

Interactive comment on “Implementation and evaluation of a new methane model within a dynamic global vegetation model: LPJ-WHyMe v1.3” by R. Wania et al.

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We would like to thank Timo Vesala for spotting this problem and raising awareness of it in the framework of the open discussion. We had an email exchange with Timo on this topic before the publication in GMDD, but we could not think of a solution to this problem and thought it would be good to draw the attention of other researchers to the problem of modelling ebullition, in order to stimulate further research on this issue and hopefully finding a solution.

I have found the fundamental problem in Section 2.7.1. Henry's law, used in 2.7.1, must be applied between the concentrations per se and not between the concentrations

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calculated from the amount of CH₄ (either gas or dissolved) per the whole system volume. The concentrations used in Eqs. 16-19 are now for the whole system. What is the concentration per se?

It is the amount of CH₄ per volume of the phase concerned, i.e. the amount of the dissolved CH₄ per the volume occupied by the liquid and the amount of gaseous CH₄ per volume of the gaseous phase, including also possibly air, water vapour etc. Henry's law gives the ratio of those concentrations. This distinction is very fundamental and important and the misuse can lead to unphysical situations.

To answer the question of what is the concentration in the gaseous and liquid phase, we would need to know the fraction of water-filled pore space and the fraction of gas-filled pore space in each peat layer. The peat layers we are talking about are fully inundated, so information concerning the water table would not help us to answer this question. In our manuscript, we made the assumption that the volume of the gas-filled pore space depends on the amount of gaseous methane, which again depends on the total methane available in any particular layer. The total amount of methane will be split between gaseous and dissolved form, as these two are found to be in equilibrium (depending on environmental conditions). Timo correctly points out that to calculate this equilibrium, we would need to know the gas-filled pore space first. However, this leads to a tricky problem since the volume of gas-filled pore space depends on the gaseous methane content of the layer.

We are thus faced with a circular problem: we cannot calculate the methane concentration in the gaseous phase correctly because we do not know how big the gaseous volume is; neither can we calculate the gaseous volume because that depends on the amount of gaseous methane (and other gases such as hydrogen, carbon dioxide and nitrous oxide). One approach to avoid this problem was taken by Tokida et al. 2007, who fixed the gaseous volume to 10% and calculated the methane concentration given that gas volume.

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I give an example: if one single, small bubble exists in the large volume of liquid, the concentration in the sense used in the manuscript (MS) would be practically zero, but the concentration per se would not be. The concentration inside the bubble (that is the concentration per se) would be determined by the Henry's law and the concentration per se of the liquid phase, which in this example would be practically the same as the concentration in the MS, since the liquid volume is much larger than the gaseous volume. Applying now the philosophy used in the MS, the formulas would predict increase of the gaseous CH₄, either increasing the size of the single bubble or formation of new bubbles. However, Henry's law does not say anything about the total amount of the gaseous phase dispersed in the liquid, Henry's law would tell only the equilibrium concentration per se for the gaseous phase, which would not depend on the bubble size or number of bubbles.

The way that we used Henry's law is to take the total methane concentration in a given layer, from which Henry's law allows us to calculate how much of that methane can be dissolved in the liquid phase given the environmental conditions and an upper limit of dissolved methane concentration. Whatever is not dissolved is in gaseous form. The volume of gaseous methane is then based on the amount of gaseous methane and the ideal gas law. The underlying assumption here is that there will always be enough nucleation sites for bubble formation and the gaseous volume will not be constrained based on nucleation site availability.

I do not know how to solve the problem in 2.7.1. To calculate the amount of gaseous phase would need the dynamic model for bubble formation by nucleation and their subsequent growth. It could be possible that the model in 2.7.1 provides reasonable results compared with the observations, but it is then coincidence since it is based on the wrong physics.

We don't know how to solve this problem either. To model bubble formation by nucleation would, in general, be very difficult, depending very strongly on soil, chemical and environmental characteristics that are difficult to predict, and, at least on a regional

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basis, may be fundamentally unknowable.

The reasonable behaviour of ebullition in our model may, as Timo says, just be a lucky coincidence. On the other hand, it may be an indication that fluxes due to ebullition are not terribly sensitive to the details of the processes of bubble formation and growth. We are not so concerned with the details of individual bubbles, but only with the gross flux due to ebullition.

I am very sorry for raising this nasty question, but I do not see how the model would work correctly at the present form. My motivation is that I would like to apply the model also in my own research and would like to know how to make it correctly, but at the moment I do not know how to calculate the total amount of gaseous CH₄ dispersed in the peat matrix. In addition, minor comment, the iteration procedure is not clear for more, it should be explained more thoroughly.

As we said above, we appreciate Timo's comment for raising this difficult question, even though we do not have a solution for it yet. We will attempt to add some material to the manuscript addressing this issue, in the hope that it may encourage other workers to think about the problem.

Minor comment:

Eq. 11 assumes the linear relationship between the gas phase diffusivity and the temperature. However, by the tools of statistical thermodynamics it can be shown the diffusivity is proportional to the temperature a $D \propto T$ where the factor a is always between 1.5 and 2 (e.g. Reid et al., 1987, Properties of gases and liquids, 4th ed., McGraw-Hill, New York). However, the unphysical linear temperature fittings may be valid for the limited temperature range of the original diffusivity data, but any values outside the fitted range must be taken by caution.

The diffusivity data we use from Lerman (1979) are based on the temperature range from 5–35°C. The upper limit of this temperature range is not exceeded in our data. The lower limit may be an issue as temperatures drop below 5°C, but we think that

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the potential error arising from this deficiency is small. First, we extend the linear relationship only down to 0°C. Beyond that point, water in the soil will freeze and no diffusion will occur. Secondly, the diffusivities of Eqn. 11 are only applied to the top layer in the model and only if the fractional air content is more than 5%. We agree that this shortcoming should be corrected in a future version of LPJ-WHyMe but we do not believe that it poses a source of significant error in its current form.

References:

Granberg et al. (1999). A simple model for simulation of water content, soil frost, and soil temperatures in boreal mixed mires. *Water Resour. Res.* 35(12), 3771-3782.

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