Answer to Referee 2 comments

We thank Referee 2 comments which we hope will help us improving our manuscript.

General Comments

Loubet and others study a new method for inferring ammonia loss from small agricultural plots. I found the modelling analysis to make for an interesting case study regarding the applicability of field experiments. The approach treats bias errors carefully. As a consequence, I feel that the manuscript makes an earnest effort to quantify biases associated with passive ammonia sampling over small agronomic field plots and will be a valuable contribution to the literature.

Minor comments:

'Further work should anyway be produced for validating this method in real conditions' at the end of the abstract does not sound hopeful. Rather, the authors should try to discuss strategies for further improving the method and reducing uncertainties.

We thank the reviewer for this comment. We have indeed identified two strategies for further improving the method: (1) using Bayesian inference which has the potential for constraining the emissions and avoiding unrealistic sources inference as shown by Yee and Flesch (2010), and (2) changing the cost function (also called objective function); instead of inferring the emission strength, we could infer the emission potential Γ (a strictly positive number). This last method has the advantage of avoiding non-plausible deposition fluxes, because the flux is calculated as the concentration above the source minus the concentration at the ground, divided by transfer resistance. With such an approach negative fluxes (deposition) can occur within the limit of plausible transfer resistances but not above. We believe that the combination of these two strategies has the potential to improve the method substantially.

We think that "calibration" of the method against controlled sources is a remaining challenge that needs to be tackled (as also suggested by the comments of A. Neftel and C. Hanni in the interactive discussion).

We hence propose to add this more positive statement at the end of the abstract: "We believe that the method could be further improved by using Bayesian inference and inferring surface concentrations rather than surface fluxes. Validating against controlled sources is also a remaining challenge."

Line 41: 55.3% sounds remarkably specific given uncertainties in measuring NH_3 flux.

This is a sound comment. We propose to change to 55%.

53: 'most of the time large fields' is awkward wording.

Indeed. We propose to change to “most of the time also requires the use of large fields”

57: agronomic trials are not necessarily of those dimensions.

This is a sound remark indeed and we should not be as general as we were. We would propose to change to: “Especially useful for measuring ammonia losses are methods that can deal with small and medium-scale fields (20-50 m on the side) that are commonly used in agronomic trials.”

Parentheses on line 67.

Thanks for spotting this. We have withdrawn the left parenthesis.

118: quotes are unnecessary.

We agree and have withdrawn them.

On 130, what is the typical reaction time (and thereby Damkoehler number?)

Typical Damkhöler numbers showed by Nemitz et al. (2009) above a cut grassland canopy fertilised with ammonium nitrate were from 0.001 to 1. Values greater than 0.1 only occurred marginally, and usually during night-time conditions (Figure 6 in Nemitz et al. 2009). We would of course expect larger Damkhöler number values for slurry application which may generate larger concentrations than those reported by Nemitz et al. (2009), or with surface canopies having larger residence times. But in any case we expect the chemical depletion of ammonia to remain small at the spatial scale we are focussing on (around 200-300 m).

I find the tau near the overbar in 2 and other equations to be a bit distracting because it could be confused with an exponential term.

This is a sound remark. We propose to remove the *taus* and just leave an explanation in the text that the overbars denote averages over the period tau.

Equation 4 could be rearranged to reflect that only the numerator of the second term on the right hand side is unknown.

It is true that the numerator of the second term is the only unknown. However we can’t see how to isolate this term apart from multiplying by D(x). Moreover, leaving the equation as it is now has the advantage of explicitly showing this term which is the bias. We hence propose to keep equation (4) as it is.

251: why is z_{ref} 3.17 m? The curly braces in $R_b\{NH_3\}$ I find to be a bit distracting.

Regarding z_{ref}, we subjectively choose to use the reference height z_{ref} as the height where our ultrasonic anemometer was placed in the field, which simplified the calculation of the aerodynamic resistance for us. This does not have much importance anyway as we assumed that atmospheric ammonia concentration was zero.

Regarding Rb, we propose to change Rb{NH3} to R_{bNH_3}.

263: is there a justification for the model in simulation 2?

Exponential decrease in emission potential is representative of strong NH_3 emissions like those happening following slurry application. The value of 4.6 and the time scale τ_0 were chosen arbitrarily and would represent emissions a little bit less intense than those for nitrogen applications reviewed by Massad et al. (2010). In fact the equation we used here would be equivalent to a time scale equal to 6 days while in Massad et al. (2010) they report a time scale of 2.88 as being representative of slurry application. We propose to add the following text in Line 270: "The time scale of the exponential decrease we used here was around 6 days, which is twice as large as the one reported by Massad et al. (2010) for slurry application (2.9 days)."

265: what are typical parameters for the Gaussian model? Also, what mechanism causes it? The urea spreader?

The Gaussian model is rather representative of urea application. Indeed, NH_3 emissions result from combined processes: first the urea is hydrolysed by urease enzymes which release ammonium which can be volatilised but can also be nitrified or absorbed by roots. This leads to typical emissions starting a few days following application and showing a maximum up to 15 days following application but also a slower decrease of the emissions following the peak.

The Gaussian model was centred on day 14 with a standard deviation of 8.4 days.

267: I understand why 4.6 now in simulation 2. . .but why does this 'best' represent NH3 emissions?

As explained in previous paragraphs and following Massad et al. (2010) this model best represents slurry applications.

302: why is the covariance term negligible at the half hourly period? The spectral gap in eddy covariance studies?

The covariance term is indeed negligible at that time scale because of the so-called spectral gap in eddy covariance studies. This gap corresponds to time scales at which there is little energy in the turbulence and surface flux spectra (see e.g. Van der Hoven {, 1957 #25437}). We propose to replace the sentence at line 302 by "In practice the concentrations were computed at each sensor location using Eq. (6) over 0.5h: at that time scale, which corresponds to the spectral-gap, the covariance term is assumed to be negligible (Van der Hoven, 1957)."

303: in 2.5.3, these are not hypotheses as they cannot be falsified, even in the model.

This is indeed an interesting remark. We propose to change to the term "scenario" instead.

327: extra point

Thanks for spotting this. We have removed it.

336: how close is 'nearby'? From the figure it looks like it was part of the larger setup.

The meteorological data were measured at around 25 m away from the edge of the central plots (Figure 2). We propose to change the sentence for clarification: "The meteorological data were measured at less than 50 m from the central plots (Figure 2)".

355: results should be written in the past tense.

Thanks for the comment. We propose to change this sentence also to clarify its meaning : "The friction velocity u* varied between 0.024 and 1.181 m s-1, and the stability parameter z/L varied between -49 and 21 m-1 (Figure 3)"

365: define Gamma for the reader in the figure legend.

Thanks for the comment. We propose to change the last part of the legend to "…with an emission potential □ = 10000"

Please avoid using red and green simultaneously in Figure 4. This figure appears to be made using R, and gray is also a default color. And honestly yellow is never a good choice on a white background.

The comment that Figure 4 was hard to read was also made by reviewer 1. We have hence simplified the Figure and we further propose to change the colors as suggested by reviewer 2:

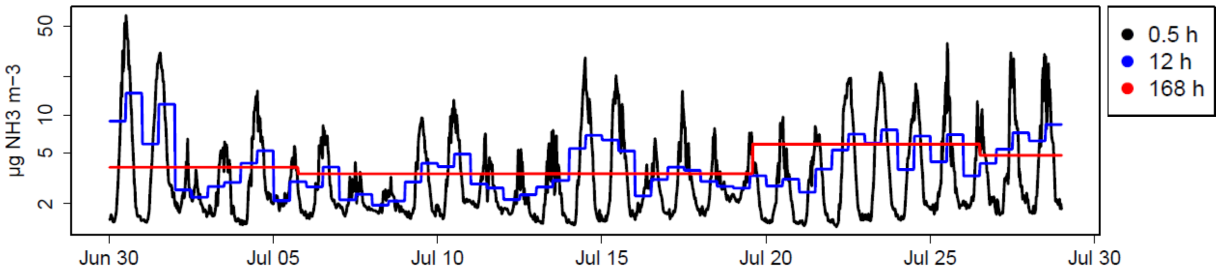

Figure 4. Example modelled concentration pattern at 1 m above a single 50 m width source for several averaging periods (0.5h, 12h and 168h) for the month of July 2008. The source Γ was set to 10^5. The y-axis is log scaled.

384: focuses

Thanks for spotting this typo. We have corrected it.

Figure 6 confuses me a bit because the 13 periods vary so strongly in their meteorological conditions from summer to winter, why are they grouped? The bars also leave the figure in the upper left subplot.

The idea for grouping the periods in Figure 6 but also in Figures 9-11 and 14 is actually to evaluate the variability of the bias due to meteorological conditions: ideally, if the method shows little variability in the bias, this bias could be characterised and even withdrawn. In Figure 6 we try to give a broad view of how the bias changes with sensor height and plot width. Figure 7 actually shows the variability of the bias due to meteorological conditions.

Regarding the scale, we chose to have a single scale for all panels to ease the comparison between heights and plot size, and we also chose to get the scale focussed enough to better see biases in the range -0.2 to 0.1. What we conclude from the upper left subplot is that the bias is much larger than all other cases which shows that that combination height-plot size is not satisfactory.

464-466: the attribution of stability with respect to continental vs. oceanic sites is too much of an approximation. There are many continental sites that are consistently windy, often due to orography.

We agree that we might have been too approximate in this statement, although we might still agree on the fact that oceanic conditions are typically windy. We propose to withdraw the reference to continental or oceanic climate to make it more general and replace the sentence for the following one: "We conclude that the inference method with a long integration period will lead to very moderate biases for locations with near-neutral conditions and high wind speed, but may lead to much larger bias under stable conditions and low wind speed as soon as the integration period gets up to 12 hours."

There is a strange x on line 468. Font sizes for figure 7 should be increased.

Thanks for spotting the x. It came from a problem when pasting Figure 7. We propose to modify Figure 7 to increase font size and improve as follows:

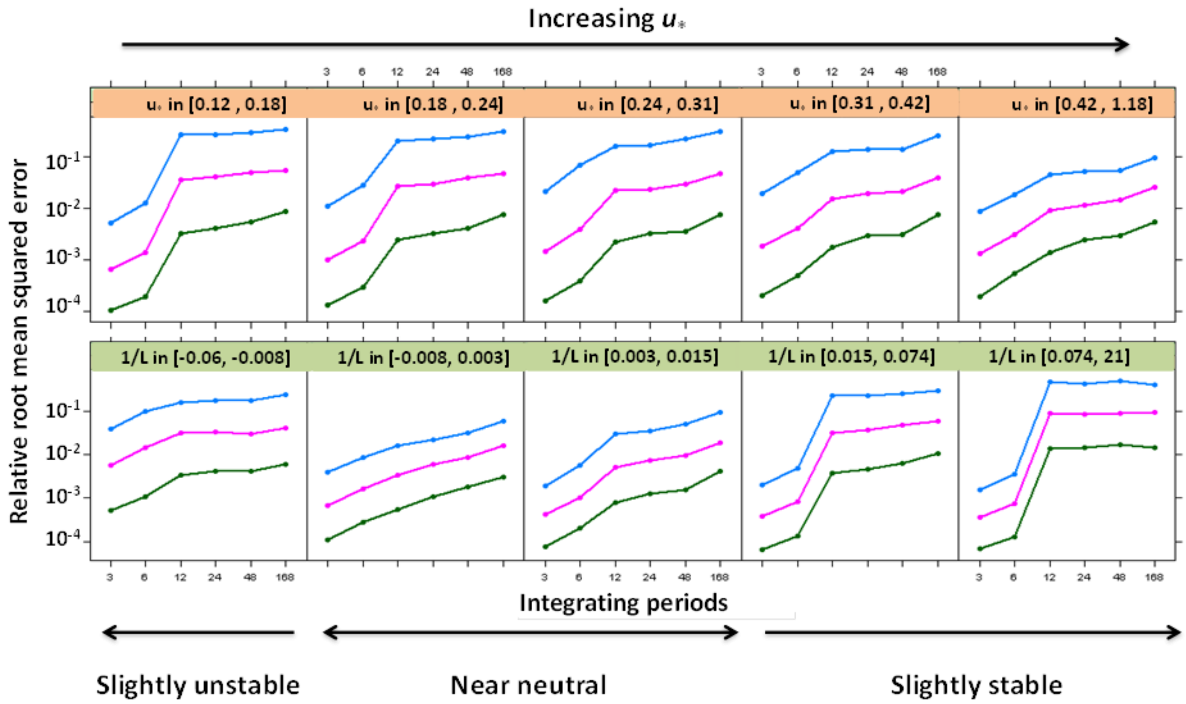

Figure 7. Relative root mean squared error as a function of integration period for stability factor and friction velocity classes for a single 25 m side field. Medians and quartiles are given for equally sized bins of u_* and $1/L$ and for the lowest sensor height (0.25 m). The blue, pink and green curves are the 3rd, 2nd and 1st quartiles, respectively.

741: why bird colonies?

Actually the only references we found where this bias was evaluated were those from emission estimates from bird colonies. We propose to withdraw the mention to bird colonies as this does not add much to the conclusion statement.

742: again, continental does not imply low wind speeds.

As in previous comment we propose to withdraw the reference to continental climate.

References

Massad, R.S., Nemitz, E. and Sutton, M.A., 2010. Review and parameterisation of bi-directional ammonia exchange between vegetation and the atmosphere. Atmospheric Chemistry and Physics, 10(21): 10359-10386.

Nemitz, E. et al., 2009. Aerosol fluxes and particle growth above managed grassland. Biogeosciences, 6(8): 1627-1645.

Yee, E. and Flesch, T.K., 2010. Inference of emission rates from multiple sources using Bayesian probability theory. J. Environ. Monit., 12(3): 622-634.