

Please see in our comments and discussion points below in blue.

General comments

This paper summarises the results and interpretation of two intensive periods of nocturnal measurement over 4 nights during 2011 and 2012 to estimate a local emission of methane where NBL estimates are compared with inventory estimates from the National Inventory (NIR) emission for methane, and two spatially explicit inventory estimates at national scale (SEI) and for local area (CHAI).

(1) P21786-Li22 It is very useful to see a comparison of NBL technique with inventory estimates, particularly for the local area "based on actual livestock data" and as the authors mention, few studies have done this. I feel it would add considerable value to this paper and broaden possible comparisons if the authors could give information about the animal numbers for Chamau farmstead for CHAI and for the local area used with SEI estimates and from this add some estimates of emission per head estimated by the NBL method and how this compares to the numbers that have been used in NIR, SEI and CHAI because many animal science and the inventories are based on emission per head (in addition to giving the per area fluxes).

The CHAI inventory estimates are calculated on the basis of actual livestock census data from the years 2011 and 2012 (see table 1), including the emissions from enteric fermentation as well as from manure management. In addition the CHAI inventories are based on the aggregation of emissions from different animals species, e.g. diary cattle, non-diary cattle, young cattle, sheep, swine and goats for 2012 additionally. For a better understandability, we will introduce these numbers with in the methodological part of the paper of P21776-Line17: „The CHAI inventory estimates include different animal headcounts for the years 2011 and 2012, respectively, . In 2011, 55 diary cattles, seven non-diary cattles, nine young cattles, 18 sheeps and 129 swines were located at the chamau farmstead. The inventory estimate for 2012 includes besides 81 diary cattles, two non-diary cattles, 18 young cattles, eight sheeps, 47 swines also 39 goats.“

Since the applied NBL budget integrates the emissions over all sources located in the respective area, the disaggregation of the estimate onto the different animal species might only be done with a generalization over all species. This will introduce high uncertainties and the resulting emission per head numbers might not be useful anymore for a reasonable comparison with the inventory estimates.

| | dairy cattle | non-diary cattle | young cattle | sheep | swine | goats |
|--|--------------|------------------|--------------|-------|-------|-------|
| Population 2011 | 55 | 7 | 9 | 18 | 129 | - |
| Population 2012 | 81 | 2 | 18 | 8 | 47 | 39 |
| Total emission per head [kg CH ₄ yr ⁻¹] | 148.13 | 94.01 | 38.16 | 11.23 | 6.77 | 11.33 |

Table 1: Population numbers and emission per head at the Chamau farmstead in 2011 and 2012.

(2) I feel that a bit more analysis is warranted given the profiles of 2012 that appear remarkably well mixed with height (in Fig. 3). As mentioned on P21784, the profiles may be influenced by a relatively large area of order hundreds km² and on P21785. The authors note the "well mixed" nature of the methane and its build through the night. There is useful discussion on P21780 and it appears that with increased wind aloft or possible jet activity may have been sufficient to keep the stable NBL stirred and as mentioned, it appears "that local sources only had a minor influence". However the CHAI would suggest that local sources were larger in 2012 than 2011; alternatives are that these sources have (i) mixed vertically through the lower atmosphere locally which seems unlikely, or (ii) that they have not been captured because too close and do not fall in measurement footprint for winds experienced at the measurement site which is solely "seeing" distant sources contributing to the NBL or (iii) both local and distant sources are well mixed. Can more analysis be done to understand the

2011 / 2012 differences add more interpretation? Figure 4 gives useful additional information for 2011, could a similar figure be looked at for 2012. Alternatively or further it would be instructive to look at the Richardson Number through each of the profiling periods; perhaps a critical number is always exceeded in 2012 to allow turbulent mixing to continue and is perhaps sufficient to mix up local emissions in spite of light wind at the surface (e.g. along lines of Grachev et al 2013) whereas perhaps turbulence is suppressed in 2011 case. Whilst this may help explain differences in the profiles, there is also no sign of a capping in the methane profiles and if the NBL flux is only integrated to Zi (50m) then it looks like a substantial component of the flux is missed as it appears that methane above this height is well coupled to the surface layer. There is some discussion of uncertainty introduced by assumptions in Zi etc. This analysis depends to a degree on whether the Chamau farmstead region a relative "hotspot" of methane emission or is it surrounded over the hundreds of km scale by land of similar or larger mean emission as this affects potential advection flux errors.

Grachev, A., Andreas, E., Fairall, C., Guest, P., and Persson, P. O.: The Critical Richardson Number and Limits of Applicability of Local Similarity Theory in the Stable Boundary Layer, *Boundary-Layer Meteorology*, 147, 51-82, 2013.

After having calculated the gradient Richardson number for each 10-m layer of the vertical profiles, it could be seen, that we did not experience any turbulent conditions which could have introduced mixing processes (see example in Figure 1). Ri tends to be decreasing with height or is highly variable when wind speeds are low (there is a division by zero problem if the vertical increase/decrease of wind speed is within the dead band of zero change with height). However, as the reviewer correctly states, the well-mixed CH₄ concentration profiles in 2012 might be influenced by further sources, which could not be excluded although having restricted the NBL budget estimates to specific wind directions and integration heights. We assume that advective effects were causing the well-mixed profiles in different manners (see Section 4.2). In order to put further emphasis on that aspect, we will change our text as suggested under point 2 in our response to reviewer's no. 2.

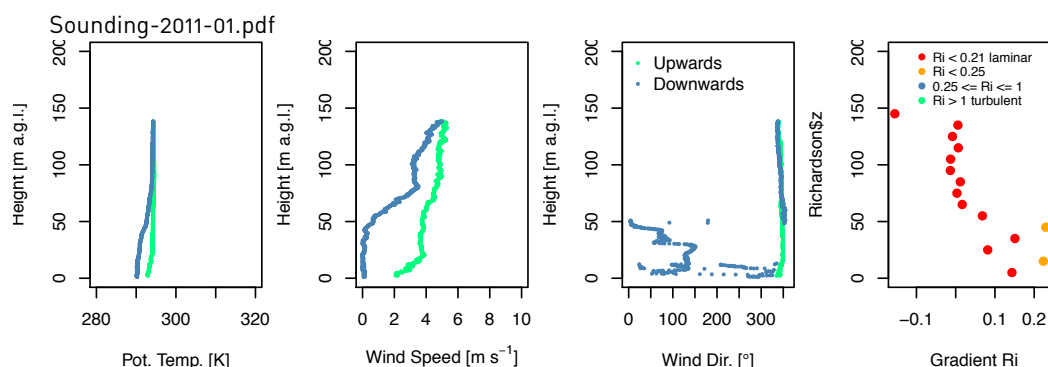


Figure 1: Profiles of potential temperature, wind speed, wind direction and gradient Richardson number, shown exemplarily for sounding 1 in 2011.

Reviewer questions: 1. Does the paper address relevant scientific questions within the scope of ACP? Yes

2. Does the paper present novel concepts, ideas, tools, or data? Yes, there are few other authors that have attempted this quite challenging method of validation

3. Are substantial conclusions reached? Yes in comparison with inventory estimates. Discussion of components of uncertainty would be valuable

We agree and will add this further aspects to the uncertainty discussion in the final version (see above our response to point 2).

4. Are the scientific methods and assumptions valid and clearly outlined? See point 2 above re interpretation of well mixed profiles and Zi assumptions

5. Are the results sufficient to support the interpretations and conclusions? A small amount of further analysis is suggested.

We carried out additional analysis (see example above) and will do our best to benefit from the constructive feedback.

6. Is the description of experiments and calculations sufficiently complete and precise to follow their reproduction by fellow scientists (traceability of results)? Yes

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution? Yes with Acknowledgements

8. Does the title clearly reflect the contents of the paper? Yes
9. Does the abstract provide a concise and complete summary? Yes
10. Is the overall presentation well structured and clear? Yes
11. Is the language fluent and precise? Yes

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? Yes

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? Not necessary

14. Are the number and quality of references appropriate? A few additional references may be useful.
15. Is the amount and quality of supplementary material appropriate? n/a

Specific remarks

P21768-Line11: the IPCC methodology for NIR could be cited: e.g. IPCC: IPCC Guide- lines for National Greenhouse Gas Inventories. In: Prepared by the National Green- house Gas Inventories Programme, Eggleston, H. S., Buendia, L., Miwa, K., Ngara, T., and Tanabe, K. (Eds.), IGES, Japan, 2006.

The IPCC reference will be included for the final version.

P21768-Line16 statement "require detailed knowledge of the transport pathways of tracers and the location of active CH₄ sources with which they must be collocated to provide realistic flux estimates" needs clarification. For the individual animal measurements, (Johnston, Deighton) the SF₆ tracer source is typically located in the rumen to be collocated with the main source location of enteric methane. It is unclear what if any short-comings the authors are suggesting with this technique.

In order to clarify this statement we suggest following rewording of the paragraph P21768-Line12: "These tracer techniques have been useful for the quantification of known CH₄ source components and especially of individual emissions per animal, requiring detailed knowledge of the transport pathways of tracers and the location of active CH₄ sources. However, such methods tend to have some limitations as the scale of integration increases to a whole farmstead or even larger spatial scale. For the validation of an emission inventory that is supposed to cover the total of all known and unknown (or neglected source components), mostly volume- integrated budget estimates are used, since such an approach has the potential to also reveal unexpected or unexpectedly large flux components of CH₄."

P21768-Line18 : "for the validation of an emission inventory that is supposed to cover the total of all known and unknown (or neglected) source components. " Atmospheric verification

will potentially see all sources of methane to the area, the emission inventory does not need to account for natural methane sources. This point is discussed by Hiller et al 2014a who note that natural and semi-natural sources make up only 3% of the total emissions in Switzerland. Can the authors comment whether emissions in the study region are likely to be affected by any natural sources?

Although natural and semi-natural methane sources are minor in Switzerland, they might have an important impact on the overall methane budget of a region. In another study currently in preparation, we address the question of source partitioning of agriculturally dominated areas using $d^{13}C$ measurements of air samples of the Chamau research station. The measurements revealed that despite increased methane mixing ratios during night, the observed $d^{13}C$ values were partially enriched by non-biogenic methane and/or sink processes. Possible non-biogenic sources were identified as vehicular methane emission from less efficient combustion of farm machinery or vintage cars, emissions from biomass burning (e.g. grilling on charcoal) and leakages in gas pipelines from upwind lying agglomerations. As a result, non-biogenic methane sources in combination with local advective transport processes contributed up to 22% to the local daytime methane budget.

J. Stieger et al. (2015): Source partitioning of atmospheric methane using carbon isotopes. In prep.

P21769-Line8: The authors state that "Little is known about the temporal variability". I note that Harvey et al, 2002 recommends that "In order to compare nocturnal measurements with daytime or 24 h measurements, any systematic diurnality in flux needs to be accounted for." They suggest that " In general, the diurnal pattern of methane emissions is linked closely to animal feeding patterns, which in turn may be linked to times of availability of fresh feed" and that systematic difference may occur where there are set feeding times or feeding habits if freely grazing. Can the authors comment on whether there could be any systematic biases in ruminant sourced methane due to any regular diurnal patterns of feeding?

Harvey, M. J., Brailsford, G. W., Bromley, A. M., Lassey, K. R., Mei, Z., Kristament, I. S., Reisinger, A. R., Walker, C. F., and Kelliher, F. M.: Boundary-layer isotope dilution/mass balance methods for measurement of nocturnal methane emissions from grazing sheep, *Atmos Environ*, 36, 4663-4678, 2002.

It is correctly stated, that feeding patterns and the general diurnal activity of animals can have an impact on the temporal methane emission strength. However, during our measurement periods, all animals were located inside the farmstead buildings resulting in fixed feeding times and identical temporal feeding patterns for each individual animal. However, the impact of the diurnal feeding pattern is neglected, as the NBL budget estimates are based on nocturnal measurements, which are not influenced by additional food availability during night. The reference of Harvey et al. (2002) will be part of the above-mentioned paper in preparation, where night- and daytime methane and $d^{13}C$ measurements are compared and used to reveal the impact of livestock methane emissions on the regional methane emission budget.

P21769-Line14: "only few projects, so far, have directly validated livestock CH₄ emission estimates via atmospheric concentration measurements without the deployment of chambers". Few projects should be clarified, and only refs to Denmean and Grobler given. There have been a number of studies at a number looking at free grazing animal emissions using a variety of techniques without the deployment of chambers e.g. Judd et al 1999, Lassey et al 2011, 2013, Laubach and Kelliher, 2004, 2005a,b, Laubach et al 2008, Wratt et al 2001 etc. and with comparison with chambers, e.g.: Grainger et al 2007.

Grainger, C., Clarke, T., McGinn, S. M., Auld, M. J., Beauchemin, K. A., Hannah, M. C., Waghorn, G. C., Clark, H., and Eckard, R. J.: Methane emissions from dairy cows measured

using the sulfur hexafluoride (SF₆) tracer and chamber techniques, *J Dairy Sci*, 90, 2755-2766, 2007.

Judd, M. J., Kellier, F. M., Ulyatt, M. J., Lassey, K. R., Tate, K. R., Shelton, I. D., Harvey, M. J., and Walker, C. F.: Net methane emissions from grazing sheep, *Global Change Biol.*, 5, 647-657(611), 1999.

Lassey, K. R., Pinares-Patiño, C. S., Martin, R. J., Molano, G., and McMillan, A. M. S.: Enteric methane emission rates determined by the SF₆ tracer technique: Temporal patterns and averaging periods, *Anim Feed Sci Technol*, 166–167, 183-191, 2011.

Lassey, K. R.: On the importance of background sampling in applications of the SF₆ tracer technique to determine ruminant methane emissions, *Anim Feed Sci Technol*, 180, 115-120, 2013.

Laubach, J. and Kelliher, F. M.: Measuring methane emission rates of a dairy cow herd by two micrometeorological techniques, *Agric For Meteorol*, 125, 279-303, 2004.

Laubach, J. and Kelliher, F. M.: Methane emissions from dairy cows: Comparing open-path laser measurements to profile-based techniques, *Agric For Meteorol*, 135, 340- 345, 2005a.

Laubach, J. and Kelliher, F. M.: Measuring methane emission rates of a dairy cow herd (II): results from a backward-Lagrangian stochastic model, *Agric For Meteorol*, 129, 137-150, 2005b.

Laubach, J., Kelliher, F. M., Knight, T., Clark, H., Molano, G., and Cavanagh, A.: Methane emissions from beef cattle – a comparison of paddock- and animal-scale measurements, *Aust. J. Exp. Agric.*, 48, 132-137, 2008.

In order to avoid a misunderstanding, we suggest to change the wording of the statement and include further references: „ So far, a fair number of studies estimated livestock CH₄ emissions via atmospheric concentration measurement without the deployment of chambers (Judd et al., 1999; Denmead et al., 2000; Laubach et al., 2004; Laubach et al., 2005a; Laubach et al., 2005b; Laubach et al., 2008; Grobler et al., 2014). Unfortunately, only few studies compare their results directly to inventory estimates (Levin et al., 1990, Lowry et al., 2001; Hsu et al., 2010; Hiller et al. 2014b).“

P21771-Line 10 Please give tube diameter as this affects Reynolds number for the tube and how fast and degree of turbulence in gas transported.

For our system with an inner tube diameter of 4 mm and an average flow rate of 1 L min⁻¹ the resulting Reynolds number is 344, which indicates laminar flow and gas transport. Since the flow was not turbulent in the intake tube a longer delay and a more pronounced smoothing of short-term concentration variations are expected, but since only mean concentrations over longer time scales of hours are needed for a boundary-layer budget, laminar flow in the intake hose is not of concern as it would be for an eddy covariance system (where turbulent flow is important to resolve the short-term variations that we actively smooth out here).

Figure 2: what is explanation for the sharp cut-off at 150m in the kriged plot, is this an artifact of the analysis? Further, in a similar way to the 2012 profiles, if Zi is found to be 50m there would appear to be a component of the methane plume that has mixed above this height and do the authors consider possibility of flux underestimation from this by not integrating the whole vertical extent or ignoring the Fent at the top of the NBL?

The reviewer correctly mentioned the sharp cut-off at the 150 m a.g.l. in the kriged plot to be an artifact of the kriging procedure. Kriging provides options to force interpolations and extrapolations to follow a prescribed direction, which we however did not use. Since we work at night when the NBL is stratified, we were not concerned about the fact that the Kriging automatically provided a pronounced layer structure above the maximum height reached during the hours after midnight on 17 August 2011. This is not unrealistic, but since we observed the unexpected fact that the inversion layer tends to exceed (slightly) our maximum height achieved by the tethered balloon, we do not have a sound basis to force the Kriging procedure to weigh the vertical direction more than the horizontal direction. Hence we left Fig. 2 as

is, but we will add a sentence in the caption to explain the feature: „Kriged time-space interpolation of the CH₄ concentrations obtained from the balloon measurements during 16/17 August 2011. Vertical and horizontal kriging directions were equally weighted, resulting sometimes in sharp vertical concentration changes.“

Regarding the underestimation of the NBL flux: The temperature profiles reveal a highly stratified atmospheric boundary layer in 2012 (see P21779-Line10). In order to include emissions from the Chamau farmstead and exclude the impact from sources further upwind, the integration height z_i was set to a height where the first transition from stable to neutral stratification was detected. Integrating the whole vertical extent for the NBL budget will introduce uncertainty about the fetch of the resulting estimate and might include emissions from sources outside the Chamau farmstead. Although the NBL budget might underestimate the effective methane flux, the results compare well to the CHA1 inventory estimate, pointing to the reasonable selection of z_i .