

## ***Interactive comment on “Trimethylamine emissions in animal husbandry” by J. Sintermann et al.***

**J. Sintermann et al.**

joerg.sintermann@art.admin.ch

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reviewer: This manuscript was a pleasure to read. It reports a well-designed experiment, to investigate a question nobody had looked at before, namely: where exactly and how originate the TMA emissions that had previously been found to be associated with the presence of cattle. The authors show an impressively broad command of the TMA-related literature, truly interdisciplinary (spanning from animal physiology to atmospheric chemistry), and the manuscript is thus excellently-placed within the scope of Biogeosciences. It is also well-organized and thereby manages to give the reader an excellent overview of the topic and its significance. The following list of suggestions is of minor editorial nature.

- authors: We are very happy about the detailed and positive assessment of the present  
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manuscript, and we would like to thank the reviewer for his valuable contribution. This was indeed a study touching a broad range of aspects of interdisciplinary topics. In the following we address the reviewer comments in detail.

reviewer: p.6521 last paragraph: details such as “Federal Research Station”, the location and the long parentheses with instrument names should not be in Introduction. They appear in Methods section anyway.

- authors: agreed; we leave out this paragraph since its content is given in the method section. The introduction now ends: “The focus lies on TMA emission pathways in dairy systems, investigated by ambient trace gas concentration as well as laboratory dynamic chamber measurements. We further discuss the fate of the identified agricultural TMA emissions and their role in secondary aerosol particle formation.”

reviewer: p.6525 r2: Schallhardt et al. “in preparation” is not a valid reference. If this is not submitted to a journal by the time of revision, then I suggest to provide an outline of the calibration and calculation procedures here as “Supplementary Material”.

- authors: The manuscript is currently in preparation, therefore we provide a short PTR-TOF calibration/calculation scheme in the supplement.

reviewer: p.6526 r19: CH<sub>4</sub> background of 7 ppm is very high. Is this a typo (Fig. 2 would suggest that)? Or is there a strong CH<sub>4</sub> source permanently upwind of the experiment location?

- authors: the expression “CH<sub>4</sub> background” is, in our context, indeed misleading and a value of 7 ppm would be too high for conditions without additional source influence. In our case, however, the measurements took place in a situation with several CH<sub>4</sub> sources rather close by, for example: the slurry pit, silage storage, polluted surfaces. Such sources elevate the local CH<sub>4</sub> concentration, and on top of that we identified concentration increase associated with the action of present cattle. Periods without strong cattle CH<sub>4</sub> emissions were before the morning milking, between milkings, and

mainly during night, which is the reason for the given 7 ppm value (CH<sub>4</sub> concentrations increased in the stable nocturnal boundary layer). In the manuscript the expression “Background concentration levels” is replaced by “Typical concentration levels without the direct influence of animals” and we specify the corresponding concentration as a range between approx. 3 and 7 ppm.

reviewer: p.6527 r11: At what height were wind speeds measured? Was temperature measured, too? Temperature affects NH<sub>3</sub> emission rates; those of TMA as well?

- authors: Wind speed was measured at 1.5 m above ground, as was air temperature. Temperature was lowest in the early morning with 13 to 15 °C, and peaked during daytime with 23 to 26 °C. Temperature affects NH<sub>3</sub> and also TMA solubility. This argument is used in the discussion, p. 6530, l.4-7.

reviewer: p.6529 r11-15: There is nothing wrong in these sentences about CH<sub>4</sub> release, but it may be worthwhile pointing out more clearly that the CH<sub>4</sub> peaks are less frequent than those of acetone, because CH<sub>4</sub> release occurs with every “burp” while acetone release (apparently) occurs with every breath. Hence, correlation of the two time series is actually quite poor (Fig. 3a).

- authors: Yes, we will state this more clearly with an additional statement

reviewer: p.6536 r9-14: If the lifetime is only 1 min, why is dispersion considered for 8.4 min? How was the dilution “down to 1 ppt” obtained, from a model calculation or a reference? Why is “mixing . . . with cleaner air” considered separately, when it is the very same process of turbulent mixing that spreads a plume and entrains air from outside its initial boundaries to the inside? Please rework this passage.

- authors: We agree with the reviewer that it would be desirable to consider all processes within one consistent dispersion-chemistry model. For simplicity, however, we split the reduction of the TMA concentration into dilution and condensation, the latter representing the lifetime given in the manuscript. The TMA lifetime is between 30-1000

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seconds due condensation to pre-existing aerosol particles. The lifetime of 1 minute was chosen as the best estimate for typical central European conditions and the very long lifetimes is estimated only for very clean environments. As stated on page 6535 line 18, when amine concentrations exceed the sulphuric acid concentration nucleation rates will be increased substantially. Lifetime is defined as the time it takes for the concentration  $c$  to be reduced to a concentration of  $1/e \cdot c$ . On average we measured approx. 10 ppb TMA. In central Europe the concentration of sulphuric acid is in the order of 0.01 ppt. On the one hand, we calculated the time of 8.4 minutes which it takes for the typical TMA concentration to be depleted below 1 ppt sulfuric acid concentration by the condensation on particles. On the other hand, the 1 : 100 dilution is an estimate based on dispersion modelling during typical daytime boundary layer conditions over the travel time of 8.4 minutes. This exercise is supposed to give an impression of the possible dispersion time window for TMA availability of a farm source to contribute to particle formation. We will rephrase the paragraph in the manuscript for clarification.

reviewer: Fig. 2: Nice display of some essential results. The NH<sub>3</sub> time series looks rather messy, though, not as convincingly affected by excretion and scraping as the TMA time series. Also, there is no clear diurnal cycle.

- authors: it is our experience that, in a high concentration regime caused by local sources such as the farm itself, it can be common that the concentration at one location fluctuates strongly around the mean value within a given time period of, say, 10 to 30 minutes (e.g. Sintermann et al., 2011, Atmos. Meas. Tech. 4, 599-616). In Fig. 2 of the present manuscript, the anyway high NH<sub>3</sub> concentrations at the farm are even more increased by the NH<sub>3</sub> emissions from the cow's excrements during and subsequent to their presence in the stable. It seems like the related concentration increase and considered temperature dependence is less pronounced for NH<sub>3</sub> than for TMA. On the baseline of these peaks (occurring in the morning and afternoon) the NH<sub>3</sub> concentrations do increase slightly again in the night, associated with a stable nocturnal boundary layer frequently forming in that area.

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reviewer: Also Fig. 2: Why is acetone elevation higher and more prolonged after the afternoon milking than it is after the morning milking? I find this counter-intuitive because stronger accumulation should occur in the morning when stratification tends to be stable, rather than in the afternoon when I would expect good mixing due to unstable stratification. This is not really the topic of this paper, but I'm still curious.

- authors: This has to do with the time period the animals spend in the stable, which was longer in the afternoon than in the morning. Also, the typical management (here: gathering of animals) resulted in a higher animal density at the open-air waiting yard in the afternoon, thus more concentrated emissions. The difference is not so pronounced for CH<sub>4</sub>. It might be the case that the animal behavior varied during the day, e.g. that the cows ruminated/eructated differently during the course of the day.

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