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Interactive comment on "Estimation of waste water treatment plant methane emissions: methodology and results from a short campaign" by C. E. Yver-Kwok et al.

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Interactive comment on "Estimation of waste water treatment plant methane emissions: methodology and results from a short campaign" by C. E. Yver-Kwok et al. W. Eugster (Referee) werner.eugster@usys.ethz.ch Received and published: 22 December 2013

The authors present an interesting methodological paper to assess the methane emissions from a waste water treatment plant in France near Valence. The field experiment uses an impressive combination of state-of-the-art gas analyzers, includes the natu-

ral 222Rn tracer and the artificial C2H2 tracer applied to the water tank. In principle, they also measured CO2 and 13C in CO2 but did not analyse these gas emissions. The most critical point is the short duration of the measurements in combination with the gappy 222Rn data from the FTIR, which makes all quantitative statements rather problematic, as the authors state themselves. A possible scaling error with natural 222Rn flux estimates, and the somewhat arbitrary approach to estimate extreme emissions (see my detailed comments below) reduce the scientific quality of the overall manuscript. However, the presented material has a relevant scientific value for advancing the methodology to measure CH4 fluxes from waste water treatment plants and thus the authors should be given a chance to revise and improve their manuscript for AMT.

We thank the reviewer for his insightful comments. As stated in the manuscript, we are aware of the fact that the duration of the campaign was too short to provide robust emission estimates. However, the main objective was to demonstrate the suitability of the different methods for different scales. Thank you for pointing out the incoherence in format and units. We will correct the wording and unit errors. Given that, hereafter, we refer to the content-related questions.

Details 9188/3: how can you be sure that it is only the temperature variablity that matters for the reproducibility? Are you sure that the 2C higher temperature does not matter, only the variability?

####The FTIR regulates the cell temperature by blowing heated air from the room on it. In the laboratory, the temperature is less than 30°C so the FTIR indeed can warm up the air. When the cell's temperature is above 30°C, the heating stops. But the FTIR keeps blowing outside air on the cell to cool it down. So, if the room's temperature is above 30°C, the FTIR will not be able to cool down. When the cell is at a stable temperature, the signal is also stable, it is the variations of temperature and the time for the FTIR to adjust to it that accounts for the variability. That is why we have increased the value to avoid having variations due to the facts that above this value, the FTIR

can not regulate the temperature. Moreover, the instrument was regularly calibrated during the campaign with the cell temperature fixed at 32°C to ensure that even if the raw measurement could be different at 32 than at 30°C the calibrated measurements are the same.

9193/6: on 9192/21 you defined H as "a well-mixed layer of height H", whereas here you call it the "boundary-layer height". This is confusing since the boundary layer is only well mixed during unstable stratification, i.e. during the day, but not at night. However, you implicitly pretend Eq. (5) can be used at any time, which is not quite correct. Please (a) be consistent in the wording, and (b) clarify the issue with non wellmixed conditions (since you do not limit your analysis to well-mixed conditions). Under nocturnal conditions Eq. (5) would need to be expanded to account for the different diffusivities of 222Rn and CH4.

####We have reformulated the text as followÂă: ÂńÂăAssuming that the gases (here CH4 and 222Rn) are released from the atmosphere at a constant rate F in a layer of averaged height integrated along the air mass trajectory H, we can write the temporal variation of their concentration above background C (with an additional radioactive decay term for 222Rn).ÂăÂż This reformulation refers to the publication from Biraud et al., 2000. As redefined, the method applies to any period of the day. In our case, the only event was in the afternoon, with a well-mixed layer.

9193/23: this is not unproblematic: you use a specific, local measurement for everything, but a very coarse, generalized 222Rn flux map. Hence, all errors in the estimate of the true 222Rn flux at your locality translates to errors in CH4 flux estimates. I am not convinced that 25% uncertainty is a realistic estimate for your environment. It is the problem that the uncertainty of a mean values is definitely lower (i.e. 25% in your estimate) than that of a specific timepoint as it is used in Eq. (5).

###Unfortunately, we do not have radon flux measurements at the site. We estimate the value from the radon map from Szegvary et al (2007,2008) with a pixel size of

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about 50 km. According to the soil type map from Zobler (http://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=540), the soil type in the area around the station is mostly agricultural, so we can assume that the radon flux does not vary much at our particular location compared to the rest of the pixel. O f course, as we have only one data point, the uncertainty is not measurable. The 25% error can only be applied to a statistically significant number of data.

9193/22 and elsewhere: Bq is sôĂĂĂ1 in SI base units, so your units are definitely wrong here – Bq already has the time reference it it, multiplying Bq with hôĂĂĂ1 gives an acceleration, which is nonesense. Please rectify, check all numbers. The value in the map under http://radon.unibas.ch is on the order of 1 Bq cmôĂĂĂ1, so for 1 môĂĂĂ1 it should be 10000 Bq môĂĂĂ1, I guess. What you wanted to do with the addition of hôĂĂĂ1 remains obscure. Please rectify.

###In all the publications, the activity of Radon is expressed as Bq m-3, so s-1 m-3 and then the flux being defined as J=C*H/t, we indeed end up with s-2 m-2 in order to be homogeneous.

9194/9–10: the selection of a threshold correlation of 0.6 seems rather arbitrary. How did you chose this threshold, how can it be justified?

###This threshold was selected in previous studies as a compromise between a good correlation and a significant number of events. We have added this explanation in the revised text.

9199/18–20: you call this a "very conservative upper limit", but from only a few days of measurement I am not sure you have the full insight into the true variability of the system. Why are you not using a (more robust) extreme values distribution approach (as e.g. the Gumbel approach, or a Pareto approach) to make a sound estimate for the extremes? We tried the Gumbel approach with eddy flux data (see Eugster et al. 2010) and this should also be a sound approach for your estimates.

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###In our case, we have very few events and thus can not apply any statistical methods. We have decided to delete this sentence in the revised version and keep only the results without further interpretation.

9203/22–26: this is the weakest aspect of the manuscript: you realized that 4 days is not enough to make a sound estimate. But since this manuscript was submitted to a journal that rather focuses on the method than on the results obtained with this method, I see the value of the manuscript to be published after revisions. However, as a reviewer, I am not supportive of this least-publishable-unit approach.

###As stated in the manuscript, our main objective was to apply and evaluate different methods of estimating emissions for a waste water treatment plant. We think this was done with success during the campaign. We have decided to also present and discuss the emissions obtained during the campaign but it was clear that these are first results, here to illustrate the methods more than to give a complete evaluation of the site. We have decided to publish these methods and results without waiting for a next campaign, as we felt that this material was interesting enough by itself.

Fig. 4: it is obvious that CH4 and 222Rn have very different time constants here, hence a correlation approach is not unproblematic (serial autocorrelation and serial cross-correlation problem). In my view Rn is to a great extent the inverse of the wind speed, which indicates the relevance of mixing during conditions where the atmosphere is stably stratified. It is also obvious that CH4 responds more strongly to abrupt changes in wind direction than Rn. I think the main problem in your approach is that you use Eq. (5) also for conditions when the atmosphere is not well mixed at all.

###Radon with its mostly constant exhalation rate is proxy for the boundary layer height, with higher concentration when the layer is low and lower concentrations when the layer is high. Methane follows also these variations but its sources are more variables in time and space. In our case theyalso vary locally while the measured radon emissions are spread regionally. The only event took place from noon to 20:00Z

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so mostly in a well-mixed layer as we were still in summer. We attempt to take into account these problems by choosing only events with a strong cross-correlation. The difference in wind speed and wind direction during the event makes the estimation of the influence area more difficult to calculate but does not change the fact that in this area, CH4 and Radon emissions were correlated.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/6/C4724/2014/amtd-6-C4724-2014-supplement.pdf

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