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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Methane and nitrous oxide emissions in The Netherlands: ambient measurements support the national inventories" *by* S. van der Laan et al.

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We thank both referees very much for their encouraging remarks and comments with which we can improve our work. Detailed responses to the comments follow below:

Anonymous Referee #1

[RC] Para 3, section 3.2, page 6: stable implies low wind speeds and low boundary layers (inversion heights), this is not consistent with increasing wind speeds especially to over 8 m/s. In this instant it is the wind direction changes that drives the event. [AC] Text adapted accordingly.

[RC] Para 4, section 3.2, page 6: there is always an atmospheric inversion (boundary layer) they are just at different heights at different times. This needs to be re-worded to be correct. [AC] Point taken. Text adapted accordingly.

[RC] Para 1, page 7: This assumes that the wind speed is constant during the transit. It should also be noted that it assumes no change in the wind speed with height, this is clearly a significant simplification. Clearly there must be some vertical mixing to mix the surface emission to the inlet height of 60m. Usually the vertical mixing is constant within the boundary layer. [AC] Point taken.

[RC] Para1, page 8: The cut-off of 0.7 is arbitrary, it would be interesting to understand the sensitivity to this parameter by say calculating the emissions with say a 0.6 and a 0.8 cut-off. Para3, page 8: You could argue that by only accepting r>0.7 the impact of big point sources has been removed and therefore using the lognormal distribution is double counting this effect. I am not convinced that the median result is not a reasonable estimate. I agree that the mean is affected by outliers but the median is not. I would include the median results in table 1 as well and not be negative to this method. [AC] Both referees raise the question about our choice of R>=0.7 as a threshold for the linear regressions fits of the events, and if choosing a different R would result in different results. As the Radon flux method assumes a relation between Radon and the other tracer, it is clear that we cannot include all regressions from 0<=R<=1. On the other hand, choosing only regressions fits with R>0.99 is also impossible since there too few of them. We therefore choose R>=0.7 in order to ensure that there was a significant relation between the two, and still have enough data for analysis. If we choose R=0.6 as a threshold, our results would be lower by about 5% and with R=0.8 the results would be higher by about 8%. The values would therefore still be in the range of the results and uncertainties that are given. The positive relation between R and the flux is probably caused by the fact that nearby (point) sources have, in general, a high R (less disturbance), and because the soil close to the tower has a high moisture content (close to the sea) and therefore a lower radon soil flux compared to the

average value of the Netherlands. The latter could cause these (point) sources to be overestimated (Eq.1.). More research is needed in the future to be sure, but a higher R does therefore not necessary guarantee more reliable results (in terms of estimation of emissions of the whole country). We have added this in the discussion section.

[RC] Equation 2, page 9: Surely this should be applied per observation during the event as each has a different time of influence. For example if there were 3 obs in an event taken at times t1, t2 and t3, Obs at t1 could be corrected using eq2 but with tmax set to t1, similarly for the other obs. The total length of the event would only be correct for obs at the end of the event.

[AC] In principle. the only correct way is to apply a decay correction for each Radon soil emission for the transit time belonging to that emission. In the case of an emission from 4 days ago this will also contain Radon from 3 days ago and 2 days etc. which al will be decayed accordingly and have to be included in the decay correction. In practice the exact transit time and soil emission is however not known. The linear regression fits contain all Radon observations versus all (i.e.) CH4 observations during one event and are therefore integrated over the whole footprint and for the total observation time. The best we can do is assume a constant radon soil emission (which is generally assumed in the radon flux method) and calculate an average decay correction based on the average decay of the radon collected during the event. This is how it is usually done, but it is not clear how the average transit time is estimated. We propose to use the total length of an event as an approximation for tau max and apply Eq.2. which then gives the average decay correction for the whole air mass. We have clarified this in the text.

[RC] Page 11, para 1: Median is not heavily influenced by a few very high fluxes. In fact the median estimates agree much better to the UNFCCC inventory than the lognormal estimates. [AC] We agree with these statements from the referee, we have however more faith in the values given by the lognormal regression fit as the distribution of the fluxes are clearly lognormal (figs. 6b & 7b). For the monthly averaged long range events (figs. 9b & 10b) this is indeed less clear since they are not subjected to nearby

point sources. The median results are therefore also included in table 1.

[RC] Page 12, para 2: What baseline is used for the long range analysis. This is not made clear. [AC] The lowest mixing ratio in the month is used as a baseline. This is clarified in the text.

[RC] Para1, page 7: what was assumed an (un)acceptable variation in trajectory during an event? [AC] The trajectories are subjected to uncertainties not well known (i.e. related to interpolation of wind fields etc.) which make high-resolution analysis difficult. The analysis is mainly used to distinguish between emissions from The Netherlands, which is a relative small country, and its surrounding countries. We aimed at selecting trajectories which have a track at least for 70% over the Netherlands.

[RC] Figure 3: What do the dots signify? Hourly points on the trajectory? [AC] Yes, this is clarified in the text.

[RC] References: No reference to Forster 2007 or Maas 2008 (or maybe I missed them?) [AC] These are in the manuscript.

line 2, page 2: “are significant contributing” to “are significantly contributing” para 2, pag2: change ”obliged” to “encouraged” para 3, page 3: change “data is” to “data are” para 4, page 3: remove reference to “middle European”, it does not aid understanding and is confusing. para 4, page 3: surely easterly winds also sample continental air? Line 2 section 3.1, page 5: remove “is used” Para1, section 3.2, page 6: An event will also terminate when wind direction changes. Para 1, page 7: again add “or wind direction change” Para2, page 7: what is the “resp.” for ? Para2, page 8: Add an extra bit to add understanding. “The Netherlands (i.e. those used in the short range analysis), as well” Section 4.1.1., para 1 page 10: change “and longer events” to “ and long range events” Section 4.1.1. , para 2 page 10: change “chapter” to “section” Page 11, section 4.1.2.: change “similar as CH4” to “similar to CH4”. Page 11, section 4.1.2.: same comment as above. Section 4.2, page 11: change “data which is” to “data which are”. Page 13, para 2: What is “resp.” for? Page 13, para 2: Missing reference to

Eckhardt 1990. Page 14, para 2 last line: Change " asses " to " assess ". Figure 1: Add that met taken from top of 60m mast. [AC] These comments have been adopted in the text.

Anonymous Referee #2:

[RC] in section 2.2 it is not clear how often samples are actually measured, so the temporal resolution of the initial dataset is not clear. This then made me less certain of the analysis methods. [AC] CH4 and N2O are measured every 6.5 minutes, 222Rn every half hour. For the analysis the temporal resolution is therefore 30 minutes. This is clarified in the text.

[RC #1] It is not clear how dependent the analysis is on the choice of a regression coefficient of 0.7 (Section 3.2). How sensitive are the final estimates (uncertainties) to this assumption. [RC #2] Para1, page 8: The cut-off of 0.7 is arbitrary, it would be interesting to understand the sensitivity to this parameter by say calculating the emissions with say a 0.6 and a 0.8 cut-off. Para3, page 8: You could argue that by only accepting r>0.7 the impact of big point sources has been removed and therefore using the lognormal distribution is double counting this effect. I am not convinced that the median result is not a reasonable estimate. I agree that the mean is affected by outliers but the median is not. I would include the median results in table 1 as well and not be negative to this method. [AC] See our response to Ref#1.

[RC] The third issue is the comments on the comparison between a log-normal distribution and Gaussian-based mean and median (Section 3.2). The text implies the median is a "Gaussian" product, whereas it is the basis of a number of non-parametric statistical analyses for the very reason that it does not assume a distribution. It is not true that it is sensitive to a few outliers (Section 4.1.1). The use of a log-normal distribution may well be justified (and there are statistical tests available to demonstrate the effectiveness of "renormalization"). I would recommend clarifying the difference between these models, and probably based on the statistical evidence not report the

mean, but report the median and log-normal results. [AC] Point taken. Text adapted accordingly. The median results are also included in table 1.

Section 1 (line 4) temperature increases, which are inevitable. Later in the paragraph all units should be W m-2. At least in my version the "2" is missing on some. Para2, Section 1, line 7 (e.g. cattle, . . .) (shift the parenthesis). Para 3, line 1 the Netherlands (removal capital on T) Section 3.1, line 5. Via the soil air, radon (add the comma and replace "it") Section 4.1.1 , paragraph 2, line y. Remove "yet". Section 4.2, para 1, line 12 "seems" means what? Please clarify [AC] These comments have been adopted in the text.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 18867, 2009.