices are unknown’
Reply: Changed according to the comment.

line 147 ’spatial distribution based on the variation observed (over a short time span of several minutes) while measuring along the downwind flanks’ [I know this is ’wordy’, but time variations implies watching a source as it varies over time (e.g. in the course of a day, from day to day, season to season, etc...)
Reply: Changed as the suggestion from the reviewer.

line 152 (although not known specifically here)
Reply: The statement is added in the revised manuscript.

line 166 needs a reference
Reply: A webpage is cited in the revised manuscript.

line 228 state estimated dilution(s), based on distance, and how much it varies across these data sets
Reply: We add a short sentence in the end of the paragraph: “(see example in section 3.4 for aircraft measurement results associated with a factor of ~10 lower concentrations than those from mobile laboratory)”.

lines 263-4 plus or minus how much
Reply: The uncertainties from the multi-variate fits are shown in the revised manuscript.

line 269 provide wind speed means
Reply: The means of measured wind speeds are provided in this sentence.

line 291 separating
Reply: Corrected.

line 316 prior to sampling

Reply: Corrected.

line 321 'strong' should be enumerated

Reply: Correlation coefficients are provided in the revised manuscript.

lines 331-2 list numbers of each, as best as is known

Reply: The numbers for each flight are included in the revised manuscript.

lines 341-4 grammar hard to follow long sentence

Reply: The long sentence is divided into two sentences in the revised manuscript.

line 346 'measured downwind air to discern VOC...'

Reply: Changed in the revised manuscript.

Reviewer #2

In this study, Yuan and his colleagues measured the emissions of different VOCs, along with methane and ammonia, from CAFOs of northeastern Colorado through two methods: mobile laboratory and aircraft measurement. The high time-resolution measurements allowed the authors to identify different sources of VOCs within the CAFOs. They found ethanol was mainly from feed storage and handling while NH3 predominately came from animals and their waste. A multivariate regression analysis was conducted to determine the relative importance of these two sources for different VOC species. The contributions of different VOCs to odor activity, NO3 reactivity, and OH reactivity were also investigated. The study was interesting and the result was well presented. However, the reviewer would recommend the authors make the following edits before the manuscript can be reconsidered for publication:

Reply: We would like to thank the reviewer for his/her work and helpful comments. Please find the response to individual comments below.

Line 33-34 "emissions of ethanol concentrations". Concentrations can't be emitted. I would recommend changing it to "concentration of emitted ethanol" or "concentrations of ethanol emissions".

Reply: We change to “emissions of ethanol” in the revised manuscript.

Line 85-86 Should be “within the facilities that contribute to VOC emissions”.
Reply: Corrected.

Figure 2 For the upper left figure, the number labeling of the time of measurement (1,2,3,4) should be on the same side of the road for easy reading. On the current figure, 1 was put on one side of the road, while 2 and 3 were put on the other. I had a hard time finding the location of time point 1 while first reading the manuscript.
Reply: The graph has been changed according to the reviewer’s suggestion.

Line 226-228 The authors stated that “We note that OAV, OH and NO3 reactivity are measured along the fence line and they decrease rapidly with downwind distance and dilution.” If the authors have data to support such a statement, please put them in the manuscript. Otherwise, please indicate “(data not shown)” at the end of the sentence.
Reply: Based on the comments from both reviewers, we add a short sentence in the end of the paragraph: “*(see example in section 3.4 for aircraft measurement results associated with a factor of ~10 lower concentrations) than those from mobile laboratory*”.

Line 234 Is it possible to separate the VOCs from animal exhalation and animal waste?
Reply: The emissions from animal exhalation and animal waste should be able to be separated from measurements performed for a longer time period (e.g. days or weeks) in a CAFO facility, using potential different time variations of the two emissions (e.g. waste removal practices), as demonstrated in the study of Sintermann et al. (2014). However, this type of data is not available in this study. We added a sentence here to reflect this discussion: “*It is worth mentioning that long-term measurements in CAFO facilities could permit separation of the two co-located sources (see example in Sintermann et al., 2014).*”

Figure 4 On the top graph, there seem to be pretty high discrepancies on the sources of VOCs for the two dairy farms, especially for carboxylic acids, alcohols, and carbonyls. Are there any explanations for that?

Reply: We thank the reviewer for the comment. We notice that there are differences in the relative fractions of the sources for some VOCs between the two dairy farms, notably carboxylic acids, methanol and carbonyls (Figure 4). We do not know the exact reason for this.

There are some further observations that may help to understand this: (1) We did not observe an emission plume that can be clearly attributed to emissions from the milking parlor for the dairy farm #1. The emissions from the milking parlor may be well-mixed with those from feed before detection. Therefore, some of the fractions of feed emissions for the dairy farm #1 in Figure 4 may be contributed by emissions from milking parlor. Considering this, the differences for carbonyls would be not large, if the feed fractions in the dairy farm #1 with feed+milk parlor for the dairy farm #2 are compared. (2) We also observed some differences in the site-averaged VOC compositions (Figure 3A) and the determined VOC compositions of source emissions (Figure 5) between the two dairy farms. The differences in the determined VOC compositions from feed emissions imply that ingredients in cattle feed may be also different in the two dairy farms.

With the above discussion in consideration, we added a sentence in the fourth paragraph of section 3.1 in the revised manuscript:

It is worth noting that we did not distinctly observe emissions from the milking parlor in the dairy farm #1, which were potentially mixed with emissions from feed prior to sampling.

We also added a sentence in the last but one paragraph of section 3.3 in the revised manuscript: *The agreements between the two dairy farms are not as good as for the two beef feed yards. The above mentioned observations of the emissions from milking parlors and potential differences in feed ingredients for dairy cattle, which are reflected by the discrepancies in VOC compositions emitted from feed storage+handling (Figure 5A), could be the reasons.*

Ling 265-268 The authors claimed that the reason for feed+handling played a major role in VOC emission of VOCs in Chicken farms was because "Chickens were raised in production houses and emissions of NH3 and VOCs may be treated when in-house air was ventilated out". Are there any supporting evidences for such a claim, like a detailed layout of the chicken farm to prove that such treatment does exist?

Reply: As mentioned in the manuscript, we had no access to the facility. The satellite images of the chicken house from Google Earth are not clear enough to locate potential odor mitigation

installations. Thus, we do not have evidence to prove that pollutant treatment does exist in this facility. However, we found through a news report that the chicken house uses a manure belt system to manage manure. For manure belt systems, chicken waste is frequently transported to a separated location for storage. The emissions of NH_3 from the chicken houses with manure belt systems were reported to be much lower than facility without manure belt system (Wood et al., 2015). We changed this paragraph to reflect this new information:

Based on a news report on the facility, a manure belt system is used to manage manure in this facility. The manure belt system catches the excreta from chicken to transport manure to a separated location for storage. The chicken houses with manure belt system usually lead to substantially lower emissions (e.g. NH_3) from animal waste (Wood et al., 2015). It is consistent with significantly lower NH_3 concentrations (0-175 ppb, Figure S3) at this site compared to other feed yards measured in this study (0-1000 ppb), although wind speed was 36%-60% higher during measurements of the chicken house (7.5 m/s) than others (4.7-5.5 m/s). It is also possible that emissions of NH_3 and VOCs were treated when in-house air was ventilated out (Wang et al., 2010).

Line 324-325 The authors stated that "the enhancement ratios of acetic acid to NH3 may be used as an indicator for emissions from different animal types" based on the observation on known beef feed yards and dairy farms. Have the authors tested this hypothesis on other CAFOs in the region that were covered by the aircraft measurement to support this claim, if the locations of other CAFOs in the region were known?

Reply: Identification of the plumes from individual facilities was difficult from aircraft measurements during SONGNEX, as the emissions have been mixed prior to sampling. We can only attribute three plumes to one single facility with known animal type, which happened to be the beef cattle feed yard #2 sampled by mobile laboratory as well. Note that the beef cattle feed yard #2 is one of the largest CAFO facilities in the area (Table S1). The enhancement ratio of acetic acid to NH_3 (33.2×10^{-3} ppb/ppb) derived from the three plumes agreed very well with that from mobile measurements (32.5×10^{-3} ppb/ppb). Unfortunately, we do not have information for other CAFO facilities in this region to validate our statement.

References:

Sintermann, J., Schallhart, S., Kajos, M., Jocher, M., Bracher, A., Münger, A., Johnson, D., Neftel, A., and Ruuskanen, T.: Trimethylamine emissions in animal husbandry, Biogeosciences, 11, 5073-5085, 10.5194/bg-11-5073-2014, 2014.

Wang, L., Oviedo-Rondón, E. O., Small, J., Liu, Z., Sheldon, B. W., Havenstein, G. B., and Williams, C. M.: Farm-Scale Evaluation of Ozonation for Mitigating Ammonia Concentrations in Broiler Houses, Journal of the Air & Waste Management Association, 60, 789-796, 10.3155/1047-3289.60.7.789, 2010.

Wood, D., Cowherd, S., and Van Heyst, B.: A summary of ammonia emission factors and quality criteria for commercial poultry production in North America, Atmospheric Environment, 115, 236-245, 10.1016/j.atmosenv.2015.05.069, 2015.