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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of the global atmospheric methane budget using ECHAM-MOZ simulations for present-day, pre-industrial time and the Last Glacial Maximum" *by* A. Basu et al.

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Referee 1 General comments:

1) The reviewer commented that in this paper, the good agreement between simulated result and the ice core records or station observation is based on the fact that OH remains unchanged over long time, which is not clear since Montzka et al., study is done over a short time.

Answer: We would like to state that in this paper, the OH variability study is carried out in two phases, first for the transition from LGM to PI and the other from PI to PD. The

conclusion that OH did not change much over the simulated period from LGM to PI and to PD is based up on the recent chemistry findings by Hofzumahaus et al., 2009, Peeters et al., 2010 and Taraborelli et al., 2012 that BVOC exert much less control on OH concentration than assumed in earlier paleo chemistry studies. Assuming no changes certainly yields an upper limit of OH and the truth is probably somewhere in the middle between the 25% change estimated by MOZART chemistry simulation (which we ran as a sensitivity case) and the no-change scenario from our study. As discussed in section 3.4, paragraph 2 and in the figure 1 that the no-change scenario is derived if we ignore the BVOC-OH oxidation chemistry following the above findings.

As far as the question of OH changes from PI to present day is concerned, , the Naik et al. paper which encompasses multi model comparison ACCMIP study, also found that the net change is minimal (-0.6$\pm$8.8%). However they considered that BVOCs contributed OH loss up to (3.1$\pm$3%). Going by this, we have considered two scenarios, one with the unchanged OH and the other with a maximum possible change of OH by 6%.

2) The reviewer does not think that the distribution of PD wetland emission is in agreement with studies that show most of the emission occurring in the tropics and high latitudes.

Answer: Please find attached the figure with wetland methane emission for present day. It clearly shows a high emission over tropics and north compared to mid latitudes. As suggested, we would add some details on the LGM climates in section 2.2

#Referee 1 specific comments:

i) Comment 1, L5-7: "analyzing methane sources and sinks at each of these climatic periods" refers analysis at PD, PI and LGM, the three individual climatic regimes, and not the period between LGM and PI or PI and modern. May be the phrase "period" is a bit confusing. We change it to "each of these three climatic regimes". The present day simulation is done unless it reaches the steady state which is taken as a global mean

methane concentration between 1999 and 2003, when it attained a steady state.

ii) Comment 2, P3195, L6: I do not understand why GWP comes here?

iii) Comment 3, P3195, L24-25: This question is answered in the General comment section. However, we are not claiming here that the OH is assumed to be constant over a long period. We referred to Montzka et al. study here since it shows less variability and uncertainty in methane sink over recent years than shown in previous studies. It indicates that sinks are relatively well constrained compared to sources which are plagued by large uncertainty.

iv) Comment 4, P3198, L1: Gedney et al. is the one which we referred here as original publication. We changed “as in the original publication” to “as in Gedney et al.”

v) Comment 5, P3198, L2-7: No, we did not use any inversion. Rather in our bottom-up approach, we carried out a sensitivity study by varying KCH4 and targeting the minimum error between model and observed methane seasonality while preserving the seasonality of the emission sources as they were provided. So starting from an arbitrary value of KCH4 , it is calibrated to a fixed value. This approach is similar to Gedney’s and discussed briefly in that paper.

vi) Comment 6, P3199, L1-4: Indeed it is re-oxidation of methane in soil by microbes. The unit used to express uptake must not be direct influx values because deposition obeys first order kinetics for atmospheric gas concentration. We should change the sentence to “we also included the soil uptake of CH4 by microbes which is expressed by a linear loss rate due to dey deposition”

vii) Comment 7, P3199, L20: Indeed so. We change it to “last 50 years”.

viii) Comment 8, P3200, L11-15: In the sensitivity test to optimize wetland emissions and produce the best model-observation seasonal match , we figured out that even a small modification in the hemispherical distribution of biomass burning proves effective to come up with an improved seasonal match, particularly over farthest south latitudinal

stations. Although a better seasonal match at those stations can also be achieved by modifying other sources e.g. eneteric fermentation and ruminants, it occurs at the expense of diminishing the seasonality at other stations. So we considered scaling biomass burning methane emissions instead of a multiple other sources.

ix) Comment9, P3203, L12: Figure 1 shows that during LGM, a reduced BVOC emission and a reduced methane flux, both individually affects in 26% OH increment. We change the structure of the sentence for better understanding.

x) Comment 10, P3204, L10-12: I attach the present day wetland emission map to actually show that we found the same as the reviewer expects.

xi) Comment 11, P3205, L8-10: Yes, the number of inter-hemispheric gradient is obtained from the final simulation carried out with an optimized methane source, which includes tuning of biomass burning emissions.

xii) Comment 12, P3205, L25: I am not sure if I understand the point correctly. We see in Figure 7, the annual maximum at the ocean station (66deg N, 2 deg E) in the month of October, which is quite different from the observation. I suppose that it occurs from the transportation of methane from continental source, among which one or more is overestimated in the model.

xiii) Comment 13, P3208, L3-5: Instead of writing that “ Sunda shelf region yields more that 450 nmol mol-1 resulting from the large wetland emissions from coastal shelves”, We change it to “ the methane concentration surrounding the Sunda Shelf region is found to higher than 450 nmol which could be influenced by high wetland emission from exposed shelves.”

xiv) Comment 14, P3208, L13-14: We do not really mean here that LGM wetland emission has to be higher than PI wetland emission in order to match the model methane to the ice core records. Here we infer that the total LGM methane emission (not the wetlands alone) should be 96 and 120Tg/yr respectively for a constant OH scenario and a

25% higher scenario, to match the model data with observation. In the next sentence, it is meant that if the uncertainty in total budget is compensated by wetlands alone, then it should either be reduced to 52 Tg/year or increased to 76 Tg/year for these two scenarios. Likewise, for PI, the wetland strength should be reduced in the range of 99-88 Tg/yr. Now since in our model set up, the strength of the non-wetland sources both in PI and LGM, are already in their lowest ends, I put the onus on wetlands alone.

xv) Comment 15, P3208, L21-22: Indeed, there is a contribution of wetlands (it grew by 17 Tg/year) behind the growth of atmospheric methane burden from PI to PD. But this is insignificant compared to the role of anthropogenic emission, which grew by almost 300 Tg/year between PI and PD (ref. Table 1).

xvi) Comment 16, P3209, L16-18: We carried out the sensitivity test with a 6% OH variability in PI (as found by Naik et al.) as given in section 3.4. The results are also discussed in section 5.

xvii) Comment 17, P3209, L16-18: Indeed yes. You are right. We change it from PD to PI in the text.

#Referee 2 General comments:

i) In this paper, we did not discuss in detail about the CARAIB simulation since its output is used offline to estimate potential wetlands. Here we provide the details, and if the reviewer feels it to be essential, we can include it in the subsection 2.1 of wetland methane modeling. Climatic fields to force the CARAIB model for present day climate are monthly mean observed fields from New et al. (2002) over the period 1931-1960. The same for LGM is obtained by adding the anomalies obtained from AGCM models from Paleo-modelling inter-comparison project to the present day climate. For the LGM, the climatic fields were from a simulation of ECHAM4 provided by Stephan Lorenz. In the simulation of ECHAM4, SST was fixed to CLIMAP northward of 30°N and southward of 30°S, between 30°S and 30°N it was fixed to CLIMAP minus 3°C. The soil texture data from Zobler (1986) were used for present simulation. For LGM, the same

was used and extrapolated to emerged shelf. There is no organic peat soils parameterized in CARAIB. Texture is only sand, silt and clay, using the old parameterisation from Saxton et al. In CARAIB, an improved bucket model of the hydrological cycle with a single soil pool is used to estimate the soil moisture content plus another reservoir for the snow. It is not exactly a bucket model, because bucket models fill up to field capacity, where any excess water is evacuated as runoff. Here, we calculate drainage rate (hydraulic conductivity) as a function of soil texture and soil water amount. However, soil water can increase above field capacity (contrary to bucket models) and even reach saturation under high precipitation events (Francois et al., 2011). ETOPO 5 topography data for present day and its modification by adding 105 m for LGM are accounted in CARAIB. The reviewer wants to know if hydrology depends on topography. The answer is no. Evapotranspiration is depending on atmospheric pressure, which depends on elevation, but this is the only dependence on elevation. Runoff or drainage does not depend on slope and there is no connection between the pixels in the hydrological module. Regarding the reviewer" question about the spin up years, we would like to mention that in the initialisation phase, vegetation is typically integrated over a few hundred years (about 300 years) and the soil is equilibrated with the litter produced by this vegetation. The number of years needed to reach this equilibrium varies from one pixel to the other (it is quicker in warm climates and longer in cold ones). The maximum number of years is 4000 years. We have used potential vegetation calculated with CARAIB both for the presnt and the LGM with 15 PFT version. PFTS were described by Galy et al. (2008). The CO2 forcing for LGM is 200 ppmv for the LGM and 360 ppmv for the present.

ii) The reviewer asked to provide details on the dataset used for LGM and PI/PD. We agree that not all the technical details regarding the dataset are given in sub-section 2.1. There we discussed more in detail the parameterization method. As pointed out by the referee, we can add the details, which follow next, in the same section. As answer to the questions, we have used the surface slope map, originally derived from ETOPO5 data of surface elevation (http://www.fao.org/nr/land) for both the PI and PD wetland

model. For LGM, except for the sea level retreat and the resulting exposed shelf areas there is no change on the mainland terrain pattern. So, we used the same surface slope data and the same condition over the mainland for LGM wetland parameterization. They were interpolated to a coarser grid of 30 minute by area conservational approach, in order to be sure that it would not impact the wetland fraction per grid. In fact we have checked the total wetland areas to be same post interpolation. However, one important change occurred during LGM was the expose of shelves during LGM for ocean retreat and the sea-level change. For that, we added 105 m to the elevation data obtained from ETOPO5. Since the shelf areas are flat enough, we do not pose any condition for surface slope for wetland estimation. The only condition comes from soil wetness.

iii) As asked by the reviewer, here we provide in brief the details of the MOZART simulation used to produce Figure 1. In this study, the present day tropospheric chemistry (Lamarque et al., 2005) is contrasted with an investigation of the chemistry during LGM. The present day CTRL run is forced with AMIP SST while the LGM run is forced with GLAMAP 2000 SSTs and sea-ice cover data. The LGM input fields of vegetation, emission and chemical transport modeling are adapted using data from Peltier(1994) to accommodate different land sea distribution. The LPJ DGVM is used to estimate LGM vegetation. LPJ is driven by climatic fields from 15 year ECHAM output for PD and LGM. The biogenic emission modeling is based on the algorithm from Guenther et al. It parameterizes various factor dependence (light, temperature, leaf area etc.) on biogenic emission. The global fire model GLOBFIRM in LPJ was used to simulate fire emissions. For Albedo, the approach of Laepple et al. (2005) was used. As per as chemical boundary conditions are concerned, for CTRL run, the standard upper boundary conditions are used. For LGM, the same were extracted from a (PI) MOZART-3/WACCM1b IPCC run. Using the PI conditions for LGM is justifiable as previous 2D studies of the atmospheric chemistry during LGM (Martinerie et al., 1995) did not find a significant stratospheric ozone change between LGM and PI. For CTRL run, the initial conditions from climatological present day MOZART run were used. For LGM, a two years spin up run was integrated using the same present day initial conditions.

The output of this spinup is used as initial conditions for all LGM experiments. Overall, MOZART-4.2 which is used in the simulation with a T21 horizontal resolution and 19 vertical levels , provides distribution of 97 chemical constituents between surface and stratosphere. The evaluation of species is calculated with a time step of 20 minutes. For each experiment, the model is run for 1.5 years, using the last 12 months for analysis. For the stratosphere troposphere exchange of ozone, the SYNOZ (Mclinden et al., 2000) parameterization is used.

iv) We agree to the reviewer that we need to highlight the change of wetland methane from LGM to PI instead of PD in the conclusion section. We also agree that this is important to discuss since our estimation of 62% increase is within the range of Weber et al., 2010 (57 to 64%). We should incorporate the said changes in the revised version.

v) As pointed out by the reviewer we should include the statement in the discussion section that apart from reduction of wetland sources, the non-wetland sources also had an important role for the reduction of atmospheric methane concentration. I also agree that the findings of Singarayer et al. (2011) where they found the early Holocene rise in methane occurred due to orbital configuration changes and not the anthropogenic emissions, should be mentioned in the text. It certainly needs a discussion as it gives a different perspective than the Ruddiman et al.

#Referee 2 specific comments:

i) Comment 1, Page 3197, line 14: Regarding the regional boundary of selection of different parameter values for wetland methane parameterization, we adopted a simple and straightforward approach to use fixed latitude boundaries. Note that this applies consistently to KCH4 and the threshold values of soil wetness. Concerning the change of the latitudinal boundary over time we acknowledge the referee's point but argue that the vast majority of today's tropical wetlands would also have shown tropical characteristics during the LGM. So, the choice of 30deg N as a regional boundary is a sensible choice across climatic regimes.

ii) Comment 2, Page 3197, line 22: The soil temperature in the CH4 model is used interactively with ECHAM. Here the temperature profile within the soil is calculated from the thermal diffusion equation connecting the volumetric heat capacity, thermal flux and conductivity. The soil temperature is estimated as for the numerical solution, the upper 10 m of the soil are divided into 5 unevenly spaced layers and used in the CH4 model.

iii) Comment3, Page 3199, line 12-15: Yes, surface potential and snow depth as obtained from COSMOS simulation was prescribed to the ECHAM model for LGM simulation. The global temperature change between LGM and PD is 4.7 deg C.

iv) Comment4, Page 3199, line 23-25: It is not very important, since the model has been run sufficiently long to rule out any influence from the initial concentrations. This piece of information only serves to say that we don't begin with present-day methane levels for a LGM simulation.

v) Comment 5, Page 3203, line 1: We will include the references for the changes in the text which are Martinerie et al., Kaplan et al., and Adams et al.

vi) Comment 6, Page 3203, line 6: We shall remove 'personal communication' from this line.

vii) Comment 7, Page 3206, line 6: We shall remove 'in' after 'further'.

viii) Comment 8, Page 3206, line 26: Soil carbon does not have a role in the seasonal variation since it is constant over the year unlike soil temperature, which shows a strong seasonal change.

ix) Comment 9, Page 3207, line 21-23: Indeed the non wetland sources for PI together contribute 81 Tg per year, but we have to admit that individually they are pretty low compared to wetland strength. In fact, the anthropogenic emissions constituted of ruminants, rice and biomass burning emissions are significantly low compared to present day, thus are less uncertain. The choice of wild animal emission to be higher

than present day also is not irrational. On the other hand, the PI wetland methane budget is more uncertain because the effect of factors like land use change etc. are not taken into account in this study. So the most logical explanation in order to obtain a low PI methane concentration is a lower wetland emission. The other possibility of all of the non-wetland sources to be consistently reduced is not very rational. That is why, we propose a low wetland emission.

x) Comment 10, Page 3208, line 8: We agree that we should update the inter-polar gradient value with Baumgartner paper, which estimates the range between 13. 4 ppbv and 43.5 ppbv.

xi) Comment 11, Page 3209, line 20: We should mention in the text that they are model inferred values of wetland at PI OH sensitivity test.

xii) Comment 12, Page 3210, line 15-19: We should change the paragraph based on LGM to PI change.

xiii) Comment 12, Page 3210, line 3-5: As discussed, Baumgartner value of the inter-polar gradient should be included in this section.

xiv) Comment 13, Page 3211, line 3: Although both Levine et al. and us argue about constant OH, Levine et al. has found OH buffering has a little effect on methane lifetime. On the other hand, we argue that it should have a large effect.

xv) Comment 14, Page 3211, line 2: Levine et al. included the OH recycling mechanism of Peeters et al., 2009 to the existing Mainz isoprene mechanism of Poschl et al., 2000 which is coupled with climate. We can include it in the text.

xvi) Comment 15, Table 1: We argue that although, the LGM soil temperature is lower which inhibits methane emission from termites even in the tropics, still there is a possibility that due to largely non-human intervened tropical plain land, the presence of termites were higher than present.

xvii) Comment 16, Figure 1: We should give a proper explanation of the method.

xviii) Comment 17, Figure 9: Apart from parts in tropics over South East Asia, there are not much wetlands produced on the coastal shelves. We agree that changing the present day coastline to show the LGM shelves would be interesting, but would be difficult to implement.

xix) Comment 18: Although we agree that a comparative analysis of wetland distribution among our study and WETCHIMP would be interesting, still we would like to emphasize the scope and primary idea of the study whish is to address the question of methane budget analysis over various climates. May be we should make the point clearly in the introduction section. Thus, we wanted to build a wetland methane parameterization based exclusively on CARAIB DGVM, which involves both the potential wetland mapping and methane estimation at different climates. We do not find the relevance to use readymade wetland map for present day methane budget analysis, as it should not be coherent for past climate study.

#Referee 3 comments:

i) Comment 1: The reviewer raised concern about changing the biomass burning emissions with latitudes to improve agreement between model and observation. Answer: We would like to mention at the outset that, we did not change the biomass burning emission by a large amount. The regional scaling causes overall only a 4 Tg deviation from the original RETRO field (from 38 Tg to 34 Tg). But it is also important to explain the rationale behind it within the scope and idea of the paper. In this paper, we tried to optimize the regional contribution of one or more methane sources by trying to improve the seasonal agreement between model derived methane concentration to the observed one. This same methodology is used in inversion studies. However, we are not doing any inversion, rather while doing this exercise in our bottom-up approach we are trying to figure out which one or two of the sources is most sensitive and whether improved model seasonality can be achieved with small perturbations in those while preserving the seasonality of the emission sources as they were provided. As there are clearly visible latitudinal differences, we scaled the Northern and Southern hemi-

sphere independently (quasi a two-region inversion), and as additional constraint we preserved the global total amount of the emissions. In this sensitivity test, we figured out that even a small modification in the hemispherical distribution of biomass burning proves effective to come up with an improved seasonal match, particularly over farthest south latitudinal stations. Although a better seasonal match at those stations can also be achieved by modifying other sources e.g. enteric fermentation and ruminants, it occurs at the expense of diminishing the seasonality at other stations. So we considered scaling biomass burning methane emissions instead of a multiple other sources. We must state that it is not blindly done “to cover up the drawback of wetland methane parameterization” but to explore the spatial uncertainties of various methane sources and test the robustness of our wetland model.

The reviewer also argued if the latitudinal boundary selected in methane parameterization can change over climate and what is the rationale behind this selection.

Answer: According to the Gedney et al. (2004) wetland methane emission parameterization, which is followed in this paper, KCH4 is a global constant, which is calibrated and set in such a way that the difference between modeled wetland emission and observed data is minimal. So, it is evident from their paper that the value of KCH4 is the constant of proportion in the empirical relation, which connects soil temperature and soil carbon to the methane emission and starting from an arbitrary value it is calibrated to a fixed value. The same approach is followed in this paper as well, however, instead of using only one global constant value, we decided to use two separate values of KCH4, one is for northern latitudinal wetlands and the other for tropical ones. Since the hydrological causes of wetlands occurrences at the northern latitudes and at the tropics are different, selecting two different KCH4 values doesn’t appear illogical to the authors and should rather be seen as an improvement over the original Gedney formula. Regarding the regional boundary of selection of different parameter values for wetland methane parameterization, we adopted a simple and straightforward approach to use fixed latitude boundaries. Note that this applies consistently to KCH4 and the

threshold values of soil wetness as well. Concerning the change of the latitudinal boundary over time we acknowledge the referee's point but argue that the vast majority of today's tropical wetlands would also have shown tropical characteristics during the LGM.

As argued by the reviewer, it is true that there are various newly developed models to parameterize methane emission from wet soils which can be cited in the text. These DGVMs in general consider different methane emission processes (plant mediated, ebullition etc.) as a fraction of simulated soil carbon and NPP in the form of heterotrophic respiration (Christensen et al. 1996). However we would like to state that basis of their calculation and that used in Gedney et al. are actually the same. The starting point in both the methods is Arrhenius equation, which relates the rate of chemical reaction to the soil temperature. In Gedney et al. the Q10 stands for that which defines the rate of reaction with 10degC temperature rise. The soil carbon in Gedney et al. is replaced by NPP (available substrate) in the vegetation models. As per as KCH4 is concerned, it is nothing but a constant of proportion and can vary depending up on soil type. Since we do not incorporate different soil types in the present method since it can be a major constraint for past climate, we chose to use two different set of values for KCH4 for regions north and south of 45deg N as described in sub-section 2.1. Regarding the optimization of KCH4 values, we already discussed it in the previous bullet.

ii) Comment 2: As the reviewer suggested, we should mention the methane lifetime for each of the simulations in the text. As answer to the reviewer's question, all the simulations are done for at least twenty years to reach the steady state. For the present day simulation, the steady state is considered to be the period between 1999 and 2003 during which the global average methane concentration was approximately 1780 ppbv and showed least inter-annual variability. The emissions are optimized in view of that. As per as the biomass emission is concerned, the climatological mean between 1990 and 2000 is used in the simulation. As for the past climatic period, we chose LGM to be

21,000 years before present and pre-industrial period to be 200 years before present.

iii) Comment 3: The methane loss due to dry deposition changes significantly from Present day to LGM, dropping to 7 Tg/year.

iv) Comment 4: The reviewer argued if the GFED fire emissions data can be reconciled in this study?

Answer: While we of course acknowledge the considerable uncertainties in the estimations of biomass burning emissions, previous research has shown that most current inventories tend to agree on the general patterns and seasonal distribution of the biomass burning source. The RETRO inventory is very similar to GFEDv2 (see Schultz et al., 2008) and for the purpose of this study there is no point in trying out other inventories just for the sake of doing so.

v) Comment 5: The reviewer suggested discussing historical biomass burning estimates from post Valdes et al. (2005) study.

Answer: This is a valid point. We should include Marlon et al. and similar other past biomass burning estimates in discussion.

vi) Comment 6: We would like to point out here that although Montzka et al. (2011) talks about recent OH variability, nevertheless it also strongly suggests that OH is well buffered against perturbations which endorses a smaller post industrial change. However having said that, we must admit that "no change" is certainly a lower limit for OH as the truth lies in between the Naik et al. estimation (-0.6$\pm$8.8%) and zero.

vii) Comment 7: We refer to figure 1 for a better understanding of the changes in LGM OH. Figure 1 shows the impact of various factors on LGM OH change including the effect of BVOC reduction. If we add up the individual impacts, the net OH change was 26%. But the recent chemistry studies (Taraborrelli et al., Hofzumahaus et al.,) raised serious doubt over the impact of BVOCs on OH. Thus, if we ignore the BVOC effect on OH, we find that OH remains unchanged during LGM. However, since it is not sure

how exact the BVOC-OH chemistry works, we chose two extreme scenarios, one with a 26% higher LGM OH and the other with an unchanged OH.

viii) Comment 8: We agree to the argument of the reviewer that PD wetland methane estimates should be compared with other estimates. We should include that in the text. However, regarding Prather et al. uncertainty study, where they have considered the potential wetlands change between PI to present based on Houweling et al. study, the tentative guess was made by considering effects of various complex factors like land use change, human influence of wetland conversion etc. In our paper, we discretely modeled wetland emission for PI and Present day which has a limitation of not inculcating these factors. As the reviewer later pointed out, indeed deriving these changes and incorporating their effects are difficult. Nevertheless, we should mention in the text that these factors are a source of uncertainty in PI wetland methane change.

ix) Comment 9: We decided to keep Figure 4 separate to Figure 5 to keep focus on the effect of temperature seasonality solely on north latitudinal wetlands.

x) Comment 10: We decided to present only the wetland maps with its occurrence frequency to highlight the differences of their distribution between LGM and PD. We did not show the methane emissions maps mainly for the reason that the locations are the same and also to avoid too many maps. However, if the reviewer finds it essential, we can show the emission maps as well.

xi) Comment 11: We already agreed to discuss the uncertainty imposed by land use changes on PI wetlands.

xii) Comment 12: We completely agree with the reviewer to mention in text that wetland area is estimated high and the methane emission in the lower end.

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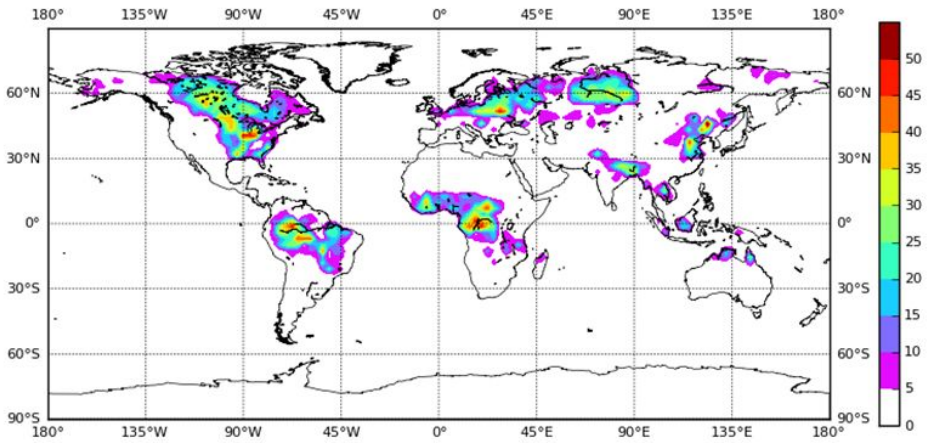

Fig. 1. Annual mean methane emission rate (mg/m2/day) for present day