Harper and coauthors present a global atmospheric chemistry-climate model with methane emissions. The paper documents the emissions of methane and other compounds, then evaluates the simulated concentrations of methane and ozone against observations. The methods are reasonable and the comparison to observations is sufficient to show that the model appears to be performing competently. The paper is written clearly. Some methods need greater explanation and discussion, which can be accomplished with modest revisions.

The model construction and budget analysis are based on the assumption that atmospheric methane was in steady state in 2005. Although the atmospheric methane concentrations were approximately stable during 2000-2007, as the authors say on p7, atmospheric methane may not have been in steady state at that time because emissions and OH may have been changing (Rigby et al. 2017; Turner et al., 2017). The steady state assumption, its limitations, and implications for model interpretation should be discussed.

In the abstract and conclusions, the emissions magnitude and especially its partitioning into natural sources are stated too confidently and simply. These estimates assume that the prescribed anthropogenic emission inventory and the simulated CH4 loss are correct. Any error in these other budget terms would alter the authors’ estimate of natural emissions. The emissions values should be presented as a best fit within the context of the other model assumptions.

The paper needs greater detail about how the natural methane emissions were optimized. The general approach is described a bit in Section 2, but lacks detail for a reader to attempt to reproduce it. I suggest providing this greater detail in Section 3. What observations were used in the optimization? Was it a formal optimization of some cost function or ad hoc trial and error with visual comparison? I would expect that the optimal emissions would produce an unbiased global mean, but Section 4.1 reports and Figs 2 and 3 show that the model is systematically higher than observations at almost all sites.

In the abstract and elsewhere, 1% model bias against surface observations is acceptable, but not excellent. It may be comparable to the performance of other models, but it is one-fifth of the interhemispheric ratio NH/SH: 1.05. For a well-mixed gas like methane, a 1% model error after optimization is substantial.

The supplement contains data in Excel xlsx format. I recommend an open source file format readable by free software, but I defer to the editor on whether this is required.

Minor comments

P2L19: CH4 is also oxidized by O(1D) in the stratosphere.
P5L13. Is version 1.1 a past model version or the new version described by this paper?
Were the LLGHG concentrations prescribed at the surface or also elsewhere?

The work of Turner et al. is slightly misrepresented. The gross magnitude of methane emissions are well constrained, with uncertainty of 10% or less in the global total (Turner et al., 2017; also Prather et al., 2012). Turner et al. (2017) and also Rigby et al. (2017) showed that observations poorly constrain partitioning and small but important trends in this total, although see Prather and Holmes (2017) for ways that exploiting spatial patterns could extract more information from existing observations.

I believe the CH4 lifetime estimate by Rigby et al. (2013) should supersede Prinn et al. (2005), although the values are similar.

References


