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Interactive Comment

Interactive comment on "Methane production from mixed tropical savanna and forest vegetation in Venezuela" *by* P. J. Crutzen et al.

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The criticism of Keith Smith concerns the magnitude of the global estimate of the CH_4 emission from tropical savannah and forest vegetation by Crutzen et al. He points out that their estimate may be an order of magnitude too large because two major assumptions may not be justified, namely:

1. that the nocturnal boundary layer (NBL) was at a height of 100 m, as reported for another occasion by Octavio et al. (1987)
2. that the methane was uniformly mixed from ground level to this height.

To support this, he states "...*it seems to be widely established within the microme-*

teorological community that, during the build-up of the NBL, gases do not mix well within the layer, and steep vertical profiles (such as those cited below) are common, and these profiles should be integrated over the NBL depth to infer the surface source strength (Culf et al., 1999)".

Furthermore, Keith Smith presents an example of weak nocturnal NBL mixing (CO_2 over an Australian pasture) resulting in a "*curvilinearly*" CO_2 concentration decrease from the ground surface to the maximum height. Additionally, he refers to very shallow NBL's observed elsewhere (in Australia and northern Italy; 40 m and 10 m, respectively), which may result (considering weak vertical mixing) in effective NBL heights of 16-17 m and 4-5 m, respectively. Since the nocturnal CH_4 emission estimate scales directly with NBL height, Keith Smith claims that the published estimate (based on a 100 m NBL) could be exaggerated 20-25 fold.

However, given additional material, specifically results of Sanhueza et al., 2000, it can be shown in detail that Keith Smith's criticism may not apply in the present case. Fig 8 of Sanhueza et al.'s paper, particularly vertical O_3 distributions on 25 October 1988, 23:00 LT and 26 October 1988, 03:00 LT (performed also at the Guri site, like the CH_4 measurements) clearly demonstrate:

1. the height of NBL at this site is 100 m above ground (at least reached 5 hours after sunset (18:00 LT), and
2. there is a marked linear decrease of vertical O_3 mixing ratio from surface to 100 m height (demonstrating nocturnal vertical mixing, which may be not perfect, but effective enough).

Given this information I calculated the nocturnal CH_4 emission rate at the Guri site. Assuming complete vertical mixing of any trace gas (as shown for many African and South American sites) in the tropical late afternoon CBL (Convective Boundary Layer), we consider a constant vertical CH_4 mixing ratio ($CH_4 \neq CH_4(z)$) from surface level to the upper end of the CBL (certainly higher than 100 m) at 17:00 - 18:00 LT. Latest at sunset (t_{SS} = 18:00 LT), the NBL will start to grow. For the sake of simplicity we assume an "instant" occurrence of NBL, i.e. a step-like function of h_{NBL} = h_{NBL}(t) (i.e., h_{NBL}=0 m at 18:00:00 LT and h_{NBL}=100 m from 18:00:01 LT onwards (until t_{SR} = 06:00 LT (sunrise) of the next day). Then, we assume that at any time $t_{SS} < t \leq t_{SR}$, the CH_4 mixing ratio at and above the NBL height (i.e., z $\geq h_{\mathrm{NBL}}$) is that CH_4 mixing ratio which has been observed at t_{SS} at all z (i.e., CH_4($t > t_{SS}$, $z \geq h_{\mathrm{NBL}}$) := CH_4(t_{SS}, z=0)). At surface (z=0), the CH_4 mixing ratio at any time $t_{SS} < t \leq t_{SR}$ is given by the results in Fig.1 of Crutzen et al.'s manuscript.

Next, we assume that the NBL over the Guri-site can be considered as a horizontally indefinitely outspread box with a tight lid at z= h_{NBL} (i.e., there is no vertical entrainment at the top and no horizontal advection). Accordingly, the methane flux deduced from the night time temporal increase of CH_4 mixing ratio is given by

$$F_{\mathrm{CH}_4} = h_{\mathrm{NBL}} * \partial \mathrm{CH}_4/\partial t \quad (1)$$

Assuming a well mixed NBL, the night time ($t_{SS} < t \leq t_{SR}$) surface CH_4 mixing ratio, CH_4(t,0), would be the same for all heights in the NBL (see red dashed curve in Fig. FXM1, available at http://www.mpch-mainz.mpg.de/~meixner/ACPD_graph/ACPD_comment_FXM_1.png).
The methane flux is then easily determined by

$$F_{\mathrm{CH}_4} = h_{\mathrm{NBL}} * M * [(\mathrm{CH}_4(\mathrm{t},0) - \mathrm{CH}_4(\mathrm{t}_{\mathrm{SS}},0))/(\mathrm{t} - \mathrm{t}_{\mathrm{SS}})] \quad (2)$$

where F_{CH_4} is in molecules cm^{-2} s^{-1}, h_{NBL} in m, CH_4 in ppm, t in h, and M (= 6.95411×10^{11}) is the conversion factor (ppm $\rightarrow$ molecules cm^{-3}, m $\rightarrow$ cm, h $\rightarrow$ s).

The nocturnal development of surface CH_4 mixing ratio (∂CH_4(t,0)/∂t) can be deter-

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mined from the data points given in Fig.1 of Crutzen et al.'s manuscript. The linear fit of CH_4(t,0) (between t_{SS} = 0 (17:00 LT) and t = 9 h (02:00 LT) (see Fig.FXM2, available at http://www.mpch-mainz.mpg.de/~meixner/ACPD_graph/ACPD_comment_FXM_2.png).

results in

$$\mathrm{CH_4}(t,0) = 1.775166 + 7.003037 * 10^{-3} * t; R^2 = 0.98822; n = 10 \tag{3}$$

(note that the decrease of the observed surface CH_4 mixing ratio after 02:00 LT (t=9h) is not consistent with the assumption of temporally constant nocturnal CH_4 emission and a well mixed NBL).
Using relation (3), CH_4(t=6,0) = 1.8172 ppm and CH_4(t_{SS}=0,0) = 1.7552ppm. For h_{NBL} = 100 m, the corresponding methane flux (in a well mixed NBL) would result in

$$F_{\mathrm{CH_4}}, \text{well mixed} = 4.87 \times 10^{11} \text{molecules cm}^{-2}\text{ s}^{-1} \tag{4}$$

To address Keith Smith's concerns about a "*not well mixed*" NBL, two additional (hypothetical) nocturnal vertical CH_4 profiles have been considered, linearly and exponentially decreasing (representative of different states of "*not well mixed*"). The "*linear*" case is the blue straight line, the "*exponential*" case is the green dashed-dotted line in Fig. FXM1.

In the "*linear case*", at any time $t_{SS} < t \leq t_{SR}$ the vertical profile of CH_4 mixing ratio is assumed to decrease linearly from z=0 to z=h_{NBL}, which is expressed as

$$\begin{aligned} &\mathrm{CH_4}(t,z) \;=\; a(t) + b(t) * z \qquad\qquad t_{SS} < t \leq t_{SR} \text{ and } 0 \leq \mathrm{z} \leq \mathrm{h_{NBL}} \\ &a(t) = \mathrm{CH_4}(t,0) \\ &b(t) = [\mathrm{CH_4}(t,h_{\mathrm{NBL}}) - \mathrm{CH_4}(t,0)]/h_{\mathrm{NBL}} \end{aligned} \tag{5}$$

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As suggested by Keith Smith, the nocturnal vertical CH_4 profile must be integrated, which leads to the following modification of (1), namely

$$F_{\mathrm{CH_4}} = \frac{\partial}{\partial t} \int_0^{h_{\mathrm{NBL}}} \mathrm{CH_4}\,(t, z)\, dz \tag{6}$$

For h_{NBL} = 100 m, CH_4(t=6,0) = 1.8172 ppm and CH_4(t_{SS}=0,0) = 1.7552ppm (from relation (3)), and integrating according to (6), the corresponding methane flux (linearly decreasing CH_4 profiles in the NBL) would result in

$$F_{\mathrm{CH_4},\text{linear decrease}} = 2.46 \times 10^{11} molecules\ cm^{-2}\ s^{-1} \tag{7}$$

which is half the rate estimated for the '*well mixed case*'. Graphically, referring Fig. FXM1, the methane flux in the "*linear*" case is equivalent to the (triangular) area surrounded by the x-axis, the purple and the blue straight lines, while in the "well mixed" case the corresponding rectangular area (surrounded by the x-axis, the purple and the dashed red lines) is just double the triangular area.

In the "*exponential case*", at any time $t_{SS} < t \leq t_{SR}$ the vertical profile of CH_4 mixing ratio is assumed to decrease exponentially from z=0 to z=h_{NBL}, which is expressed as

$$\mathrm{CH_4}(t, z) = \mathrm{CH_4}(t, h_{\mathrm{NBL}}) + [\mathrm{CH_4}(t, 0)] - \mathrm{CH_4}(t, h_{\mathrm{NBL}})] * exp(-k * z) \tag{8}$$

The "*decay*" factor k has to be chosen to 0.095, which is equivalent to a "*1/e decay height*" of 10.5 m.

For h_{NBL} = 100 m, CH_4(t=6,0) = 1.8172 ppm and CH_4(t_{SS}=0,0) = 1.7552ppm (from relation (3)), and integrating according to (6), the corresponding methane flux (linearly decreasing CH_4 profiles in the NBL) would result in

$$F_{\mathrm{CH_4,\ exponential\ decrease}} = 5.35 \times 10^{10} molecules\ cm^{-2}\ s^{-1} \quad (9)$$

Then, corresponding "exaggeration ratios" (c.f. Keith Smith) would read as follows:

$$\frac{F_{\mathrm{CH_4,\ well\ mixed}}}{F_{\mathrm{CH_4,\ linear\ decrease}}} = \frac{4.86999 \times 10^{11} molecules\ cm^{-2}\ s^{-1}}{2.45934 \times 10^{11} molecules\ cm^{-2}\ s^{-1}} = 2 \quad (10)$$

and

$$\frac{F_{\mathrm{CH_4,\ well\ mixed}}}{F_{\mathrm{CH_4,\ exponential\ decrease}}} = \frac{4.86999 \times 10^{11} molecules\ cm^{-2}\ s^{-1}}{5.35326 \times 10^{10} molecules\ cm^{-2}\ s^{-1}} \approx 9 \quad (11)$$

As already mentioned above, the results of Sanhueza et al. (2000), namely that (1) h_{NBL} = 100 m, and (2) a linear increase of nocturnal vertical ozone mixing ratio have been observed at the Guri-site, favours the application of the (hypothetical) linearly decreasing nocturnal CH_4 profile (eq. (5)). Then, the resulting CH_4 flux of 2.46×10^{11} molecules cm^{-2} s^{-1} is just below the lower end of the estimate by Crutzen et al. (2006), namely 3×10^{11} molecules cm^{-2} s^{-1}, but still sufficient to support the main conclusion of Crutzen et al..

References

Sanhueza, E., Fernandez, E., Donoso, L. and Romero, J., Boundary layer ozone in the tropical America northern hemisphere region, J. Atmos. Chem. 35, 249-272, 2

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3093, 2006.