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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric impact of methane emissions from clathrates in the Arctic Region" *by* S. Bhattacharyya et al.

S. Bhattacharyya et al.

bhattacharyy1@llnl.gov

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Response to Reviewer2's comments:

Review of the paper by Bhattacharyya et al. entitled: Tropospheric impact of methane emissions from clathrates in the Arctic Region

.This paper by Bhattacharyya et al. presents and analyse the impact of potential additional emissions from hydrates on methane concentrations, temperature, ozone, and precipitation. To do so, the authors compare 2 simulations of the community Earth system model CESM model, which includes a fully interactive physical ocean and a fast atmospheric chemistry mechanism with explicit methane emissions. One simulation includes additional CH4 emissions to mimic possible future hydrates emissions. The

paper addresses the scientific question of the impact of an increase of CH4 emissions in the Arctic region in the context of a changing climate. The paper is well organized and is pleasant to read, except for the conclusion (see specific comments)

.General comments -My main concern is about the hypothesis of additional emission of 139 TgCH4/yr advanced by the authors. I find the section justifying this choice rather poor. Few references are cited, both for CH4 emissions at seabed and for the fraction going to the atmosphere. The fraction going to the atmosphere is from 1% to 100% but can the authors narrow a bit this range based on litterature ? Is it more 1% or more 100% ? What is the extent in the case of 100% ? Same questions for the amount emitted at the seabed ? A 22% perturbation of the CH4 cycle is a large one. I kind of agree with the authors that it could also reflect possible perturbation from other sources (e.g. permafrost). Indeed, the precise location of CH4 emission perturbation in the arctic is probably less critical than at lower latitudes because of the fast horizontal transport and because CH4 has a 10y lifetime. However, I think the paper should be presented more in this sense then and not only focusing on (very uncertain) hydrate emissions. In a warming climate, additional emissions from permafrost and wetlands are likely to happen. In other words I am not convinced of a 139TgCH4/yr due to hydrates only but including all possible sources it becomes a more “plausible” scenario. Anyway, this part has to be reinforced largely (see also specific comments).

We would like to thank the reviewer for the in depth review and insightful comments on the paper. The study reported in this paper is more of a sensitivity analysis to assess whether a large clathrate event is even important, rather than modeling a particular scenario. Also, see more extensive comments in our response to review #1. The revised text is: The methane emissions that we are using are the first physically meaningful calculations of dissociation and methane flux, based on an estimated (high-end) ocean warming scenario. The predicted emissions are not a simple function of the warming. Rather, in response to gradual warming at the sea-floor there is no significant methane release for a few decades while heat propagates into the sediment, hydrates dissoci-

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ate, and methane is transported up to the seafloor. There is then an abrupt increase in the emission rate as gas-phase methane reaches the seafloor, followed by a slow decline (see Figures 1 (a,b) of the manuscript). Note also the log scale in Figure 1, reflecting that only the shallowest hydrates contribute significantly to the total flux. The net flux, can thus be approximated by a step function. The methane emission rate of 139 Tg[CH4]/yr was the outcome of one of the higher emission scenarios (100-yr linear increases of +5 C per century at 350 m, +3 C per century at 400-600 m, and +1 C per century below 600 m simulated by Reagan et al. (Reagan et al., 2011a)). Because the emissions from different temperature scenarios scaled sub-linearly, the exact temperature scenario is less important than might be expected. For our steady-state simulation with enhanced Arctic emissions we added 100% (Elliott et al., 2011b) of the seafloor flux to the regular atmospheric methane sources in our present-day control. Note that the emissions are located only in three representative grid boxes,each of about 2.5 degree x 2.5 degree in areal extent, namely in the Barents Sea, Canadian Archipelago, and the sea of Okhotsk.

The authors agree that although the sensitivity analysis of methane emissions from hydrate dissociation is reported in this paper, that the atmospheric impacts of methane emission would be similar for other high latitude methane emission sources and in that sense the results reported here have more general implications.

.-Abstract does not reflect the quantitative results of the paper. Please rewrite to be more precise about the results obtained in the paper

The abstract has now been changed to indicate the quantitative results of the paper as below:

A highly potent greenhouse gas, methane, is locked in the solid phase as ice-like deposits called clathrates in both ocean sediments and underneath permafrost regions. Clathrates contain a small amount of methane trapped in cages made of water molecules (Sloan and Koh, 2008). Clathrates are stable under high pressures and low

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temperatures. In a warming climate, increases in ocean temperatures could lead to dissociation of the clathrates and release methane into the ocean and subsequently the atmosphere. This is of particular importance in the shallow parts of the Arctic Ocean, since clathrates are expected to start outgassing abruptly at depths of around 300 m. In this paper, we present a sensitivity analysis of the atmospheric impact of methane emission in the Arctic by simulating a plausible release of clathrates in the Arctic (based on a state-of-the-art ocean sediment model that enables the first physically meaningful calculations of dissociation and methane flux, using a synthetic (high-end) ocean warming scenario) and comparing it to the present-day control simulations using the Community Earth System Model (CESM1). The CESM model includes a fully interactive physical ocean and we added a fast atmospheric chemistry mechanism that represents methane as a fully interactive tracer (with emissions rather than concentration boundary conditions) along with the main chemical reactions for methane, ozone, and nitrous nitrogen oxides. The results show that such Arctic clathrate emissions increase global methane concentrations by an average of 38%, but non-uniformly; the increase in surface air temperature is statistically significant almost everywhere (signal-to-noise ratio greater than 3); and that surface ozone concentrations increase by more than 10% globally, particularly in polluted regions. We also find that the interannual variability in surface methane and ozone increases.

.-Sentences in the conclusions are too long. They have to be shortened and sometimes clarified. In the main text, some of the sentences can also be shortened and clarified.

Conclusion has been modified accordingly.

(see specific comments).

Specific comments 1. p26478-l22 : define long lifetime

($\sim$10 years) incorporated in the revised text.

2. p26479-l6 : considering the large range I suggest to remove “precise”

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"Precise" replaced by "Such" in the revised text.

3. p26479-l8 : why separating "bubbles" Explain or modify. 4. It is important to have an estimate of possible methane emission from 'bubbling' since it is one of the most plausible mechanisms through which methane locked in the clathrates in the ocean sediment can make its way up to the surface of the ocean without being consumed by methanotrophs and be released to the atmosphere. (incorporated in the text.)

5. p26480-l14 : "large increases in atmospheric methane concentration" : please give orders of magnitude and appreciation of the spatial extension of such increases (probably very local around the bubbling)

We have modified the text as follows: Extensive venting of methane from the East Siberian Arctic Shelf has also been observed (Shakhova et al., 2010), with bubbles reaching the atmosphere through the shallow ocean and producing large increases (of the order of 8TgC-CH4 per year) in atmospheric methane emissions over the East Siberian Arctic Shelf area of 2.1x106 km2, as reported by Shakhova et al., 2010.

6. p26482-l9 : "with a uniform initial clathrate saturation of 0.03, reflecting the high end of the estimated global average saturation for such deposits" : unclear sentence (to me). Please be more clear.

We have modified the text as follows: The code was used to model disperse, low-saturation (stratigraphic) deposits with a uniform initial hydrate distribution at 3% of empty pore space, reflecting the high end of the estimated global average saturation for such deposits.

7. p26482-l20-25: the origin of these scenario is unclear to me. Why 5/3/1_ ? Adding 139 TgCH4/y is a huge change compared to the 500-600 TgCH4/yr global emissions. Please give the present global emissions in the for comparison. This is a 20% change ! Where does this number come from ? It needs justification and/or References. Also, can you relate this number with your figure 1.

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We have modified the text as follows: Recent global estimates for methane are about 503-610 Tg(CH4)/yr, (see Table 7.6 of IPCC AR4: Solomon et al., 2007). The details of the calculation of the emission scenario are detailed in Reagan et al. 2011a; 2011b and our paper (we have revised the description to be easier to understand – see above)

8. P26483-l9-12 : The hypothesis of 100% transmission from the seafloor to the atmosphere seems a bit extreme to me. Arguing that error in the transmission could be compensated by additional emisssion at the seabed cant be turned back : what if seafloor emissions are overestimated already ? This part needs more work : 1/1% to 100% range leaves large uncertainties. The authors have to give more literature about this to justify their approach of a maximum transmission. A 100% transmission implies is only credible for bubbling probably, as diffusion implies loss by oxidation. What is the part of bubbling in emission reaching the atmosphere ? 2//The authors can use maximum emissions to the atmosphere (large seabed emissions + 100% transmission) but they have to write clearly that this is a 'maximum' scenario.

We certainly do not mean to claim that 100% transmission is the most likely scenario, and indeed this is a 'maximum' scenario for ocean transmission. However, as discussed in greater detail in our response to review #1, this was designed as a sensitivity study so our main conclusions about the non-uniform increase in methane concentration and changes in variability should be largely independent of the chosen emission rate. As the reviewer notes, other high latitude emission sources are likely to produce similar effects, so including those in the emission estimate will make the scenario more plausible for the 21st century. It is also possible that the sea-floor clathrate emission will be larger than we estimate (eg due to higher pore saturation than we assume). We are modifying the manuscript to make this clearer.

9. P26483-l17 : add somewhere in the text the duration of the run and the machine used. 10.

All the simulations were run on the machine 'Franklin' at National Energy Research

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Scientific Computing (NERSC) at Berkeley. Corrections added in the text of the manuscript. The durations for AE and Control were provided on p26484-line12, and are 449 yrs and 420yrs, respectively.

11. P26483-l21 : add a reference for the present-day scenario of methane emissions. What is the total of surface emissions ? P26484-l6 : 629 TgCH4/yr is the total emissions ? total sink ? Please be more precise. If total emissions, this is larger than current estimates more around 550 TgCH4/yr. Please justify more what source causes this large number. Is this number kept constant all along the 12. run ?

The recent estimates for global annual emission of methane as documented in IPCC AR 4 report (see Table 7.6 of IPCC AR4: Solomon et al., 2007) have the range 503-610 Tg(CH4)/yr, however in order to match the present day methane surface concentration of 1.79*10-6 mol/mol as seen from recent observations (Rigby et al. 2008, Dlugokencky et al., 2009), we had to tweak the total emissions in the model to 629 Tg(CH4)/yr. (incorporated in the text.)

The methane emissions we used were the default emissions available in the version of CESM that we used. We have not been able to determine the exact provenance of those emissions, however, we are now adding a figure to the manuscript that shows a comparison of the control simulation against atmospheric methane observations, which shows that the control simulation is reasonable, albeit with a slightly high inter-hemispheric gradient. Because the simulation is in steady-state, the total sink must be equal to the total emissions.

Yes this number is kept constant all along the run. The idea here is that once the clathrate emissions start, they continue to emit at a roughly steady rate over a long time (see figure 1). It is also necessary for this to be a steady-state run in order to achieve sufficient signal-to-noise ratio in the results, since the cost of achieving this with a large ensemble of trend simulations would be prohibitive (see response to review #1).

13. What would be the impact of considering an accidental event (increasing and then

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decreasing pulse of additional emissions ?

An event like an oil-spill accident, or an underwater slide depressuring a sediment layer containing clathrates (all of which have been observed), is quite probable, so it is quite possible for methane for outgas for some time due to a localised event (localised both in space and time). We speculate that the spatial distribution and increased variability we observed in our stead-state simulations would still apply, and probably be even more prominent. However we note from our spin-ups (and the methane pulse lifetime) that it takes a couple of decades for the methane concentration to get close to its steady-state amplitude, so a lot will depend on the precise magnitude and duration of the emission pulse.

14. P26684-l15 : did you add point emissions in 3 pixels of the model? In three zones covering several pixels ? Please be more precise on the method to inject methane in the atmosphere. Arrows on figure 1a are not visible.

The emissions were added in 3 specific grid boxes of the emission grid, which happened to be a T42 rectangular Gaussian grid. The emission grid then gets interpolated by CESM bilinearly onto the model grid, which was a 1.9x2.5 degree regular rectangular grid. Because the bilinear interpolation does not conserve total emissions, the emission values we provide in the manuscript are the values after interpolation on to the model grid.

Figure 2a shows the specific locations marked with red arrows. The arrows are very clear on our version of the document, so we assume this must be a glitch in the version the reviewer received. We will check that the arrows show up correctly in the proofs.

15. P26684-l21 : I would say more that this is an “extreme” scenario. Else the authors need to justify the fact that this is plausible with more references (see before). 16. Please see comments above and our response to review #1.

17. P26485 – l22 : OH is also controlled by CO and VOCs. How do you treat these

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gases in your modelling exercise? 18. The atmospheric chemistry mechanism used in the model already accounts for OH reacting with CO. VOCs are not included in the model. However, we have confirmed that the response of OH to a methane increase is similar to the response of a more complete chemical mechanism that does include higher hydrocarbons.

19. P26486 – l1-4 : What do you mean ? Unclear sentence, please rephrase to be less technical.

Our statement was correct, but confusing and unnecessary, so we are deleting it. See also review #1.

20. P26486 – l28 : "but the SNR is still generally greater than 3 over the poles" This statement is not clear for the color scale of figure 3d

Figure 3d is modified to indicate regions of SNR greater than 3.

21. P26487 – l7 : Would it be useful to have"coloured" methane chemical equations to partition stratosphere from troposphere for instance ? At least a suggestion of what to do to understand what happen here seems necessary

This is a good suggestion, and we can add such a comment to the text. Unfortunately, rerunning the code with this change would take too long for this manuscript.

22. P26487 – l18 : 39% or 38% ? 23. 38%

24. P26488 – l5-10 : Can you be more precise on the patterns high lats vs low lats ? It seems also that organized patterns appear at mid latitudes for precipitation. Can it be related to changes in atmospheric dynamics? 25.

Our results are in general agreement with the climate projections from CMIP3 reported in IPCC AR4, where warming scenarios generally cause mean precipitation to increase in tropical monsoon regimes and the tropical Pacific, with general decreases in the subtropics, and increases at high latitudes. The precise reasons do not seem to be

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well understood, and the detailed behavior is not totally consistent across the CMIP3 models.

26. P26488 – l24 : Recall in brackets what shows figure 2C. 27. Figure 2C in the above sentence should be changed to figure 2d. Inserted in brackets next to figure 2d (which shows the standard deviation of the difference in methane surface concentration between AE and C).

28. P28490 – l4 : Which regions are still significant for ozone ? Are southern regions still significant ? Why then this southern sensitivity ? Again even if no clear cause appear, ideas to diagnose why the observed changes occur should be given by the authors.

Although many polluted regions show increase in ozone variability, the greatest variability is still seen in the southern ocean. This is likely due to the fact that this region is generally a less-polluted region, so the ratio of CH4 to CO will be higher than in other regions. Hence, the relative importance of CO to the ozone chemistry in such regions will be less, and the importance of CH4 will be proportionally greater than elsewhere, resulting in greater sensitivity of ozone concentrations to methane emissions in less-polluted regions. Although our mechanism does not include higher hydrocarbons, a similar argument should apply to them too.

29. P28490 – l26 : What would be the impact of emitting methane elsewhere in the Arctic regions (permafrost, gas, wetlands: : :) ? One can assume that the fast horizontal transport at high latitudes may limit the impact of the location of emissions. This indeed makes your study rather generic. This point may be discussed further in the discussion.

We agree, and this broadens the importance our work. We have modified the text as follows:

Indeed our study can be considered as studying the impact of a generic methane emis-

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sion in the Arctic using an Earth System model with interactive atmospheric chemistry and a full ocean model. The importance of the precise emission locations can be seen in figure 2a by the intense peaks in methane concentration around the emission locations, but as can be seen in figures 2c and 2d, away from the emission locations the impact is more zonally symmetric, indicating that the precise emission location is not important. This is presumably because fast horizontal transport at high latitudes limits the importance of the precise emission location.

30. P26491 – l6-end : the sentences of the conclusion are too long making them hard to read and understand. Please rephrase with shorter and clearer statements. Figures : Character size is too small on axes.

Corrections done.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 26477, 2012.