

## **Response to Reviewer #1**

We thank the reviewer for their helpful comments, which have led us to an improved version of the paper. Here, the reviewer's comments are shown in boldfaced black text, and our responses are shown in non-boldfaced blue text. The page and line numbers to which we refer in our responses correspond to the updated manuscript (the comments of both reviewers are taken into account in this updated manuscript).

**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.**

- 1. 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.**

We agree with the reviewer that although the atmospheric methane concentrations were approximately stable during 2000–2007, atmospheric methane may not have been in steady state at that time because emissions and OH may have been changing at the same time.

We modified the description of the experimental set-up (Page 7, Line 22): “The E2005 simulation was run until atmospheric methane reached steady state, such that the global chemical sink came into balance with the net global source (prescribed sources minus prescribed soil sink), resulting in a relatively stable atmospheric methane abundance. Steady-state conditions were diagnosed using the global annual-mean atmospheric burden of methane. The final 10 years of the 45 year simulation are used for analysis. Year-to-year variation in the methane burden for the final 10 model years is < 3.2 Tg CH<sub>4</sub>. Year-to-year variation in the global-average surface methane concentration is < 1.3 ppbv. The year of interest for this study, 2005, fell within a roughly 8 year period that witnessed a largely stable global-mean concentration of methane in Earth's atmosphere (Dlugokencky et al., 2009). The observed stability in the concentration of methane does not necessarily indicate temporally invariant global sources and sinks over this era (Rigby et al., 2017; Turner et al., 2017). For example, a recent analysis by Turner et al. (2017) suggests that simultaneous counterbalancing changes in methane emissions and loss to OH may be responsible for the observed stability in the methane concentration in the early 2000s. Therefore, the methane budget derived in this study by assuming steady state conditions represents just one plausible solution that can lead to a stable atmospheric methane concentration. This assumption is convenient in global chemistry–climate modeling where the simulated climate state does not correspond to an exact meteorological year. The derived solution is constrained by both the prescribed methane fluxes and other forcing data that can affect atmospheric methane, such as: emissions of other short-lived compounds; the prescribed ocean conditions, which influence the physical climate state; and the concentrations of the non-methane long-lived greenhouse gases, which influence the radiation budget.

The non-wetland natural methane fluxes that are prescribed are based on published estimates (Sect. 3) and are representative of the 2000s contemporary era but are not necessarily specific to year 2005. Likewise, the prescribed sea ice distribution and sea surface temperatures are observation-based five year means centered on year 2005. The derived methane budget, therefore, represents a 2000s climatology and is approximately, but not precisely, representative of year 2005 conditions.”

- 2. 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 CH<sub>4</sub> 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.**

Following the reviewer’s suggestion, in the Abstract (Page 1, Line 18), we have added this sentence: “The wetland methane flux is calculated as a best fit; thus, the accuracy of this derived flux assumes accurate simulation of methane chemical loss in the atmosphere and accurate prescription of the other methane fluxes (anthropogenic and natural).”

We have altered the first sentence of the conclusions section (Page 26, Line 11): “The results of the optimization process using atmospheric modeling indicate global annual methane emissions of 140 Tg CH<sub>4</sub> y<sup>-1</sup> from wetlands; **this derivation assumes accurate representation of the other methane fluxes and atmospheric chemical loss in the model.** The global annual methane emissions magnitude from all natural sources is 181 Tg CH<sub>4</sub> y<sup>-1</sup>.”

The manuscript does already describe the limitation in our ability to partition between the various methane sources. Following the reviewer’s suggestion, we have further extended the key paragraph (Page 4, Line 16): “Using ModelE2, Shindell et al. (2013) previously used a similar procedure of modifying the wetland methane source to achieve a modeled methane concentration that is in line with present-day observations, noting that the accuracy of the magnitude of the wetland flux that is derived in this way depends on whether the other prescribed fluxes have been accurately assigned. **That is, the applied methodology calculates the wetland methane emission magnitude as a best fit under the assumption that the other methane fluxes and simulated atmospheric chemical loss are accurately represented in the global model.**” And here (Page 4, Line 21): “Relative to the Shindell et al. (2013) study, this study updates the natural non-wetland methane fluxes; applies a different anthropogenic emissions inventory; includes a new land surface model with interactive computation of isoprene and monoterpene emissions; and applies observed ocean boundary conditions. This methodology permits harmonization of the modeled methane mole fractions with contemporary observations, but can potentially misattribute the methane fluxes among the various source categories. Planned chemistry–climate simulations that will make use of the natural methane inventory developed here are specifically designed to investigate perturbations in anthropogenic methane emissions.”

- 3. 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.**

Please see Response to Reviewer #2 Point (3) for an updated description of the optimization methodology. We address the model–measurement comparisons in our response to the next point.

- 4. 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.**

At no place in the manuscript do we refer to the model performance as “excellent.” The reviewer may be confusing the different purposes of global chemistry–climate models (CCMs) versus chemistry–transport models (CTMs). We clarify this distinction here. We work with a global chemistry–climate model that has biases in the climate simulation itself (like all global climate models). Consider that we would actually be slightly more worried if we achieved an almost zero bias or an “excellent” comparison with observations. The ultimate purpose of a CCM is to study feedbacks and linkages between changes in atmospheric composition, radiation, and climate dynamics; there is a focus on understanding the role of interactive Earth system processes in determining the global climate sensitivity. In contrast, CTMs (with “correct meteorology,” e.g., GEOS-Chem) can and must be used for formal optimization procedures to constrain surface emissions. We completely understand that for methane a 1% model error after a formal optimization process in a CTM would be considered substantial. That is not the case for a CCM. Certainly, we could go on and on applying additional iterations of our optimization process to further minimize discrepancies between modeled and measured methane mixing ratios. However, we argue that additional iterations are not justified at this point we have achieved (1) because this framework is for coupled CCM studies and (2) because of the existing limitations and uncertainties in model–measurement comparisons. Indeed, we show that our methane simulation is reasonable and realistic compared to and within the limitations of existing measurement comparisons.

- 5. 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.**

We now use the comma-separated values (CSV) file format for all of the datasets included as part of the Supplementary Information.

#### **Minor comments**

- 6. P2L19: CH<sub>4</sub> is also oxidized by O(1D) in the stratosphere.**

We have added this methane sink to the indicated sentence. The Kirschke et al. (2013) reference, already cited in the original version of the sentence, covers this reaction, so no references were added (Page 2, Line 19): “Additional chemical loss occurs in the stratosphere via reactions with chlorine radicals and excited-state oxygen radicals (O<sup>1</sup>D) (Kirschke et al., 2013; Portmann et al., 2012).”

**7. P5L13. Is version 1.1 a past model version or the new version described by this paper?**

Version 1.1 is the new version of the model that is described in this paper. We have improved the description of the various model versions to make this clear (Page 5, Line 22): “This paper describes the new version 1.1 of ModelE2-YIBs. ModelE2-YIBs version 1.1 refers to the use of interactive methane chemistry and dynamic methane emissions (including application of the final contemporary natural methane flux inventory described in Sect. 3) within the framework of ModelE2-YIBs version 1.0. ModelE2-YIBs version 1.0 refers to YIBs version 1.0 (Yue and Unger, 2015) coupled to the version of ModelE2 described by Schmidt et al. (2014).”

**8. P6L21. Were the LLGHG concentrations prescribed at the surface or also elsewhere?**

The concentrations are prescribed for the non-methane long-lived greenhouse gases (e.g., CO<sub>2</sub>, N<sub>2</sub>O, and CFCs) only in the first model layer (i.e., the layer closest to the surface). We have added the term “surface-level” to this sentence to clarify (Page 7, Line 3): “Prescribed global annual-mean surface-level mixing ratios of the non-methane well-mixed greenhouse gases are likewise from the RCP8.5 scenario (Meinshausen et al., 2011; Riahi et al., 2007): 379.3 ppmv CO<sub>2</sub>, 319.4 ppbv N<sub>2</sub>O, and 793 pptv chlorofluorocarbons (CFCs = CFC-11 + CFC-12).”

**9. P10L9 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.**

We have modified the sentence (Page 11, Line 7): “While the gross magnitude of methane emissions is well constrained, substantial uncertainties remain regarding the partitioning of methane emissions among source categories (Rigby et al., Turner et al., 2017). The interpretation of isotope composition measurements is currently ambiguous and complex (Turner et al., 2017). Prather and Holmes (2017) have recently suggested new approaches to extract more useful information from existing observations by exploiting spatial patterns.”

We already have stated (Page 2, Line 32): “Together, these estimates provide a constraint on the total methane flux into the atmosphere; however, apportionment of this total into contributions from the individual source sectors is highly uncertain (Kirschke et al., 2013; Saunio et al., 2016).”

**10. P17L25. I believe the CH<sub>4</sub> lifetime estimate by Rigby et al. (2013) should supersede Prinn et al. (2005), although the values are similar.**

The methane lifetimes against OH are similar from the two references: 10.6 ± 0.4 years from Rigby et al. (2013) and 10.2 (+0.9, -0.7) years from Prinn et al. (2005). The estimate by Rigby et al. (2013) is based upon the same general modeling framework as is used by Prinn et al. (2005), and therefore can be considered to be an update of the earlier work. We now use the methane lifetime estimate made by Rigby et al. (2013) in place of that made by Prinn et al. (2005):

In Sect. 4.3, we use the Rigby et al. (2013) estimates in place of the Prinn et al. (2005) estimates (Page 22, Line 29): “Further evidence of the model’s skill in capturing methane-relevant processes is found through the close agreement of methane lifetime in the model with that derived from observations. The chemical lifetime of methane in E2005 is  $10.4 \pm 0.1$  years, which is nearly identical to the present-day methane chemical lifetime against OH of  $10.6 \pm 0.4$  years that was derived from OH estimates based on methyl chloroform observations (Rigby et al., 2013). The methane chemical lifetime in the model is only slightly shorter than – but well within the 1 standard deviation range of – a second observation-based estimate that is likewise based on methyl chloroform loss to OH:  $11.2 \pm 1.3$  years for 2010 (Prather et al., 2012). The total lifetime of methane in E2005, taking into account both chemical loss and the soil sink, is  $9.2 \pm 0.04$  years. This closely matches the present-day methyl chloroform-based estimates of total methane lifetime  $9.7 \pm 0.4$  years (Rigby et al., 2013) and  $9.1 \pm 0.9$  years (Prather et al., 2012), derivation of which makes use of estimates of the loss rates for the other minor methyl chloroform and methane sinks. Importantly, the close agreement between the modeled and observation-based methane lifetimes is a strong indicator that the model appropriately captures the processes that control atmospheric methane.”

In the Introduction (Page 2, Line 28): “Methane chemical lifetime is not directly measured in the atmosphere, but has been derived from knowledge of the synthetic compound methyl chloroform ( $\text{CH}_3\text{CCl}_3$ ; Prather et al., 2012; Prinn et al., 2005; Rigby et al., 2013).”

## References

Dlugokencky, E.J., Bruhwiler, L., White, J.W.C., Emmons, L.K., Novelli, P.C., Montzka, S.A., Masarie, K.A., Lang, P.M., Crotwell, A.M., Miller, J.B., and Gatti, L.V.: Observational constraints on recent increases in the atmospheric  $\text{CH}_4$  burden, *Geophys. Res. Lett.*, 36, L18803, doi: 10.1029/2009GL039780, 2009.

Kirschke, S., Bousquet, P., Ciais, P., Saunoy, M., Canadell, J.G., Dlugokencky, E.J., Bergamaschi, P., Bergmann, D., Blake, D.R., Bruhwiler, L., Cameron-Smith, P., Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E.L., Houweling, S., Josse, B., Fraser, P.J., Krummel, P.B., Lamarque, J.-F., Langenfelds, R.L., Le Quéré, C., Naik, V., O’Doherty, S., Palmer, P.I., Pison, I., Plummer, D., Poulter, B., Prinn, R.G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M., Shindell, D.T., Simpson, I.J., Spahni, R., Steele, L.P., Strode, S.A., Sudo, K., Szopa, S., van der Werf, G.R., Voulgarakis, A., van Weele, M., Weiss, R.F., Williams, J.E., and Zeng, G.: Three decades of global methane sources and sinks, *Nat. Geosci.*, 6, 813–823, doi: 10.1038/ngeo1955, 2013.

Meinshausen, M., Smith, S.J., Calvin, K., Daniel, J.S., Kainuma, M.L.T., Lamarque, J.-F., Matsumoto, K., Montzka, S.A., Raper, S.C.B., Riahi, K., Thomson, A., Velders, G.J.M., and van Vuuren, D.P.P.: The RCP greenhouse gas concentrations and their extensions from 1765 to 2300, *Climatic Change*, 109, 213–241, doi: 10.1007/s10584-011-0156-z, 2011.

Portmann, R.W., Daniel, J.S., and Ravishankara, A.R.: Stratospheric ozone depletion due to nitrous oxide: Influences of other gases, *Philos. T. Roy. Soc. B*, 367, 1256–1264, doi: 10.1098/rstb.2011.0377, 2012.

Prather, M.J. and Holmes, C.D.: Overexplaining or underexplaining methane’s role in climate change, *P. Natl. Acad. Sci. USA*, 114, 5324–5326, doi: 10.1073/pnas.1704884114, 2017.

Prather, M.J., Holmes, C.D., and Hsu, J.: Reactive greenhouse gas scenarios: Systematic exploration of uncertainties and the role of atmospheric chemistry, *Geophys. Res. Lett.*, 39, L09803, doi: 10.1029/2012GL051440, 2012.

Prinn, R.G., Huang, J., Weiss, R.F., Cunnold, D.M., Fraser, P.J., Simmonds, P.G., McCulloch, A., Harth, C., Reimann, S., Salameh, P., O'Doherty, S., Wang, R.H.J., Porter, L.W., Miller, B.R., and Krummel, P.B.: Evidence for variability of atmospheric hydroxyl radicals over the past quarter century, *Geophys. Res. Lett.*, 32, L07809, doi: 10.1029/2004GL022228, 2005.

Riahi, K., Grübler, A., and Nakicenovic, N.: Scenarios of long-term socio-economic and environmental development under climate stabilization, *Technol. Forecast. Soc.*, 74, 887–935, doi: 10.1016/j.techfore.2006.05.026, 2007.

Rigby, M., Montzka, S.A., Prinn, R.G., White, J.W.C., Young, D., O'Doherty, S., Lunt, M.F., Ganesan, A.L., Manning, A.J., Simmonds, P.G., Salameh, P.K., Harth, C.M., Mühle, J., Weiss, R.F., Fraser, P.J., Steele, L.P., Krummel, P.B., McCulloch, A., and Park, S.: Role of atmospheric oxidation in recent methane growth, *P. Natl. Acad. Sci. USA*, 114, 5373–5377, doi: 10.1073/pnas.1616426114, 2017.

Rigby, M., Prinn, R.G., O'Doherty, S., Montzka, S.A., McCulloch, A., Harth, C.M., Mühle, J., Salameh, P.K., Weiss, R.F., Young, D., Simmonds, P.G., Hall, B.D., Dutton, G.S., Nance, D., Mondeel, D.J., Elkins, J.W., Krummel, P.B., Steele, L.P., and Fraser, P.J.: Re-evaluation of the lifetimes of the major CFCs and CH<sub>3</sub>CCl<sub>3</sub> using atmospheric trends, *Atmos. Chem. Phys.*, 13, 2691–2702, doi: 10.5194/acp-13-2691-2013, 2013.

Saunio, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J.G., Dlugokencky, E.J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F.N., Castaldi, S., Jackson, R.B., Alexe, M., Arora, V.K., Beerling, D.J., Bergamaschi, P., Blake, D.R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Höglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H.-S., Kleinen, T., Krummel, P., Lamarque, J.-F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K.C., Marshall, J., Melton, J.R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F.-J.W., Patra, P.K., Peng, C., Peng, S., Peters, G.P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W.J., Saito, M., Santini, M., Schroeder, R., Simpson, I.J., Spahni, R., Steele, P., Takizawa, A., Thornton, B.F., Tian, H., Tohjima, Y., Viovy, N., Voulgarakis, A., van Weele, M., van der Werf, G.R., Weiss, R., Wiedinmyer, C., Wilton, D.J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z., Zhu, Q.: The global methane budget 2000–2012, *Earth Syst. Sci. Data*, 8, 697–751, doi: 10.5194/essd-8-697-2016, 2016.

Schmidt, G.A., Kelley, M., Nazarenko, L., Ruedy, R., Russell, G.L., Aleinov, I., Bauer, M., Bauer, S.E., Bhat, M.K., Bleck, R., Canuto, V., Chen, Y.-H., Cheng, Y., Clune, T.L., Del Genio, A., de Fainchtein, R., Faluvegi, G., Hansen, J.E., Healy, R.J., Kiang, N.Y., Koch, D., Lacis, A.A., LeGrande, A.N., Lerner, J., Lo, K.K., Matthews, E.E., Menon, S., Miller, R.L., Oinas, V., Olosio, A.O., Perlwitz, J.P., Puma, M.J., Putman, W.M., Rind, D., Romanou, A., Sato, M., Shindell, D.T., Sun, S., Syed, R.A., Tausnev, N., Tsigaridis, K., Unger, N., Voulgarakis, A., Yao, M.-S., and Zhang, J.: Configuration and assessment of the GISS ModelE2 contributions to the CMIP5 archive, *J. Adv. Model. Earth Sy.*, 6, 141–184, doi: 10.1002/2013MS000265, 2014.

Shindell, D.T., Pechony, O., Voulgarakis, A., Faluvegi, G., Nazarenko, L., Lamarque, J.-F., Bowman, K., Milly, G., Kovari, B., Ruedy, R., and Schmidt, G.A.: Interactive ozone and methane chemistry in GISS-E2

historical and future climate simulations, *Atmos. Chem. Phys.*, 13, 2653–2689, doi: 10.5194/acp-13-2653-2013, 2013.

Turner, A.J., Frankenberg, C., Wennberg, P.O., and Jacob, D.J.: Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl, *P. Natl. Acad. Sci. USA*, 114, 5367–5372, doi: 10.1073/pnas.1616020114, 2017.

Yue, X. and Unger, N.: The Yale Interactive terrestrial Biosphere model version 1.0: Description, evaluation and implementation into NASA GISS ModelE2, *Geosci. Model Dev.*, 8, 2399–2417, doi: 10.5194/gmd-8-2399-2015, 2015.