Atmos. Chem. Phys. Discuss., 11, 30757–30772, 2011
www.atmos-chem-phys-discuss.net/11/30757/2011/
doi:10.5194/acpd-11-30757-2011

Atmospheric Chemistry and Physics Discussions

Renewed methane increase for five years (2007–2011) observed by solar FTIR spectrometry

R. Sussmann[1], **F. Forster**[1], **M. Rettinger**[1], **and P. Bousquet**[2,3]

[1]Karlsruhe Institute of Technology, IMK-IFU, Garmisch-Partenkirchen, Germany
[2]Laboratoire des Sciences du Climat et de l'Environnement, IPSL-LSCE, CEA-CNRS-UVSQ, UMR8212, 91191 Gif-sur-Yvette, France
[3]Université de Versailles, Saint Quentin en Yvelines, France

Received: 11 October 2011 – Accepted: 4 November 2011 – Published: 16 November 2011

Correspondence to: R. Sussmann (ralf.sussmann@kit.edu)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Trends of column-averaged methane for the time period [1996, September 2011] are derived from the mid-infrared (mid-IR) solar FTIR time series at the Zugspitze (47.42° N, 10.98° E, 2964 m a.s.l.) and Garmisch (47.48° N, 11.06° E, 743 m a.s.l.). Trend analysis comprises a fit to the de-seasonalized time series along with bootstrap resampling for quantifying trend uncertainties. We find a positive trend during [1996, 1998] (9.0 [3.2, 14.7] ppb yr^{-1}, Zugspitze, 95 % confidence interval), a non-significant growth during [1999, mid 2006] (0.8 [−0.1, 1.7] ppb yr^{-1}, Zugspitze), and a significant renewed increase during [mid 2006, September 2011] of 5.1 [4.2, 6.0] ppb yr^{-1} for Garmisch, which is in agreement with 4.8 [3.8, 5.9] ppb yr^{-1} for Zugspitze.

The agreement of methane trends at the two closely neighboring FTIR sites with strongly differing levels of integrated water vapor (min/max = 0.2 mm/12.7 mm for Zugspitze, 1.9 mm/34.9 mm for Garmisch) proves that potentially significant water-vapor-methane interference errors do not affect the trend results, if the updated mid-IR retrieval strategy MIR-GBM v1.0 is used. Furthermore, agreement of the trend of 6.6 ppb yr^{-1} derived from SCIAMACHY (WFMD v2.0) data for the time period [mid 2006, mid 2009] is found within the 95 % confidence interval of the ground-based FTIR result.

While earlier studies using surface network data revealed changes of 8.0±0.6 ppb in 2007 and 6.4±0.6 ppb in 2008 (update from Dlugokencky et al., 2009), our updated result proves that meanwhile, the renewed methane increase has been persisting for >5 yr [mid 2006, September 2011]. This is either the longest and largest positive trend anomaly since >25 yr when systematic observations began or the onset of a new period of strongly increasing CH_4 levels in the atmosphere.

The 2007–2008 part of the anomaly was previously attributed to increased natural wetland emissions. For the full period from 2007 to 2011, our analysis of ECMWF ERA-INTERIM precipitations and 2-m temperatures shows that precipitations above tropical wetland areas increased in 2007–2008, decreased in 2009, and have

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been increasing again since 2010, while tropical land temperatures increased only slightly. As recent estimates of anthropogenic emissions are not yet available, it is not possible to finally conclude that the 2009–2011 period of methane increase was dominated by natural wetland emissions, although they probably play a significant role.

1 Introduction

The molecular symmetry of methane (CH_4) allows vibration-rotation excitation by infrared absorption to occur much more actively compared to carbon dioxide (CO_2). This is the principal reason why methane is the second important anthropogenic greenhouse gas in spite of its still relatively small abundance in the atmosphere.

Methane concentrations in the atmosphere have more than doubled since the beginning of industrialization (Forster et al., 2007). After a period of near-zero growth at the beginning of this century (Dlugokencky et al., 2003; Bousquet et al., 2006), the growth rate of atmospheric methane started to increase strongly again after 2006 (Rigby et al., 2008; Dlugokencky et al., 2009). Recent publications quantified renewed methane increase up to the year 2009 and discussed the possible causes of the increase observed in the years 2007 and 2008 (Rigby et al., 2008; Duglokencky et al., 2009; Bousquet et al., 2011; Frankenberg et al., 2011; Schneising et al., 2011; Spahni et al., 2011; Montzka et al., 2011). Significant contributions from a reduced OH sink or from biomass burning could not be identified, but evidence was found of an increase in emissions from northern wetlands in 2007 due to unusually high temperatures. In addition, an increase in tropical wetland emissions during 2007–2008 was found, which was mainly due to precipitation changes associated with the La Niña event in these years.

All in all, the 2007–2008 anomaly was attributed mainly to increased natural emissions from wet ecosystems. The question of how long the 2007–2008 anomaly will persist after 2008 is related to the interpretation of the mechanisms driving the trend until today (2007–2011). Wetland emissions are widely assumed to drive inter-annual

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variability (time scale of 1–2 yr), while more gradual changes on the time scale of several years are understood to be due to changing anthropogenic emissions.

It is therefore the goal of this paper to address the question of persistence of the 2007–2008 anomaly via an updated trend analysis including data until today (fall 2011). Contrary to most previous trend studies based on in-situ surface network data, we use ground-based solar absorption Fourier-Transform-Infrared (FTIR) spectrometry. This technique measures methane columns representative of large geographical areas. Measurements at a single FTIR station may be comparable to a set of in-situ measurements (Ohlsen and Randerson, 2004).

Columnar methane retrieved from ground-based FTIR spectrometry in the mid-infrared (mid-IR) within the Network for the Detection of Atmospheric Composition Change (NDACC, http://www.ndacc.org) has already been used for trend studies (Zander et al., 1989; Angelbratt et al., 2011) or satellite validation (e.g., Sussmann et al., 2005). However, it became obvious recently that methane-column retrievals at high-humidity (low-altitude) sites may be dominated by water-vapor (H_2O/HDO)-CH_4 interference errors of up to 5 %. Consequently, artifacts dominated the seasonal cycle of methane retrieved with the standard NDACC retrieval strategy. Dry (high-altitude) sites were not affected. Note that a general formulation of the interference problem was given before (Sussmann and Borsdorff, 2007) and water vapor interference also perturbed methane retrievals from SCIAMACHY (Frankenberg et al., 2008). To improve the standard retrieval strategy used within NDACC, we recently developed a new retrieval strategy (MIR-GBM v1.0) using systematically selected spectral micro windows and spectroscopic parameters (Sussmann et al., 2011). MIR-GBM v1.0 was shown to eliminate H_2O/HDO-CH_4 interference errors down to the ≈0.1 % level at the wettest sites. In return, a good agreement of the retrieved Northern Hemisphere seasonal cycle with SCIAMACHY results was achieved for the first time (Sussmann et al., 2011). Therefore, another goal of this paper is to further validate MIR-GBM v1.0 by comparing the methane trend results from two neighboring sites with strongly differing levels of integrated water vapor (IWV), namely, the Zugspitze (2964 m a.s.l.) and Garmisch

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(743 m a.s.l.).

After describing the mid-IR sounding technique (Sect. 2), methane time series and trends (Sect. 3.1) will be presented, the MIR-GBM v1.0 retrieval strategy will be validated by comparing Zugspitze and Garmisch trend results, and the trend results will be related to earlier findings using other techniques (Sect. 3.2). Section 4 will provide a tentative explanation of the trend behavior derived for the 2007–2011 time period along with some conclusions.

2 Sounding technique

Time series of column-averaged dry-air mole fractions of methane (XCH_4) were retrieved from long-term FTIR solar absorption measurements at two northern mid-latitude sites, namely, the Zugspitze high-altitude site (47.42° N, 10.98° E, 2964 m a.s.l.) and Garmisch (47.48° N, 11.06° E, 743 m a.s.l.). The two sites are located only a few kilometers apart in horizontal distance. However, the IWV levels differ strongly (min/max = 0.2 mm/12.7 mm for Zugspitze, 1.9 mm/34.9 mm for Garmisch) because of the altitude difference. This helps to validate the methane retrievals and in particular to exclude a potential impact of water-vapor interference errors on the methane trend results.

The Zugspitze FTIR system has been conducting continuous measurements since 1995 as part of NDACC. It is operated by the Group "Variability and Trends" of IMK-IFU[1], Karlsruhe Institute of Technology, at the Zugspitze[2]. The FTIR system is based on a Bruker IFS125HR interferometer; details can be found in Sussmann and Schäfer (1997). The interferograms for the methane retrievals were recorded with an InSb detector using an optical path difference of typically 175 cm. 6 scans were averaged (≈7-

[1] Institute for Meteorology and Climate Research – Atmospheric Environmental Research, http://www.imk-ifu.kit.edu/atmospheric_variability.php

[2] Zugspitze site details can be found at http://www.imk-ifu.kit.edu/311.php

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min integration time). The pressure-temperature profiles necessary for the inversion were taken from the National Center for Environmental Prediction (NCEP) automailer. The Garmisch solar FTIR system was set up in 2004 at the Garmisch site[3] and is part of the Total Carbon Column Observing Network (TCCON, http://www.tccon.caltech.edu/) operating in the near-infrared for high-precision retrieval of column-averaged mixing ratios of carbon dioxide and methane. The system performs mid-IR NDACC-type measurements in parallel (in alternating mode on the time scale of several minutes). The latter are utilized for this study. The measurement settings for the Garmisch mid-IR methane measurements are the same as for the Zugspitze.

For inversion of XCH_4 from the mid-IR solar spectra, the MIR-GBM v1.0 retrieval strategy is applied, which minimizes interference errors from water vapor and HDO down to the ≈0.1 % level. The basic features of this strategy are given in Table 1, more details can be found in Sussmann et al. (2011).

3 Results

3.1 Time series and trends

Figure 1a shows the time series of monthly-mean column-averaged methane above Zugspitze and Garmisch. Typically, $n = 40$–60 individual column measurements per month are recorded on 6–8 clear sky days. Shaded bars in Fig. 1a indicate the statistical uncertainties of the monthly means calculated from the individual measurements ($\pm 3\sigma/sqrt(n)$). These suggest that statistically stable monthly means are obtained for the trend analysis.

The de-seasonalized time series (Fig. 1b) shows a significant increase during the 3-yr time interval [1996, 1998], followed by a period with no significant growth [1999, mid 2006]. Afterwards, a renewed strong increase can be observed, which has been persisting for 5 yr now [mid 2006, September 2011]. This behavior is quantified by a trend

[3] Garmisch site details can be found at http://www.imk-ifu.kit.edu/315.php

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analysis based on the approach described in Gardiner et al. (2008). See Table 2 for the resulting figures. Briefly, the approach augments a basic linear trend model applied to the indicated parts of the time series after subtraction of a fitted intra-annual function and uses least squares regression in conjunction with a bootstrap resampling of the residuals in order to determine confidence limits associated with the trend estimates. For the intra-annual model, a 3rd-order Fourier series is used.

As a main result from Table 2, the original anomaly of the period [mid 2006, 2008] with a Zugspitze trend of 6.6 [3.5, 9.8] ppb yr^{-1} and a Garmisch trend of 5.1 [2.0, 8.3] ppby yr^{-1} is found to persist also during the extended period [mid 2006, September 2011] investigated now (Zugspitze trend 4.8 [3.8, 5.9] ppb yr^{-1}, Garmisch trend 5.1 [4.2, 6.0] ppb yr^{-1}).

3.2 Validation and consistency with other results

Development of MIR-GBM v1.0 (Sussmann et al., 2011) was motivated by the finding that the standard retrieval strategy used within the NDACC network until then was subject to H_2O-CH_4 interference errors ranging up to 5 % at high-humidity (low-altitude) sites like Garmisch. This error may even affect the retrieved methane trends, e.g., if there is a trend in columnar water vapor at one site and no trend at another. A particular result from Table 2, namely, very good agreement of the FTIR trends retrieved with MIR-GBM v1.0 for Zugspitze and Garmisch, shall therefore be discussed: For the [mid 2006, 2008] period above Zugspitze, a trend of 4.9 ppb yr^{-1} with a 95 % confidence interval of [3.8, 6.0] ppb yr^{-1} is found, which agrees with 5.1 [2.0, 8.3] ppb yr^{-1} retrieved from Garmisch measurements for the same period. For the [mid 2006, September 2011] period, the trend of 4.8 [3.8, 5.9] ppb yr^{-1} above Zugspitze is close to the Garmisch trend of 5.1 [4.2, 6.0] ppb yr^{-1}. This excellent agreement between two neighboring FTIR sites has to be considered in the context of their altitude difference (more than 2000 m), as this implies that typically about two thirds of the total IWV are contained in the partial column between Garmisch and Zugspitze (see figures in Sect. 2). Given the fact that we found a significant trend of IWV in the Zugspitze time series (Sussmann

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et al., 2009), this agreement in methane trends proves that the trend results obtained with the MIR-GBM v1.0 are not influenced by H_2O-CH_4 interference errors.

The trend of 6.6 ppb yr^{-1} derived from our Zugspitze time series for the period [mid 2006, 2008] agrees qualitatively with earlier studies using in-situ data for both hemispheres, according to which the growth rate of atmospheric methane started to increase after 2006 with changes of 8.0±0.6 ppb in 2007 and 6.4±0.6 ppb in 2008 (update from Dlugokencky et al., 2009).

For a more quantitative intercomparison using SCIAMACHY retrievals of column-averaged mole fractions for the 30° N–90° N latitudinal band, we also calculated the Zugspitze trend for the period [mid 2006, mid 2009], i.e., 5.6 [3.1, 8.2] ppb yr^{-1}. This agrees well with the SCIAMACHY (WFM-DOAS v2.0) result of 6.6 ppb yr^{-1} for the same period (Table 3 in Schneising et al., 2011).

4 Discussion and conclusion

The methane increase persisting for 5 yr [mid 2006, September 2011] as documented in Fig. 1 and Table 2 is either the longest and largest positive trend anomaly since more than 25 yr when systematic observations began or marks the onset of a new period of strongly increasing CH_4 levels in the atmosphere. Our study cannot identify the mechanisms behind this increase, but our findings shall be discussed briefly in relation to previous studies on this subject and most reasonable causes shall be proposed.

Previous studies discussed possible causes of the increase in the years 2007 and 2008 (Rigby et al., 2008; Duglokencky et al., 2009; Bousquet et al., 2011; Frankenberg et al., 2011; Spahni et al., 2011; Montzka et al., 2011; van der Werf et al., 2011). Rigby et al. (2008) and Montzka et al. (2011) find a generally declining OH concentration between 2004 and 2007. However, the large but uncertain 4±14% drop between 2006 and 2007 inferred by Rigby is much larger than the small drop found by Montzka using several modeling approaches. Having in mind that atmospheric chemistry models can hardly infer large year-to-year changes in OH concentrations (Dentener et al., 2002),

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OH loss may be assumed to play a minor role in the recent methane increase. The biomass burning contribution also turned out to be insignificant, because no large CO anomaly is observed in 2007–2008 (van der Werf et al., 2010). Bousquet et al. (2011) invoke an increase in emissions from natural, mostly tropical wetlands in 2007–2008, but with a significant contribution from northern wetlands in 2007. The land surface models ORCHIDEE (Ringeval et al., 2010) and LPJ (Spahni et la., 2011) both show a positive trend in natural wetlands emissions between 2005 and 2008, which is mainly due to tropical precipitation changes associated with the La Niña event in these years and to boreal positive temperature anomalies in 2007.

The question is whether the continued increase shown in Fig. 1 for the years 2009, 2010, and 2011 can still be attributed to increased wetland emissions. The analysis of ECMWF ERA-INTERIM (ECMWF, 2011) precipitations and 2-m temperatures in the tropics reveals a contrasted picture for 2007–2011. While tropical wetland areas in South America and Asia experience increasing precipitations on the average with a drop in 2009 (Tropical Asia) and in 2010 (South America), decreasing precipitations are found over Africa. All together, tropical precipitations above wetland areas increase in 2007–2008, decrease in 2009, and have been increasing again since 2010. Meanwhile, tropical land temperatures have increased only slightly. As recent estimates of anthropogenic emissions are not yet available (EDGAR4 stops in 2005 and atmospheric inversion do not cover the past 2 yr), it is not possible to explain the 2009–2011 period of methane increase by the predominance of natural wetlands, although they probably play a significant role for increasing precipitations in 2007, 2008, and since 2010.

However, a persisting natural wetland anomaly over more than a few years may be in conflict with the understanding that wetland emissions typically cause inter-annual variability (i.e., 1–2 yr time scale), while more gradual changes on the time scale of several years would be due to changing anthropogenic emissions. In this context, it is worth considering the causes of the near-zero growth during 1999–2006 (Duglokencky, et al., 2003). While one might be tempted to attribute this trend stagnation to the collapse

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of industry in the former USSR, Bousquet et al. (2006) pointed out that global emissions continued to increase during this period in spite of this negative trend component. According to the EDGAR 4.1 database (European Commission 2009), anthropogenic emissions between [1999, 2005] totaled 31 Tg and were dominated by strongly increasing emissions in China, which is more than sufficient to explain the post-2006 anomaly in atmospheric concentrations. This leads to the question why no continued increase of methane had been observed after 1999 already, as was pointed out by Frankenberg et al. (2011). In contrast to the EDGAR inventory, Aydin et al. (2011) recently proposed on the basis of an ethane-based fossil fuel emission history that decreasing fossil fuel emissions of methane may explain the observed atmospheric slowdown. The answer by Bousquet et al. (2006) was that the expected increase had been masked from 1999 to 2005 by continuously decreased wetland emissions due to long-lasting drier conditions encountered in various regions of the Northern Hemisphere. According to this finding, Bousquet et al. stated in 2006 (just before the renewed methane increase was detected by measurements) that a renewed increase of global CH_4 emissions would have to be expected as soon as wetland emissions rise back to a normal level. This explanation is in line with recent simulations by the ORCHIDEE global dynamical vegetation model (Ringeval et al., 2010) and by the LPJ-WHyMe global dynamical vegetation model using model parameters derived from 2003–2005 inversions constrained by SCIAMACHY and surface network data (Spahni et al., 2011). The latter found a strong increase in ecosystem CH_4 emissions ($3.62\,Tg\,yr^{-1}$) for the period [2005, 2008] compared to emissions of $1.1\,Tg\,yr^{-1}$ during the [1900, 2008] period, although they pointed out that their simulations can only partly explain the long-term decline of the atmospheric growth rate during [1990, 2006].

According to the different possible interpretations, the renewed CH_4 increase after 2006 is expected to continue on the decadal time scale as long as no large-scale and long-term droughts will follow. Anyway, there is no need for an explanation of the actual methane trend by strongly increased emissions from stores of carbon in melting permafrost and from marine hydrates as a reaction to climate change. However, both

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remain to be potential sources of severely increasing methane emissions in the future, which should be closely monitored by remote sensing at the surface or from space.

Acknowledgements. We thank H. P. Schmid (IMK-IFU) for his continual interest in this work. Our work was performed as part of the ESA GHG-cci project under a subcontract with the University of Bremen. Earlier studies which contributed to this paper were funded by the EC within the HYMN project (contract 037048). We acknowledge support by the Deutsche Forschungsgemeinschaft and Open Access Publishing Fund of Karlsruhe Institute of Technology.

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Table 1. Characteristics of the strategy for retrieval of column-averaged methane from mid-IR solar spectra (MIR-GBM v1.0). For more details, see Sussmann et al. (2011).

micro windows (interfering species fitted)	2613.70–2615.40 (HDO, CO_2) 2835.50–2835.80 (HDO) 2921.00–2921.60 (HDO, H_2O, NO_2)
line list	HITRAN 2000 including 2001 update release
retrieval constraint	Tikhonov L_1
regularization strength α	optimized via L-curve/minimum diurnal variation (dofs[1] ≈2 degrees of freedom for signal)
altitude dependency of reg. strength	altitude constant on per-cent-vmr[2] scale
a priori vmr profiles	WACCM[3]
background fit	linear slope
retrieval quality selection	threshold (0.15 %) for rms-noise/dofs
calculation of column-averaged dry-air mole fractions	use 4-times-daily-NCEP[4] PTU profiles, interpolate to FTIR measurement time, calculate air column, subtract water vapor column
precision (1-σ diurnal variation)[5]	< 0.3 %
seasonal bias (H_2O/HDO-CH_4 interference error)	<0.14 %

[1] dofs – **d**egrees **o**f **f**reedom **f**or **s**ignal
[2] vmr – **v**olume **m**ixing **r**atio
[3] WACCM – **W**hole **A**tmosphere **C**hemistry **C**limate **M**odel
[4] NCEP – **N**ational **C**enter for **E**nvironmental **P**rediction
[5] for 7-min integration.

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Table 2. Existence and significance of trends of column-averaged mole fractions of CH_4.

		trend (ppb yr^{-1})	uncertainty interval (2.5th percentile, 97.5th percentile)[1] (ppb yr^{-1})	significant non-zero trend? (95 % confidence)
Zugspitze FTIR				
	[1996, 1998]	9.0	[3.2, 14.7]	yes
	[1999, mid 2006]	0.8	[−0.1, 1.7]	no
	[mid 2006, 2008]	6.6	[3.5, 9.8]	yes
	[mid 2006, Sep 2011]	4.8	[3.8, 5.9]	yes
Garmisch FTIR				
	[mid 2006, 2008]	5.1	[2.0, 8.3]	yes
	[mid 2006, Sep 2011]	5.1	[4.2, 6.0]	yes
SCIAMACHY[2] 30° N–90° N	[mid 2006, mid 2009]	6.6	–	–
Zugspitze FTIR				
	[mid 2006, mid 2009]	5.6	[3.1, 8.2]	yes

[1] underlying uncertainty distributions constructed via 5000 bootstrap resamplings for each trend
[2] WFM-DOAS version 2.0, number taken from Schneising et al. (2011).

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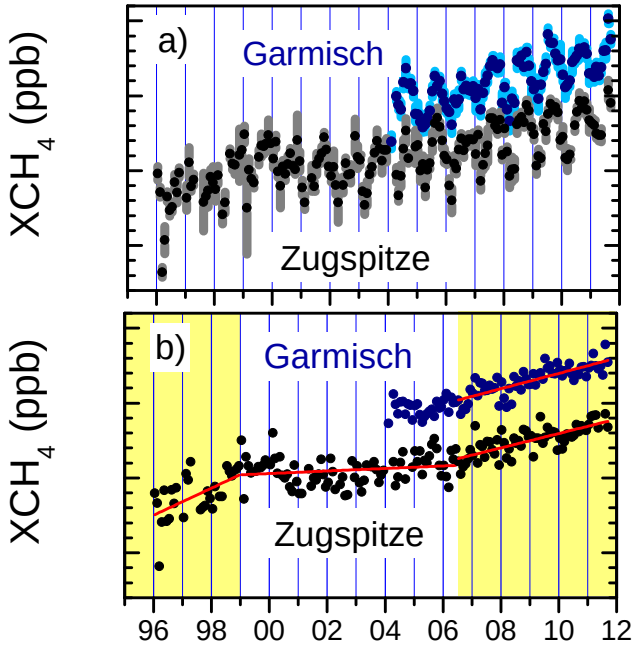

Fig. 1. **(a)** Time series of methane column-averaged mole fractions above Zugspitze and Garmisch (monthly means). Shaded bars indicate the statistical error of the monthly means calculated from the individual measurements (±3 $\sigma/sqrt(n)$), where n is the number of FTIR measurements per month. **(b)** De-trended time series and linear trends (red lines). See Table 2 for trend magnitudes and significance.

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