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[1] We assess the contribution made to the interannual variability of the global methane accumulation rate from its atmospheric sink using the STOCHEM tropospheric chemistry model coupled to the HadCM3 climate model. For both control and climate change scenarios, the standard deviation of the detrended accumulation rate was 1.4 ppbv/yr for the period 1990–2009, compared with the measured standard deviation of 3.1 ppbv/yr for the period 1984–1999. As the model emissions have no variability, the methane sink processes in the model are responsible for all the simulated variability of the methane accumulation rate. This appears to explain a significant fraction of the observed variability and was well correlated with simulated water vapour. The largest component of the model interannual variability is derived from the El-Niño Southern Oscillation cycle in the coupled Ocean-Atmosphere model, and this mode of variation is shown to be present in the methane accumulation rate. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0330 Atmospheric Composition and Structure: Geochemical cycles. **Citation:** Johnson, C. E., D. S. Stevenson, W. J. Collins, and R. G. Derwent, Interannual variability in methane growth rate simulated with a coupled Ocean-Atmosphere-Chemistry model, *Geophys. Res. Lett.*, *29*(19), 1903, doi:10.1029/2002GL015269, 2002.

1. Introduction

[2] Methane is the most important greenhouse gas after carbon dioxide, with both anthropogenic and natural emission sources and with its reaction with the hydroxyl radical (OH) in the troposphere as its primary sink. To improve the confidence in future predictions of methane, we need to understand the recent changes in its global concentration. The growth rate of atmospheric methane displays considerable interannual variability as well as long term decrease [*Dlugokencky et al.*, 1998, 2001]. These variations have been attributed to a variety of causes including variations in wetland emissions caused by changes to surface temperature and precipitation [*Bekki and Law*, 1997; *Dlugokencky et al.*, 2001], anthropogenic emissions [*Law and Nisbet*, 1996; *Karlsdottir and Isaksen*, 2000], tropical UV fluxes after volcanic eruptions [*Dlugokencky et al.*, 1996], and in its atmospheric sink with OH [*Bekki and Law*, 1997].

[3] Primary production of OH takes place through the reaction of $O(^1D)$ with water vapour and comparison of model and measured OH [*Chen et al.*, 2001] confirms the dominant role of this process in the lower troposphere where most methane destruction occurs. *Khalil and Rasmussen* [1986] postulated that the interannual variation of methane measurements at Cape Meares (45°N, 124°W) were in response to El Niño Southern Oscillation (ENSO) events which promote enhanced removal of methane through higher concentrations of water vapour. *Bekki and Law* [1997] explored the response of the atmospheric growth rate with a 2-D global model of atmospheric chemistry to the variability of the zonal mean temperature measurements in the lower troposphere and found that the resultant OH changes could cause variations in methane growth rate of a few ppb per year. Increases to temperature and humidity due to climate change significantly increase the rate of methane oxidation [*Johnson et al.*, 1999; *Stevenson et al.*, 2000; *Johnson et al.*, 2001]; here we consider the role of the internal variability of the climate system on methane oxidation using a coupled ocean-atmosphere-chemistry model.

2. Coupled Ocean-Atmosphere-Chemistry Model

[4] We used a version (HadCM3) of the Hadley Centre ocean-atmosphere GCM [*Gordon et al.*, 2000] coupled to the Met. Office Lagrangian model of tropospheric chemistry (STOCHEM). The HadCM3 model does not require adjustment of ocean heat and salinity fluxes and simulates a stable and realistic present day mean climate [*Gordon et al.*, 2000]. The internal variability of this model is discussed by *Collins et al.* [2001] who conclude that the magnitude and frequency characteristics of the variability of global surface temperature on annual to decadal time scales is in good agreement with the observations. The model simulates an El Niño Southern Oscillation ENSO with an irregular 3–4 year cycle and with a realistic teleconnection pattern.

[5] The STOCHEM model and the scenarios used are described by *Johnson et al.* [2001] and *Stevenson et al.*, [2000]. *Collins et al.*, [2000] and other studies [*Kanakidou et al.*, 1999a, 1999b] show validation of model results against observations of O_3, NO_x, CO, and HO_x precursors. The results from two scenarios were used. The first scenario uses the control HadCM3 model with constant greenhouse gases set at their pre-industrial values while in the climate change scenario greenhouse gases in HadCM3 evolve in accordance with the A2 scenario from the Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios (SRES) [*Nakicenovic et al.*, 2000]. Trace gas emissions used by STOCHEM in both experiments evolve

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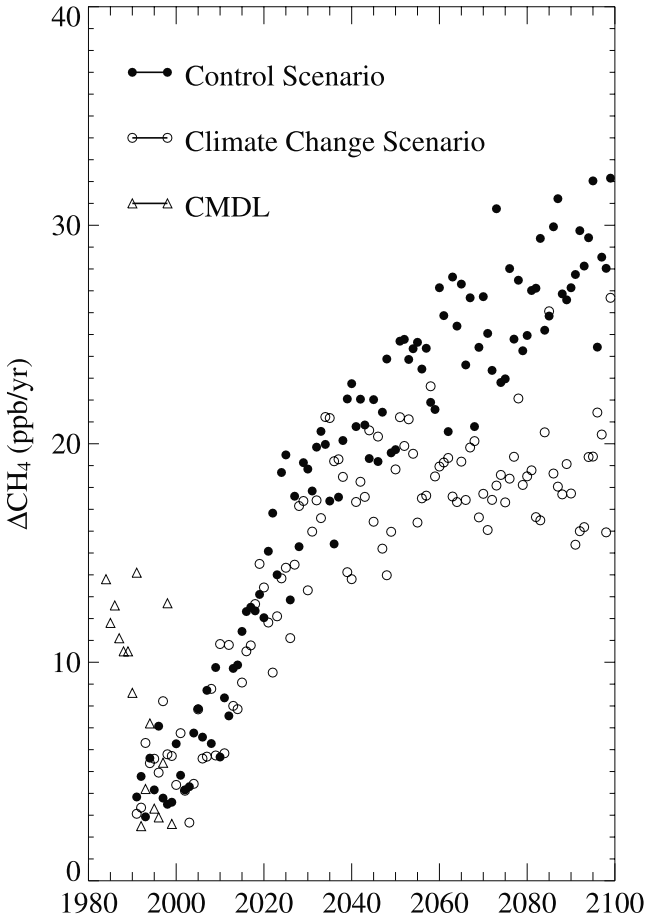

Figure 1. The growth rate of global-average atmospheric methane simulated by the HadCM3/STOCHEM model over the period 1990–2100 for the control (●) and climate change (○) scenarios, with the observed [*Dlugokencky et al*., 2001] annual growth rate for 1984–1999 (Δ).

as prescribed by the SRES A2 scenario. Natural emissions, including the NO_x source from lightning were kept constant in both experiments. Other details of the emission scenario were essentially the same as described in *Stevenson et al.* [2000].

3. Results and Discussion

[6] Figure 1 shows the annual methane growth increment (ΔCH_4) from recent observations [*Dlugokencky et al.*, 2001] together with the simulations over the 110 year period of the two scenarios. There is a considerable increase in the simulated future growth rate which is due to increases in the A2 scenario methane emissions, and there are significant interannual variations throughout the period. These are not derived from variation in the methane emission rates as annual changes to man made emissions were represented as smooth functions and natural biogenic emissions were kept constant. The variations shown are therefore a manifestation of the internal variability of the climate model expressed in the variation in the methane sink rate through its reaction with OH. While the observed downwards trend in ΔCH_4 is incompatible with the upwards trend in the SRES emissions used in the model, it is the variability in the model results which forms the subject of the present study.

[7] We are concerned here with the simulated variability of the model system, not with differences between the two scenarios and have therefore chosen to analyse in detail only the first twenty years of each scenario where differences between the methane oxidation in each case were small [*Johnson et al.*, 2001]. Both scenarios were used in order to ensure that the results were robust. The model derived standard deviation of the detrended annual ΔCH_4 predictions shown in figure 1 is 1.4 ppb/year in both experiments for the period 1990–2009 compared to the standard deviation of the detrended observations [*Dlugokencky et al.*, 2001] of 3.1 ppb/yr for 1984–1999. The model is therefore generating significant variability in atmospheric methane growth entirely from sink and transport processes, lacking any variability in emissions or UV fluxes from the stratosphere.

[8] The interannual variability of the model in the tropics, where much of the oxidation of methane is believed to take place, is expected to be dominated by the ENSO variability, represented here by the Niño-3 index: the mean sea surface temperature anomaly in the region 5°N–5°S, 150°–90°W.

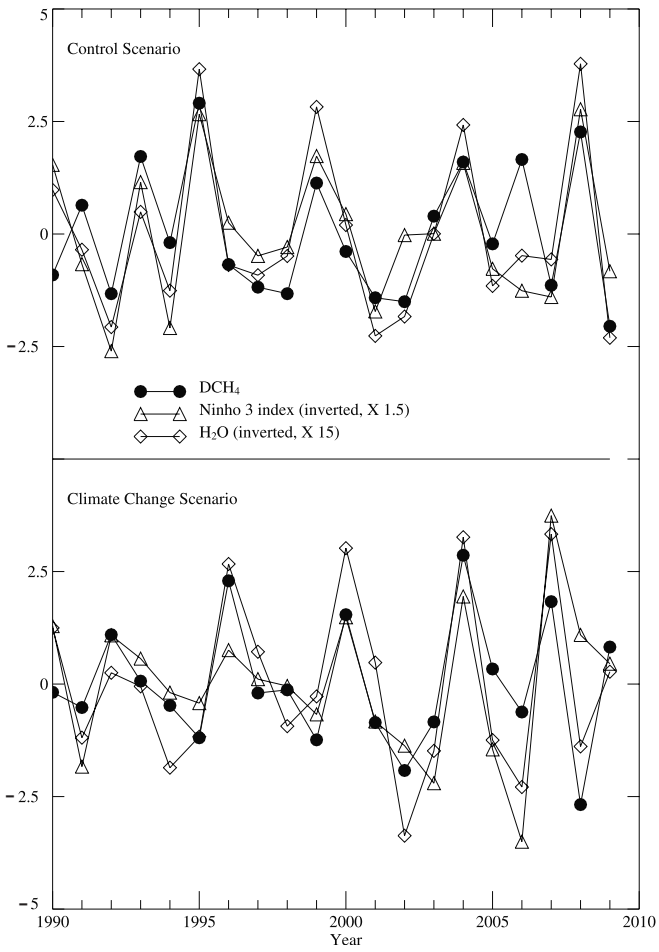

Figure 2. Variation of globally-averaged methane growth rate (●) in units of ppb/yr, total water vapour (✧) in units of Eg scaled by a factor of 15, and Niño-3 index (Δ) scaled by a factor of 1.5 simulated for 1990–2009 for the control (a) and climate change (b) scenarios. All data have been detrended and the water vapour and Niño-3 index have been scaled and inverted for plotting.

In order to characterise the influence of various model factors on the methane sink, we plotted annual ΔCH_4 values against annual average Niño-3 index, global mean near-surface air temperature, total mass of water vapour, OH inventory, and the chemical fluxes, CH_4 + OH and H_2O + $O(^1D)$. The first 20 years of each scenario were used, and the results were detrended using a third order polynomial fit before the regression analysis. Figure 2 shows the high degree of coherence between the time series of CH_4, Niño-3 index and water vapour.

[9] Table 1 shows linear correlation coefficients between ΔCH_4 simulated by both experiments and the other variables. As expected, nearly all the simulated variation of ΔCH_4 is explained by the variation in the CH_4 + OH flux, with a small residual component from sampling errors in STOCHEM. The next highest correlations are with OH and the H_2O + $O(^1D)$ flux. Correlations with temperature are not so strong, but there is still a significant correlation between ΔCH_4 and the Niño-3 index. Figure 3 shows scatter plots to illustrate the relationships between ΔCH_4 and some of the variables.

[10] From the model standard deviations for near-surface tropical temperatures (0.07% of the mean in Kelvin) compared to that for specific humidity (11% of the mean), we expect variations in water vapour to be more significant than variations in the CH_4 + OH rate coefficient. The strong correlation between the CH_4 + OH flux and water vapour shown in Table 1 for both scenarios also suggests that the ultimate source of the methane sink variability is derived from the humidity variations. This indicates that variation in the OH source from the reaction of water vapour with O^1D is important and this is supported by the high correlation between ΔCH_4 and the H_2O + $O(^1D)$ flux.

[11] The simulations above rely critically on the degree of realism of the ENSO promoted variability in the model. The amplitude of ENSO was assessed by *Collins et al.*, [2001] from the Niño-3 temperature anomalies and lies within the range of uncertainty of the observations. While HadCM3 captures the periodic nature of ENSO with the correct frequency, there is too much variability on interannual time scales, and there is too strong a reponse in the West Pacific. In addition to the temperature data discussed above, *Jones et al.*, [2001] used precipitation data to compare with the results from HadCM3L, a version with a lower ocean resolution than used here. This model captures many features of the precipitation patterns in response to ENSO, including negative correlations in northern South America and Australia. The main discrepancy is North Africa which showed a correlation of the wrong sign.

Table. 1. Correlation Coefficients Between Detrended Model Variables for the Control (CON) and Climate Change (CC) Scenarios for the Period 1990–2009

Model Variables	Correlation Coeff. (CON)	Correlation Coeff. (CC)
ΔCH_4; H_2O	−0.82***	−0.82***
ΔCH_4; OH	−0.81***	−0.87***
ΔCH_4; $T_{1.5}$	−0.66**	−0.35 ns
ΔCH_4; T_s (Niño-3)	−0.64**	−0.55*
ΔCH_4; CH_4 + OH	−0.97***	−0.97***
ΔCH_4; H_2O + $O(^1D)$	−0.88***	−0.84***
ΔCH_4 + OH; $T_{1\cdot5}$	0.67**	0.40 ns
ΔCH_4 + OH; H_2O	0.85***	0.86***
ΔCH_4 + OH; OH	0.83***	0.92***
ΔCH_4 + OH; H_2O + $O(^1D)$	0.95***	0.93***

The significance level of the correlation coefficient is represented as: 5% (*), 1% (**), 0.1% (***), not significant (ns).

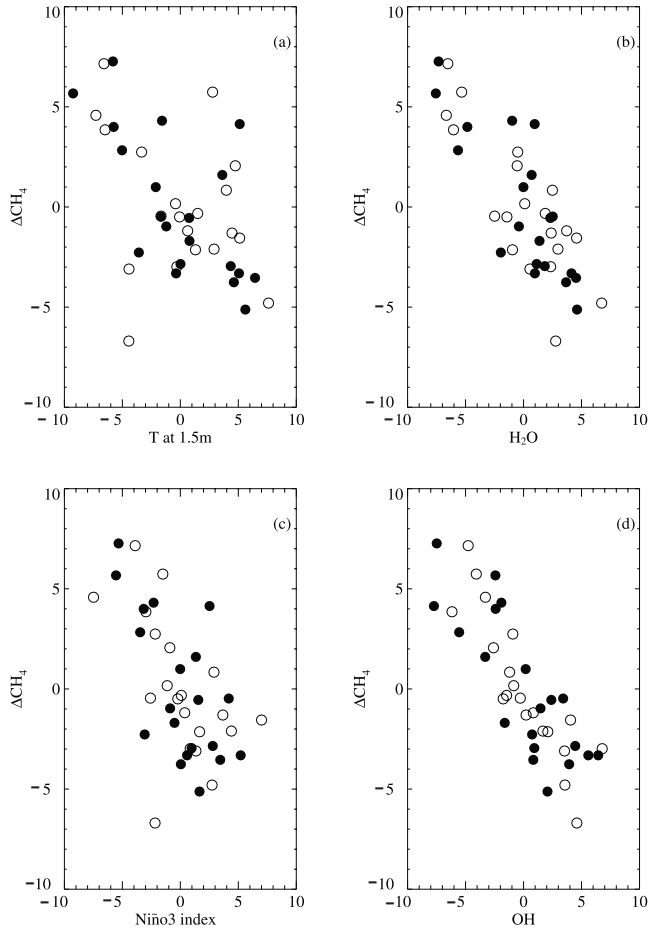

Figure 3. Scatter plots of ΔCH_4 with (a) $T_{1.5}$, (b) H_2O (c) Niño-3 index, and (d) OH for control (●) and climate change scenarios (○). All data have been detrended and scaled before plotting.

[12] The variability of the methane growth rate assumes a new importance in the assessment of model mechanisms which are aimed at reproducing the response of methane to climate change. The results reported above are a first attempt at describing the variability from the methane sink process in a coupled chemistry-climate model and point to limitations in the realism of the climate response to ENSO.

4. Conclusion

[13] We have shown here that the variations on an annual basis in the chemical removal of methane exert an important impact in the variability of the methane growth rate and suggest that variations in both source and sink processes may be linked through the ENSO cycle. The variations in CH_4 simulated in the HadCM3/STOCHEM model are from sink processes alone and are sufficient in themselves to explain a significant fraction of the observed variations in the annual accumulation rate for methane. The largest component of this variability is derived from ENSO varia-

bility in the HadCM3 model. The variations in the methane sink rate which are estimated here may act as a constraint to the interpretation of observations of methane growth rate, and point to the need for a new generation of coupled models which include realistic simulation of the variations of both natural emission and sink processes.

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