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Sensitivity of the atmospheric CH_4 growth rate to global temperature changes observed from 1980 to 1992

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ABSTRACT

The response of the atmospheric CH_4 growth rate to observed global temperature fluctuations recorded from 1980 to 1992 is explored using a 2-dimensional (2-D) chemistry/transport model. Two competing mechanisms are considered in this study: the temperature dependency of CH_4 emissions from wetlands and the temperature dependency of the CH_4 photochemical sink, principally via the positive temperature dependencies in the the reaction rate between CH_4 and OH and in the tropospheric content of water vapour, the source of OH. The results provide some bounds on these temperature effects and the regions where they are most effective at perturbing the CH_4 growth rate. The OH-induced effect could have caused interannual variations in the CH_4 growth rate of the order of few ppbv yr^{-1}; their pattern is consistent with the variations observed during the 1980s, but not in 1991/1992, when the Mount Pinatubo eruption led to global perturbations of not only the temperature but also of stratospheric ozone, stratosphere-tropospheric exchanges and the general circulation. Using the current range of temperature sensitivity of wetland emissions, the wetland effect could explain the entire CH_4 anomaly observed in 1991–1992 in the northern hemisphere. However, this mechanism cannot account for any of the CH_4 anomalies observed during the 1980s. When these temperature mechanisms are combined, we find that wetland effects dominate in the northern hemisphere and the OH effect being more important in the tropics. Any future model calculations addressing the impact of temperature fluctuations on CH_4 concentrations should be compared to the entire observational record instead of focusing on a particular CH_4 anomaly.

1. Introduction

Atmospheric methane (CH_4) is of great importance for tropospheric photochemistry and the climate. Depending on the concentration of nitric oxide (NO), the oxidation of CH_4 represents a net source or sink of odd-hydrogen ($HO_x = OH + HO_2$) and ozone (Crutzen and Zimmermann, 1991). As such, CH_4 exerts some control over the oxidising capacity of the troposphere. CH_4 oxidation is also a non-negligible source of ozone in the lower stratosphere and an important greenhouse gas (IPCC, 1994). CH_4 concentrations have increased rapidly during the last 150 years. However, it has been shown that the CH_4 increase slowed down during the 1980s (Steele et al., 1992). Furthermore, measurements at some sites in the northern hemisphere seem to suggest that the CH_4 growth rate dropped unexpectedly in 1991/1992 with, for the first time, a negative growth rate in the northern hemisphere (Dlugokencky et al., 1994a). It must be pointed out that it has not yet been demonstrated that these short-term CH_4 anomalies observed at few sites are fully representative of global changes.

The 1980s decline in the CH_4 growth rate has been partially attributed to reductions in emissions from cattle and rice paddies (Khalil and

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Rasmussen, 1993). An increase in OH, the major sink for CH_4, has also been suggested. This could have been brought about by a decrease in stratospheric ozone during the 1980s (Madronich and Granier, 1991). However, transient model calculations suggest that stratospheric ozone changes would have had the opposite effect on the CH_4 growth rate in the late 1980s (Bekki et al., 1994; Fuglestvedt et al., 1994). No observational evidence of an OH increase has been found yet. For example, Prinn et al. (1995) did not infer a statistically significant trend in global OH from methylchloroform data collected during 1980s. On shorter timescales, changes in interhemispheric transport caused by the El Nino/Southern Oscillation (ENSO) could have caused an interannual variability in the hemispherically averaged CH_4 growth rate (Dlugokencky et al., 1994a).

A number of mechanisms have been put forward to explain the sudden and unexpected drop in the CH_4 growth rate observed at some sites in 1991/1992, mostly because it coincided with unusually large perturbations in stratospheric ozone (Gleason et al., 1992), tropospheric and stratospheric temperatures (Duton and Christy, 1992; Labitzke and McCormick, 1992) and possibly the general circulation (Pitari, 1993) including changes in stratosphere-tropospheric exchange (Schauffler and Daniel, 1994), all linked to the Mount Pinatubo eruption in June 1991. Part of the 1991/1992 anomaly could be accounted for by enhanced tropospheric OH resulting from stratospheric ozone depletion (Bekki et al., 1994; Fuglestvedt et al., 1994). There is also the possibility of rapid reductions in anthropogenic emissions from biomass burning (Lowe et al., 1994) and from former Soviet Union natural gas emissions (Dlugokencky et al., 1994a; Law and Nisbet, 1996), although much of the evidence is anecdotal.

The tropospheric cooling following the eruption of Mount Pinatubo (Fig. 1) could also have affected levels of atmospheric CH_4. Hogan and Harriss (1994) suggested that lower temperatures might have led to reduced CH_4 emissions from wetlands. However, this effect is expected to have been mitigated by a decrease in the oxidation by OH, the major sink for CH_4, because of the positive temperature dependence of the reaction (Rudolph, 1994) and because of the likely decrease in atmospheric water vapour following the Pinatubo-induced tropospheric cooling. GCM experiments (IPCC, 1992) and satellite data (Rind et al., 1991) indicate that a cooling should lead to a drier atmosphere (e.g., lower specific humidity, approximately constant relative humidity). As OH is primarily produced by the reaction of H_2O with O^1D, reduced water vapour content is accompanied by a decrease in OH levels. OH levels are also slightly dependent on the temperature via the chemistry, but it is a relatively small effect (Fuglestvedt et al., 1995).

Here, a global 2-D model is used to explore the response of the CH_4 growth rate to the global temperature fluctuations recorded from 1980 to 1992 and to estimate some bounds on various temperature effects and the regions where they are most effective at perturbing the CH_4 growth rate. The mechanisms considered in this study are the temperature dependency of the wetland emissions, of the reaction rate of CH_4 with OH and of the water vapour content. These mechanisms are first treated independently before combining them and assessing their relative importance in perturbing the CH_4 growth rate over the period 1980–1992.

2. Modelling

The model used in this study is a classical Eulerian zonally averaged two-dimensional model of the atmosphere (Harwood and Pyle, 1975; Law and Pyle, 1993a). It extends from pole to pole and from the ground to 60 km with an horizontal resolution of 9.5° and a vertical resolution of half a pressure scale ($\sim$3.5 km). The model contains detailed representations of atmospheric photochemistry, transport and radiative processes. The mean circulation is calculated, based on forcing by latent and radiative heating and eddy transport processes. The vertical diffusion coefficients given by Hidalgo and Crutzen (1977) are used in the troposphere, albeit reduced in the tropics. A comprehensive description of O_x, NO_x, HO_x, ClO_x, BrO_x, SO_x, CH_4 and non-methane hydrocarbon (NMHC) photochemistry is included (Law and Pyle, 1993a, 1993b; Bekki and Pyle, 1993). Reaction rates and photolysis cross sections are taken generally from the recommendations of DeMore et al. (1992) and Atkinson et al. (1992). The photolysis scheme has been improved to make it more appropriate for tropospheric studies based on Hough (1991). The photochemistry has been

Table 1. *Model scenarios*

Run	Chemistry $=f(\Delta T^*)$	Wetlands $=f(\Delta T^*)$ (7.5% K^{-1})
control	no	no
A	yes	no
B	no	yes
C	yes	yes

* MSU temperature anomalies.

validated in model intercomparaison (IPCC, 1995). The model-calculated global OH concentration compares well with estimates inferred from methylchloroform measurements (Prinn et al., 1995). Climatological TOMS ozone columns are specified in the calculation of the tropospheric photolysis rates (Bekki et al., 1994). Tropospheric water vapour, calculated from the local humidity, is also slightly dependent on the surface temperature following the expression provided by Cess (1976). The boundary conditions for source gases such as CO_2, nitrous oxide (N_2O) and CFCs are from a recommended scenario (WMO, 1994). Anthropogenic emissions of CH_4, CO and NO_x have been updated using Muller (1992). Biomass burning emissions are taken from Hao and Ward (1993) and follow the seasonal variations reported in Law and Pyle (1993a). The soil sink for CH_4 is also taken into account (−30 Tg per year). Wetlands emissions of CH_4 are set to 100 Tg/year out of a total of 460 Tg/year. The temporal and spatial distribution of the wetland emissions follows that given by (Aselmann and Crutzen, 1989). Emissions from high northern wetlands are highly seasonal peaking in the late summer/autumn. Because of this seasonality, one has be cautious when working out changes in wetland emissions from annual mean temperature anomalies.

Zonal monthly mean temperature anomalies, calculated from microwave sounding units (MSU) data (Dutton and Christy, 1992; J. Christy, U. of Alabama, USA, unpublished data), are interpolated onto the model grid. They are centred on the lowest region of the troposphere (~700 mbar) (Dutton and Christy, 1992). As a first approximation, temperature anomalies at the surface and the lowest model layer are assumed to be equal (Dlugokencky et al., 1994b). The use of this approximation and of a zonally average temperature dataset prevents a wholly accurate quantification of the actual temperature feedbacks on the CH_4 growth rate. However, the aim of this study is to estimate the sensitivity of the CH_4 growth rate to temperature changes rather than to accurately reproduce actual changes in the CH_4 growth rate, particularly when other potentially more important perturbations to CH_4 sources and sinks have been omitted in these calculations, particularly for the 1991/1992 period. The model runs are summarised in Table 1. The model was integrated from 1974 to 1992. No temperatures anomalies are prescribed in the control run. In a perturbation run A, MSU temperature anomalies are prescribed between the surface and ~3.5 km to examine the feedback between temperature, humidity, OH and CH_4 concentrations. In run B, wetland emissions of CH_4 are varied according to MSU temperature anomalies assuming a temperature sensitivity of 15% increase for 2 K increase (i.e., $7.5\%^{-1}$) (Matthews, 1993).

The feedback between temperature changes and chemistry is not included in this run. Based on other estimates (Hogan and Harriss, 1994), higher temperature sensitivities were also considered. In run A, temperature changes should be most effective at perturbing CH_4 levels during the summer when destruction of CH_4 by OH becomes important. In run B, the northern hemisphere (NH) CH_4 growth rate is expected to be most sensitive to temperature changes in the NH during late summer when wetland emissions peak. Run C combines both temperature effects.

3. Results

Fig. 1 shows area-weighted mean MSU temperature anomalies for the tropics and the mid to high northern hemisphere (north of ~24°N) as a function of time. Several of the sustained temperature changes coincide with ENSO events or volcanic eruptions (Dutton and Christy, 1992).

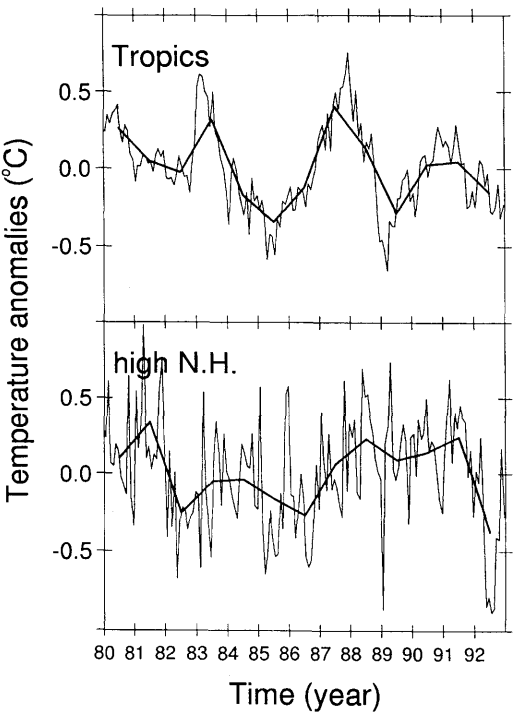

Fig. 1. Time series of area-weighted mean temperature anomalies for the globe, tropics and mid/high northern hemisphere (~24°N).

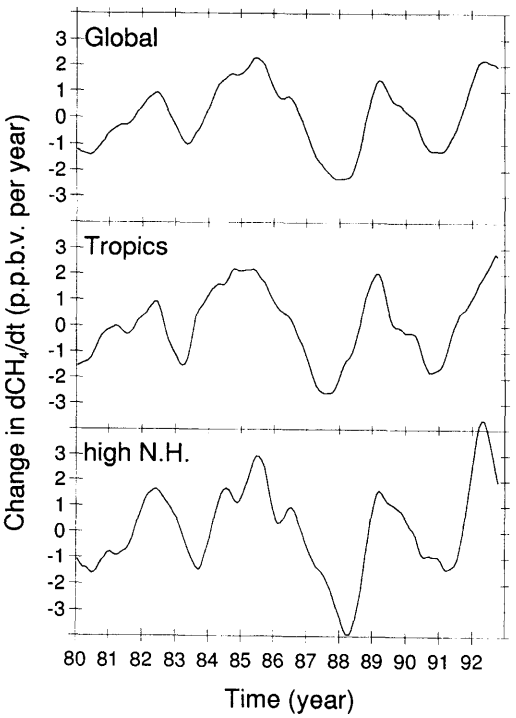

Fig. 2. Time series of the differences between run A and control run in the growth rates of global, tropical and north hemispheric (~24°N) mean CH_4 mixing ratios at the surface level (~800 mbar). Growth rates are calculated from the deseasonalised monthly means using the method of linear moving slopes described in Khalil and Rasmussen (1993) with a time span of 1 year.

Short-term variability is much larger at high latitudes than in the tropics. The prominent features in the mean tropical temperature are cool periods peaking in 1985, 1989 and 1992 and warmer periods in 1983, 1987 and 1991. The cool and warm periods are less well defined in the mean NH temperatures, partly because of much larger short-term variability. The cool periods occur in 1982, 1986 and 1992; warmer periods in 1983, 1988 and 1991 in the NH. The strong and rapid cooling of 1992 was most likely due to the presence of Pinatubo aerosols in the stratosphere which backscattered incoming solar radiation.

The changes in the tropical, the mid to high northern hemisphere, and global CH_4 growth rate with respect to the control run are plotted against time in Figs 2, 3 and 4 for runs A, B and C respectively. Since almost half of the atmospheric CH_4 burden is contained in the tropics and most of the CH_4 removal takes place in the tropics, the global mean CH_4 growth rate changes follow somewhat closely the tropical CH_4 growth rate for all the runs.

In run A, the variations in the tropical CH_4 growth rate are inversely correlated with the changes in tropical lower tropospheric temperatures, reflecting the positive temperature dependency of CH_4+OH and formation of OH from reaction of H_2O with O^1D. The contributions of the two mechanisms to the positive temperature dependency of the CH_4 removal rate are comparable with the H_2O effect slightly dominant. Overall, the CH_4 growth rate changes show similar features in the NH and in tropics. The model predicts minima for the NH CH_4 growth rate in 1983 and 1988, which were also seen in the observations (Dlugokencky et al., 1994a). However, the modelled CH_4 growth rate increases sharply between 1991 and 1992 in the NH and

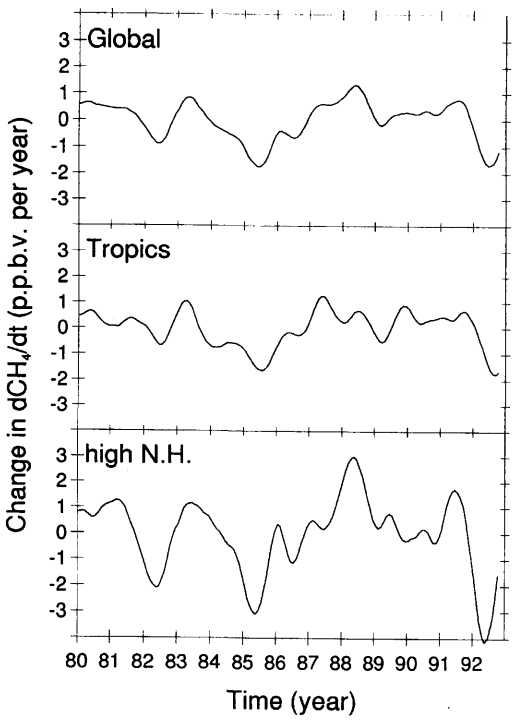

Fig. 3. Same as Fig. 2 except of the differences between run B and control run.

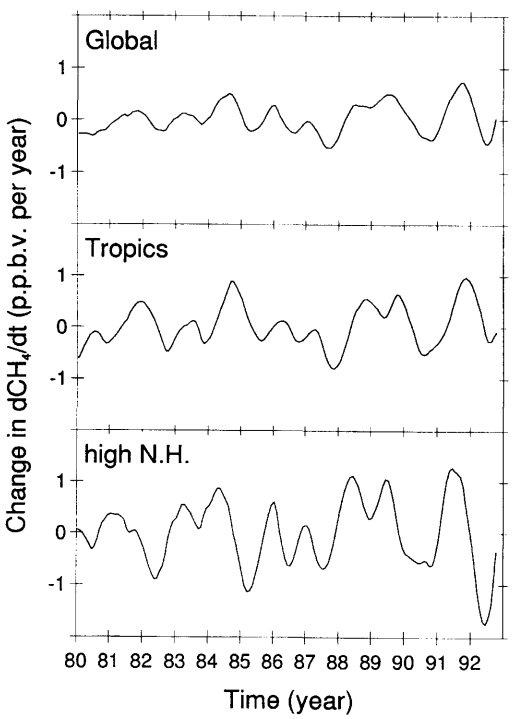

Fig. 4. Same as Fig. 2 except of the differences between run C and control run.

the tropics, which is in apparent contradiction with observations. The 1991/1992 period saw unprecedentedly large changes in stratospheric ozone and in the general circulation following the volcanic eruption of Mount Pinatubo. The resulting perturbations of the CH_4 budget (Bekki et al., 1994; Fuglestvedt et al., 1994; Schauffler and Daniel, 1994) could easily have masked the perturbations calculated in run A.

In run B, the variations in the CH_4 growth rate are mostly positively correlated with temperature changes. The variations in the CH_4 growth rate are much larger in the NH than in the tropics, indicating a greater sensitivity of CH_4 in the northern hemisphere to changes in wetland emissions. This is not surprising given that wetlands are a major component of CH_4 budget in the NH whereas they are largely outweighed in the tropics by other CH_4 emissions such as rice paddies. The most pronounced drop in the modelled instantaneous CH_4 growth rate (from 1–2 ppbv yr^{-1} to -4 ppbv yr^{-1}) occurs in the northern hemisphere (N.H) between mid 1991 and the 1992 spring; there is also a decrease, albeit much smaller, in the tropics. This particular change is consistent with available observations which show a reduction in the CH_4 growth rate taking place primarily in the NH during the same period. The reduction inferred from observations was at least 3–4 times larger than the reduction calculated here for a temperature sensitivity of 7.5% K^{-1}. Further runs have shown that increasing the temperature sensitivity of wetland emissions produce approximatively the same relative increase in the CH_4 growth rate. Therefore, the results of run B can be directly extended to higher temperature sensitivities. Some field measurements suggest that the temperature sensitivity could be as high as a 5-fold increase for each 10K increase (i.e., 50% K^{-1}) (Hogan and Harriss, 1994). In that case, the entire 91/92 drop could easily be accounted for by temperature-induced reductions in wetland emissions. However, the modelled variations of the NH CH_4 growth rate do not match at all the observations during the 1980s. The model predicts a small peak in 1983, a very substantial decrease in 1985 and a

peak in 1988. All theses features are absent in the observations (see Fig. 4 of Dlugokencky et al., 1994a). If the 1991/1992 drop is to to be attributed to wetland emissions (Hogan and Harriss, 1994), one has to explain why wetland emissions became the driving force behind the variations in the CH_4 growth rate during the highly perturbed period of 1991/1992 whereas they appear to be outweighed by other factors during the 1980s.

Run C illustrates the competitive temperature-induced effects of OH chemistry and wetland emissions on the CH_4 growth rate. Interestingly, for a wetlands temperature sensitivity of 7.5% K^{-1}, the two effects appear to largely cancel out globally with a CH_4 growth rate spanning between about ± 0.5 ppbv yr^{-1}. However, the wetlands effect dominates in the northern hemisphere whereas the OH effect is more important in the tropics. If a wetlands temperature sensitivity as high as 50.% K^{-1} is considered, the wetlands effect dominates largely the OH effect, even in the tropics.

4. Conclusions

Using a photochemical 2-D model, we have explored the response of the CH_4 growth rate to global temperature fluctuations recorded during the past decade. We find that temperature-induced perturbations in the CH_4 sink via the temperature/water vapour/OH feedback and via the temperature dependency of the CH_4 plus OH reaction, are both significant, particularly in the tropics. They should have caused interannual variations in CH_4 growth rate of the order of few ppbv yr^{-1}. The model-calculated CH_4 growth rate anomalies are consistent with the observations during the 1980s. However, the CH_4 growth rate anomaly predicted in the NH during the 1991/1992 period is opposite to the anomaly inferred from the observations, indicating that other perturbations in CH_4 sources and sinks have been dominant during this very particular period (Bekki et al., 1994; Fuglestvedt et al., 1994; Schauffler and Daniel, 1994; Lowe et al., 1994; Dlugokencky et al., 1994a; Law and Nisbet, 1996). It is worth pointing out that the OH response to changes in H_2O calculated by our 2-D zonally-averaged model should be rather comparable to the one calculated with a 3-D model. Thompson et al. (1989) studied the sensitivity of OH to changes in H_2O and found that it varied little with the type of air mass (continental and marine mid-latitudes air, northern hemisphere low latitudes air and southern hemisphere mid-latitudes air). Only for polluted urban conditions did they find a substantial difference. But these regions will only make a very small contribution to the global CH_4 sink.

Temperature-induced perturbations in the CH_4 growth rate via the temperature dependency of the wetland emissions have also been investigated. The temperature sensitivity of wetland emissions is very uncertain. Using the current range of estimates, one can explain little or all of the dramatic 1991–1992 drop in the NH CH_4 growth rate. However, attributing the entire 1991/1992 drop of the NH CH_4 growth rate to temperature-induced reductions of wetland emissions is not easy to reconcile with the fact the variations in the CH_4 growth rate predicted in the the NH are inconsistent with observations during the 1980s. In addition, the CH_4 anomaly of 1991/1992 is probably not the best period to test the existence of a "wetland effect". Unusual decreases in the trends of N_2O, and CO_2 were also detected in 1992, showing that other mechanisms, possibly dynamical, were operating to reduce the CH_4 growth rate (Schauffler and Daniel, 1994). Model calculations have also demonstrated that the unprecedented large depletion in stratospheric ozone during the same period should have contributed to the sharp decrease in the growth rates of CH_4 (Bekki et al., 1994; Fuglestvedt et al., 1994).

The CH_4 emissions used in this work lie at the lower end of the range recommended by IPCC (1995). Increasing total emissions by 15% to set them in line with these recommendations (530 Tg/year of total emissions and 115 Tg/year of wetlands emissions) has very little impact on run A and increases the absolute CH_4 changes in run B by approximatively 15%. Ideally, the temperature effect of wetland emissions should be assessed with a 3-D model in order to account for the spatial heterogeneity of wetlands and temperature anomalies (Hogan and Harriss, 1994). However, as suggested by the model calculations, the key uncertainty in assessing the temperature effect of wetland emissions on the CH_4 growth rate is probably the temperature sensitivity of wetland emissions. Note also that the possible

temperature dependency of other CH_4 sources such as rice paddies emissions was not taken into account (Shearer and Khalil, 1993). When all the temperature mechanisms are combined, we find that, for a moderate temperature sensitivity of wetland emissions, the wetland emissions effect dominates in the NH whereas the chemistry effect is more important in the tropics.

From the model calculations presented here, it is not possible to reach definite conclusions on the processes responsible for the actual variations in the CH_4 growth rate, partially because other potentially important perturbations of CH_4 sources and sinks, especially for the period 1991/1992, were not included (Bekki et al., 1994; Fuglestvedt et al., 1994; Schauffler and Daniel, 1994; Lowe et al., 1994; Dlugokencky et al., 1994a; Law and Nisbet, 1996). Nonetheless, this study shows that the contribution of temperature effects to interannual variations in CH_4 is very significant and should be taken into account. Furthermore, if model assessments of the impact of temperature fluctuations on CH_4 are to be validated, they should be tested against the entire record of CH_4 observations rather than focusing on a particular CH_4 anomaly.

The results highlight the possible link between natural variability in temperature and CH_4. Changes in wetland emissions have already been put forward to explain variations in CH_4 concentrations at the end of the last ice age (Chapellaz et al., 1990). With the expected CO_2-induced global warming, more work is needed on the identification and quantification of the factors controlling wetland emissions. Chemistry-climate feedbacks, emphasized here, will need to be considered in such studies of past and future climates, ideally in a 3-D framework.

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