Reply to Referee #1

Referee comment in bold, reply in plain text, *modified text for manuscript in italics*.

Henne et al. present a comprehensive analysis of 2013 Swiss methane emissions using atmospheric observations from the new CarboCount-CH measurement network. They perform a suite of inversions using different specifications of the uncertainty, definitions of the background, and inversion frameworks. Importantly, they find emissions that are consistent with the Swiss Greenhouse Gas Inventory (SGHGI). This is an excellent manuscript. It is well written, uses state-of-the-art inversion techniques, the figures are high quality, and demonstrates that well-informed bottom-up inventories can accurately represent the emissions. This manuscript should be published in ACP. The only comments I have are, seemingly, minor.

We would like to thank the referee for the positive review and will address all minor issues in the revised manuscript.

Best inversion?

It could be useful for the authors to suggest a "best" setup. This would help inform others doing similar atmospheric inversions. The authors may want to perform an additional inversion using the "best" setup. They currently say the best estimate is the mean over all sensitivity inversions (Page 35452, Lines 19-21). However, this seems a little odd because it means they don't actually have a map of their "best emissions". It seems like they could try one additional inversion using the setups that performed best (extKF, ML method, COSMO transport, seasonality, all sites, MAIOLICA, and Grid baseline).

Also, why don't the extended Kalman Filter inversions have Skill scores for FRU and GIM in Table 4?

Although there might be a best setup in the sense that its results are closest to the truth, this best setup is not known (as little as the truth is known). The ML method applied here is an objective method to tune the free parameters of an inversion but this doesn't necessarily correspond to the "best setup" since it cannot account for potential biases arising from transport errors or the problems in representing the release height of the particles. Our approach therefore was to provide a reference setup corresponding to our expert judgment of a best setup (as in many previous Bayesian inversion studies) but, in addition, to test a range of other setups and configurations that we consider similarly likely. We argue that the range of estimates from these individual setups provides a more realistic estimate of uncertainty than the analytical uncertainty of a single setup. We included these arguments more prominently at the beginning of the results section in the revised manuscript.

Our Kalman Filter inversion system is currently not able to consider additional validation sites and FRU and GIM would need to be validated manually. We did not consider this additional effort proportionate to the gained insights.

Unidentified source in north-eastern Switzerland

Page 35456, Lines 23-26: The authors mention that different manure management methods could lead to slight variations in emission factors. However, the differences due to manure management can be dramatic. Owen & Silver (2014) find orders of magnitude differences in the methane emission rate between anaerobic lagoons, slurry tanks, and manure piles (see their Fig. 3; Anaerobic lagoons emit at a rate about 35× larger than manure piles!). They also include a revised liquid manure slurry manure conversion factor that's about 2× larger than what's currently used by the European Union.

Have there been changes in the regional manure management practices (maybe due to a shift to more concentrated animal feeding operations)?

According to Daniel Bretscher from the Swiss institute Agroscope for agricultural research (which is responsible for collecting the information on Swiss GHG emissions from agriculture), no such changes in regional manure management practices occurred in Switzerland recently. Although different manure management methods may result in different emission factors, we have no indication that manure management practices vary strongly within Switzerland. Furthermore, the average emission rate for anaerobic lagoons given by Owen & Silver (2014) is associated with a rather large uncertainty as can be seen in their Fig. 3 but especially when comparing the range of individual values given in their table 2a. Considering this range the given average value should be taken with care. Most importantly, manure storage in anaerobic lagoons does not exist in Switzerland, where the largest contribution to CH_4 emissions originates from liquid/slurry manure (90 %, FOEN, 2015). The manure conversion factor given by Owen & Silver (2014) for liquid/slurry is about a factor of 2 larger than the one used in the Swiss inventory. However, the latter is based on country specific emission factors (Hiller et al., 2014) and some Swiss studies even suggest considerably lower emission factors (Zeitz et al., 2012). In addition, there seems to be no indication that liquid/slurry storage is more frequently used in Eastern Switzerland than elsewhere in the country. On the contrary, the discussed region can be characterised by more traditional farming practices, which may suggest a larger fraction of manure piles and, hence, smaller CH_4 emissions from manure storage. We included a reference to Owen & Silver (2014) in our revised discussion. The unidentified source will be subject of a follow-up project, which started earlier this year and will include additional CH_4 measurements to target this source.

Discussion of US ruminant and natural gas emissions

In Section 4.2 (Page 35454, Lines 23-25) and Section 5 (Page 35459, Lines 5-7), the authors mention that the findings of reduced ruminant emissions are in contrast to studies from the USA (Miller et al., 2013; Wecht et al., 2014; Turner et al., 2015). It's not clear to me that these results are actually inconsistent. Miller et al. (2013), Wecht et al. (2014), and Turner et al. (2015) all find that US methane emissions from ruminants are underestimated. However, the Turner et al. (2015) shows little-to-no change throughout Europe (see their Fig.3) in their coarse global inversion. Another paper (Alexe et al.,2015) also find a a large increase in US emissions and a decrease in the emissions across Europe (see their Fig. 6). This would imply that Swiss ruminant emissions are not underestimated (in agreement with the finding from Henne et al.). It seems

that it may just be different regional features, not something pervasive in inventories (which would make sense because countries have different reporting requirements).

We revised the text accordingly and included a reference Alexe et al. (2015). Note that the emission increments reported by Turner et al. (2015) and Alexe et al. (2015) for Europe and the US depend on the chosen a priori (EDGAR v4.2). While EDGAR v4.2 is somewhat lower than the official inventory in the US (Turner et al., 2015), it is about 35% higher than the total of all European inventories reported to UNFCCC (Bergamaschi et al., 2015). This needs to be taken into account when interpreting their results.

The authors present a similar argument in Section 4.2 (Page 35455, Lines 2-5) and Section 5 (Page 35459, Lines 16-20) for natural gas emissions from the distribution sector. The authors claim their findings are in contrast to studies from the USA, "This is in contrast to recent studies from the USA where a large underesimation of fugitive emissions was established in the inventories for different metropolitan areas (Wennberg et al., 2012; McKain et al., 2015) and fractional loss rates between 2.5 and 6% were established." However, the fractional loss rates from Wennberg et al. (2012) and McKain et al. (2015) are probably not representative of the average US leak rate. Wennberg et al. (2012) examined emissions from Los Angeles, a city with a lot of oil and gas activity (the Aliso Canyon leak is, anecdotal, evidence of this), and McKain et al. (2015) examined emissions from Boston, an old city with a lot of cast-iron pipes. So these studies are not necessarily indicative of a pervasive problem in the inventory, as implied by Henne et al.

We added a comment towards the lacking representativeness of the mentioned US studies. However, we still feel that this is a more general problem for the US. Even if Boston is a relatively old city (obviously not as old as Basel or Zurich, but maybe that is not true for the gas distribution network) it is also one of the wealthier cities in the US and one may therefore expect a certain degree of modernisation. The main point we wanted to raise it that findings such as an underestimation of gas leak emissions observed in one country cannot easily be transferred to another country. We think this is an important message for the community. Note that the national inventories are often applying the same emission estimation methods based on default equations and parameters provided by the IPCC guidelines (known as "Tier 1" method) and thus one may expect that results obtained in one country may also be applicable in other countries. For losses from the gas distribution network, however, Switzerland applies a country-specific "Tier 2" method. It accounts, for example, for the fact that cast-iron pipes have gradually been replaced by polyethylene pipes which is expected to have reduced CH4 emissions from gas transmission and distribution by 36%. There are thus significant differences between countries regarding infrastructure and emission estimation methodologies, which is a strong motivation for country-specific studies as the one presented here.

Consider adding a brief discussion of Zavala-Araiza et al.

I feel like one of the biggest strengths of this paper is demonstrating the consistency between a well-informed bottom-up inventory and the top-down estimates. This was the main point of the

recent paper: Zavala-Araiza et al. (2015). It seems that this paper could be another one that demonstrates this point.

We added the findings of Zavala-Araiza et al. (2015) in our conclusions emphasising the common reconciliation of top-down and bottom-up.

Implementation of the inversion

On Page 35430 the authors state, "In our implementation of the inverse of S = MBMT − R, a L × L matrix, was calculated using LU factorisation (function DGESVX in LAPACK)." This seems like it may be an inefficient implementation. The DGESVX routine in LAPACK is for dense linear algebra. However, the matrices in atmospheric inversions are typically sparse (usually less than 1% of the elements are non-zero). I've found that I get a 31× speedup and use 17× less memory by just switching from dense to sparse matrices for large atmospheric inversions (KE > 2, 000, 000). There are also routines that allow for efficient solutions to large inverse problems with covariance matrices that can be represented as a Kronecker product (cf. Yadav & Michalak, 2013; they provide source code for the routines). The authors could also look into some of the multi-scale state vector design methods from Bocquet et al. (2011).

Bocquet et al. (2011) presented methods for optimally designing a multi-scale state vector that allowed information to transfer across scales. These are merely suggestions for future code development that may allow the authors to expand the scale of the problem (Increase the number of observation sites? Estimate monthly emissions? Expand the inversion domain?). The implementation presented here is sufficient to tackle the problem they are interested in.

We thank the reviewer for these very interesting remarks on the technical aspects of the inverse algorithm and will certainly follow up on them during future code developments.

Specific comments

Pages 35421-35422, Lines 29-1: Should add a reference to Zhao et al. (2009), Jeong et al. (2012), Jeong et al. (2013), and Ganesan et al. (2015).

We added Zhao et al. (2009), Jeong et al. (2012) and Jeong et al. (2013) at this point in the manuscript.

The study by Ganesan et al. (2015) was not carried out at the high spatial resolution that we were referring to. They used a transport model at approximately 25 km horizontal resolution and also their inversion grid was rather coarse (estimating emissions for approximately 60 regions across the UK). Nevertheless, we added a reference to Ganesan et al. (2015) in our discussion of European agricultural emissions.

Page 35425, Line 9: Is this an issue if wind direction changes? 3hr seems like a long time.

We used 3-hourly aggregates to reduce the computation time for the transport model, which was also only started for 3-hourly intervals. However, the model is driven by hourly wind data, so changes in advection direction will still be reflected in the model result. Also particles are released continuously within each 3-hour interval, so that changes in advection are considered in the model. The aggregation certainly will smooth some of the observable variability but we feel that we don't have a strong loss of

information from the aggregation, especially since we also consider a temporal correlation length in the model/observation covariances of half a day, which adds additional smoothing of the contained information.

Page 35426, Lines 9-30: How are the PBL heights? I didn't see any mention of evaluating them.

PBL heights are a critical parameter in FLEXPART, since they are used as a scaling parameter for the turbulence parameterisation. We use the standard implementation within standard FLEXPART to diagnose PBL heights from a Bulk-Richardson method (Stohl et al., 2005;Vogelezang and Holtslag, 1996). In contrast to standard FLEXPART we did not use 2-metre temperatures from COSMO in the PBL estimation but the lowest model level temperature (~10 m above ground), since MeteoSwiss COSMO 2-meter temperatures are known to have a warm bias during convective conditions. COSMO internal and FLEXPART diagnosed PBL heights were compared by MeteoSwiss for the sounding site Payerne on the Swiss Plateau. The comparison showed a positive bias for COSMO and also for FLEXPART derived PBL heights when using 2-m temperatures. This bias mostly vanished when using the first level temperature as done in our simulations. We added this information to the manuscript.

Page 35428, Lines 8-13: Why are you using a fixed height? Shouldn't it be the mixed layer height? It seems that using a percentage of the PBL height would be more reasonable.

While we agree with the referee, FLEXPART does not allow a variable sampling height (yet). The chosen sampling heights agree with the minimal PBL used by the different model versions, therefore avoiding that the sampling height is ever larger than the mixing height. During convective mixing the particles should be homogeneously distributed throughout the PBL and a sampling height smaller than the PBL height should be valid. However, we agree that particle statistics might improve with a variable sampling(Zeitz et al., 2012). We did tests with doubled sampling heights that did not indicate systematic changes in simulated sensitivities. Therefore, we don't think that the current sampling height introduces systematic biases in the transport description.

Page 35429, Line 21: I think it should be "K_E _ 1000", not "J_E _ 1000".

Yes. We corrected this in the revised manuscript.

Page 35430, Line 9: How many observations? I'm assuming L < K since you're using the L-form.

Actually L > K (approximately L~3000 observations versus K~1400 elements in the state vector) for most of the presented inversions. We realise that under these conditions equation 5 may be solved more efficiently when reformulated. This will be subject of further code development as already mentioned above.

Page 35431, Line 17: What is f_E? I didn't see it in the table. What is the resulting grid-scale uncertainty? Is there a floor on the uncertainty?

f_E is the proportionally constant between prior emissions and prior uncertainties. f_E was set constant for all land grid cells with significant emissions and the value was chosen so that the country total emission uncertainty for Switzerland became 16 %. The resulting value for f_E was 0.3 and this information

was given on page 35437 line 1. We clarified this connection in the revised manuscript. To avoid very low uncertainties in grid cells with low emissions we adjusted the uncertainty for land grid cells with less than 10% of average land cell emissions and for ocean grid cells to a minimum uncertainty corresponding to 10 % of the average land cell uncertainty. This procedure was tested with several threshold values and did not at all affect the results for Switzerland and hence this information was not given in the manuscript.

Page 35432, Line 25: Are observations only correlated for a given site or all sites?

No correlations between sites were treated as mentioned on line 5 of page 35433.

Page 35435, Line 10: Spent a lot of time explaining the Bayesian, maybe give the equation for the extKF or explain how Q was specified.

We added the following lines at the end of Section 2.5.3:

Accordingly, uncertainties of the state vector are allowed to grow from one time step to the next which introduces an additional amount of prior uncertainty as compared to the Bayesian approach. The matrices B and R were parameterized according to equations (9) and (12), respectively. The chosen parameter values are listed in Table 3. The forecast uncertainty matrix Q was also parameterized according to equation (9), notably with the same spatial correlation length. The diagonal elements of Q were set to a relative forecast uncertainty of the emissions of 0.6% per 24 hours which resulted in fairly constant a posteriori emissions with only a small seasonal cycle.

Page 35437, Line 1: How? How is summing diagonals with 30% uncertainty giving you a smaller uncertainty (16%) at the national scale (without anti-correlations in the off-diagonal)?

The uncertainty of the sum (E) over two independent emission elements (a,b) with a relative uncertainty of 30% would only be 21 % if a=b. The following equations should illustrate this.

Total emissions:

$$E = a + b = 2a$$

Uncertainty:

$$\sigma_a = f_E a$$

Total uncertainty (Gaussian error propagation for uncorrelated uncertainties):

$$\sigma_E = \sqrt{\sigma_a^2 + \sigma_b^2} = \sqrt{2}\sigma_a \ = \sqrt{2} f_E a$$

Total relative uncertainty:

$$\frac{\sigma_E}{E} = \frac{\sqrt{2} f_E a}{2a} = \frac{1}{\sqrt{2}} f_E = 0.707 f_E$$

So the relative uncertainty of the sum is a factor of 0.707 smaller than the relative uncertainty of the each individual emission element. Hence, even in the case of positive covariances the relative uncertainty of the sum can be smaller than the relative uncertainty of the individual elements. This calculation is also reflected in equation 7.

Page 35438, Line 15: Should say “separated”, not “separating”.

Corrected

Page 35438, Line 16: Should say “ensure”, not “assure”.

Corrected

Page 35438, Line 19: Should say “ensures”, not “assures”.

Corrected

Page 35448, Line 15: Should say “distinct”, not “destinct”.

Corrected

Page 35448, Line 27: Should say “sensitivity”, not “sensititivy”.

Corrected

Page 35453, Line 15: Should cite Ganesan et al. (2014).

Added

Page 35454, Line 21: Should say “inferred”, not “infered”

Corrected

Additional References

Alexe, M., Bergamaschi, P., Segers, A., Detmers, R., Butz, A., Hasekamp, O., Guerlet, S., Parker, R., Boesch, H., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Sweeney, C., Wofsy, S. C., and Kort, E. A.: Inverse modelling of CH4 emissions for 2010–2011 using different satellite retrieval products from GOSAT and SCIAMACHY, Atmos. Chem. Phys., 15, 113-133, doi: 10.5194/acp-15-113-2015, 2015.

Bergamaschi, P., Corazza, M., Karstens, U., Athanassiadou, M., Thompson, R. L., Pison, I., Manning, A. J., Bousquet, P., Segers, A., Vermeulen, A. T., Janssens-Maenhout, G., Schmidt, M., Ramonet, M., Meinhardt, F., Aalto, T., Haszpra, L., Moncrieff, J., Popa, M. E., Lowry, D., Steinbacher, M., Jordan, A., O'Doherty, S., Piacentino, S., and Dlugokencky, E.: Top-down estimates of European CH4 and N2O emissions based on four different inverse models, Atmos. Chem. Phys., 15, 715-736, doi: 10.5194/acp-15-715-2015, 2015.

FOEN: Switzerland's greenhouse gas inventory 1990-2013, Submission of April 2015 under the United Nations Framework Convention on Climate Change and under the Kyoto Protocol, Federal Office for the Environment (FOEN), Bern, 2015.

Hiller, R. V., Bretscher, D., DelSontro, T., Diem, T., Eugster, W., Henneberger, R., Hobi, S., Hodson, E., Imer, D., Kreuzer, M., Künzle, T., Merbold, L., Niklaus, P. A., Rihm, B., Schellenberger, A., Schroth, M. H., Schubert, C. J., Siegrist, H., Stieger, J., Buchmann, N., and Brunner, D.: Anthropogenic and natural methane fluxes in Switzerland synthesized within a spatially explicit inventory, Biogeosciences, 11, 1941-1959, doi: 10.5194/bg-11-1941-2014, 2014.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461-2474, doi: 10.5194/acp-5-2461-2005, 2005.

Turner, A. J., Jacob, D. J., Wecht, K. J., Maasakkers, J. D., Lundgren, E., Andrews, A. E., Biraud, S. C., Boesch, H., Bowman, K. W., Deutscher, N. M., Dubey, M. K., Griffith, D. W. T., Hase, F., Kuze, A., Notholt, J., Ohyama, H., Parker, R., Payne, V. H., Sussmann, R., Sweeney, C., Velazco, V. A., Warneke, T., Wennberg, P. O., and Wunch, D.: Estimating global and North American methane emissions with high spatial resolution using GOSAT satellite data, Atmos. Chem. Phys., 15, 7049-7069, doi: 10.5194/acp-15-7049-2015, 2015.

Vogelezang, D. H. P., and Holtslag, A. A. M.: Evaluation and model impacts of alternative boundary-layer height formulations, Bound. Lay. Met., 81, 245-269, doi: 10.1007/BF02430331, 1996.

Zeitz, J. O., Soliva, C. R., and Kreuzer, M.: Swiss diet types for cattle: how accurately are they reflected by the Intergovernmental Panel on Climate Change default values?, Journal of Integrative Environmental Sciences, 9, 199-216, doi: 10.1080/1943815X.2012.709253, 2012.