INTER-COMPARISON on TRANSPORT and CHEMISTRY

report on model inter-comparison performed within European Commission FP5 project EVERGREEN

("Global satellite observation of greenhouse gas emissions")





joint research centre EUROPEAN COMMISSIC

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CONTENTS

1	EX	ECUTIVE SUMMARY	2		
2	AT	MOSPHERIC MODELS	3		
3	²²² RN INTERCOMPARISON				
	3.1 3.2	²²² RN MODELLING PROTOCOL	4 4		
4	SF_6	INTERCOMPARISON	19		
	4.1 4.2	SF_6 Modelling protocol SF_6 results	19 19		
5	СН	4 INTERCOMPARISON	32		
	5.1 5.2	CH_4 Modelling protocol CH_4 results	32 32		
6	OH	INTERCOMPARISON	48		
	6.1 6.2	OH MODELLING PROTOCOL OH results	48 48		
7	RE	FERENCES	52		

1 Executive summary

A comprehensive inter-comparison of 5 atmospheric chemistry transport models (TM5, TM4, TM3, IMAGES, and LMDZ) has been performed. The main objective was to analyze differences in model transport, in particular vertical mixing (boundary layer and convective transport), synoptic variations, and large scale global circulation (including inter-hemispheric exchange and stratospheric tropospheric exchange (STE)). For this purpose simulations of various tracers with very different atmospheric lifetimes τ have been carried out: ²²²Rn ($\tau = 3.8$ days), SF₆ ($\tau = \sim 3000$ years), and CH₄ ($\tau = \sim 9$ years), using prescribed boundary conditions for all models. Furthermore, OH fields from various model simulations with full chemistry have been compared.

²²²Rn simulations show significant differences in vertical transport between models, leading to differences of simulated ²²²Rn concentrations near the surface of up to a factor of ~3. The TM5 and TM4 model have generally the highest ²²²Rn concentrations near the surface, while the other models tend to stronger vertical mixing. Comparison with in-situ measurements at 9 surface monitoring sites show that synoptic variations are simulated relatively well by all models which use (re)analyzed meteorological fields (i.e. all models except IMAGES, which is using monthly mean climatological fields). Comparison of TM5 and TM4 simulations (which have the same parameterization of atmospheric transport) illustrate that increasing horizontal model resolution significantly improves agreement with observations.

Simulations of SF₆ show significant differences in inter-hemispheric transport between the applied models, ranging between 6 and 12 months. This range is consistent with previous model inter-comparisons, e. g. within TransCom2 [*Denning et al.*, 1999]. STE is weaker and probably more realistic (15–16 months) in TM5, TM4, and LMDZ than in TM3 and IMAGES (7-8 months). The difference in STE between TM3 vs. TM5/TM4 is probably largely due to the different vertical resolution of the applied model versions.

CH₄ tracer simulations with prescribed OH fields were performed for TM5, TM4 and IMAGES. Consistent with the ²²²Rn simulations, TM5 and TM4 show higher CH₄ mixing ratios near the surface over CH₄ source regions compared to IMAGES. Both TM5 and TM4 simulate synoptic variations very well at most surface monitoring sites. Similar as for the ²²²Rn experiments agreement with CH₄ surface observations is improving with increasing horizontal model resolution. The large difference in STE between TM5/4 and IMAGES is also clearly reflected in the CH₄ simulations.

Furthermore, OH distributions have been compared from model simulations with full chemistry. For these simulations the applied models (TM5, TM4, IMAGES) used different emission inventories (representing typical standard configurations of the corresponding models). Simulated OH fields show significant differences near the surface, probably largely due to the applied different emission inventories (CO, NMHC, NO_x). In the free troposphere, however, the spatial OH distribution are relatively similar. In addition, also the seasonal OH variation is very consistent for all model runs. Global CH_4 + OH lifetimes in the range of 8.3 - 11.4 years have been calculated for the different OH fields. All models suggest 20-40% higher CH_4 lifetime in the SH, compared to NH.

2 Atmospheric models

The applied atmospheric transport and chemistry models are listed in Table 2.1.

model	TM5 (JRC)	TM4 ¹ (KNMI)	ТМ3 (МРІ)	IMAGES (BIRA-IASB)	LMDZ (CNRS-LSCE) external partner
horizontal resolution	6 x 4 (global) 1 x 1 (zoom over Europe)	3 x 2	5 x 3.8	5 x 5	3.75 x 2.5
vertical layers	25 hybrid	25 hybrid	19 sigma	25 sigma	38
advection	slopes scheme [<i>Russell and Lerner</i> , 1981]	slopes scheme [<i>Russell and Lerner,</i> 1981]	slopes scheme [<i>Russell and Lerner</i> , 1981]	Semi-Lagrangian (2nd order) [Smolarkiewicz and Rasch, 1991]	finite-volume second- order scheme [Van Leer , 1977; Hourdin and Armengaud, 1999]
vertical diffusion	PBL:[<i>Holtslag and Moeng</i> , 1991] FT: [<i>Louis</i> , 1979]	PBL: [<i>Holtslag and Moeng</i> , 1991] FT:[<i>Louis</i> , 1979]	[<i>Louis</i> , 1979]	PBL: [Muller and Brasseur, 1995]	PBL: Mass Flux Representation of Thermals [Hourdin et al., 2002]
convective cumulus clouds	[<i>Tiedtke</i> , 1987]	[<i>Tiedtke</i> , 1987]	[<i>Tiedtke</i> , 1987]	[Muller and Brasseur, 1995] [Costen et al., 1988]	[<i>Tiedtke</i> , 1987]
meteorology	ECMWF	ECMWF	NCEP	ECMF (climatological)	GCM, nudged to ECMWF
chemistry	- offline - CBM4 based chem	- offline - CBM4 based chem	- offline	- offline - IMAGES-chemistry	- offline - [Hauglustaine et al., 2004]
model reference	[<i>Krol et al.</i> , 2005]	[Dentener et al., 2003]	[Heimann and Koerner, 2003]	[Muller and Brasseur, 1995]	
²²² Rn	•	•	•	•	•
SF ₆	•	•	•	•	•
ОН	•	•		•	
CH₄	•	•		•	

Table 2.1: Applied atmospheric models

¹ in some plots of this report the previous name 'TM3_KNMI' is still used; however the applied model version is identical as described for 'TM4_KNMI'.

3 ²²²Rn intercomparison

3.1 ²²²Rn modelling protocol

²²²Rn emissions

Similar to other ²²²Rn studies constant ²²²Rn emissions are assumed over land surfaces (between -60° S and 60° N ²²²Rn emissions of 1 atom cm⁻² s⁻¹; between 60° N and 70° N emissions of 0.5 atom cm⁻² s⁻¹). For all other land surfaces (incl. Greenland and Antarctica) and for the ocean emissions are set to zero.

²²²Rn sink

radioactive decay: rate constant k = 2.11E-6 [s⁻¹]

Initialization and simulation period

simulation period: 2001 (1 full year, spin-up 1 month) initialization: ²²²Rn (01.12.2000) = 0.0

3.2 ²²²Rn results

The 3D distributions of simulated ²²²Rn concentrations are illustrated in Fig. 3.1 (surface), Fig 3.2 (free troposphere at 500 hPa), and Fig 3.3 (vertical distribution). In particular near the surface significant differences are visible between the models. The TM5 and TM4 model have generally the highest ²²²Rn concentrations near the surface, while the other models tend to stronger vertical mixing. In December, the LMDZ model has similar surface ²²²Rn concentrations as TM5 and TM4 over the extratropical NH landmasses, but much lower values over the tropics. The displayed surface values represent the concentrations of the lowest model layer. Therefore, beside differences in vertical mixing, also differences in vertical model resolution may play a role, e.g. when comparing TM5/TM4 with TM3. While many parameterizations are identical or similar between TM5/TM4 and TM3 (such as vertical diffusion in the free troposphere, and convective cumulus clouds), TM5/TM4 apply the diffusion parameterization of [Holtslag and Moeng, 1991] in the PBL, use ECMWF meteorological fields (compared to NCEP in TM3) and have higher vertical resolution. As expected 3D distributions of TM5 and TM4 are very similar (as for the other tracer experiments with SF₆ and CH₄), but effects due to the different horizontal resolution (TM5 $6^{\circ}x4^{\circ}$; TM4: $3^{\circ}x2^{\circ}$) are clearly visible near the surface. Model simulations have been compared with ²²²Rn measurements at 9 European and global monitoring sites (Fig. 3.4). Synoptic variations are simulated relatively well by all models which use (re)analyzed meteorological fields (i.e. all models except IMAGES, which is using monthly mean climatological fields). At the two European continental sites Freiburg and Milano, also the simulated average diurnal variations (mainly due to the diurnally varying boundary layer) agree very well with observations, in particular for TM5 and TM4. TM3 and LMDZ also show these diurnal cycles, but simulate lower mean concentrations than observations. In general very difficult to simulate are sites at the land-sea border (as Mace Head) and mountain sites. For some of these sites, sampling locations were slightly shifted in some models. E.g. at Mace Head, the sampling point has been shifted 2-3° westwards in some models (avoiding or reducing the effect of local ²²²Rn emissions of the

corresponding model grid cell), leading to significant improvements with observations.

Several mountain sites (in particular Zugspitze) exhibit significant influence of local mountain-valley winds, which with the applied models cannot be reproduced. Comparison of TM5 and TM4 simulations (which have the same parameterization of atmospheric transport) show that increasing horizontal model resolution significantly improves agreement with observations (Fig. 3.5). On the global domain TM4 achieves higher correlation with measurements than TM5 (resolution $3^{\circ}x2^{\circ}$ (TM4) vs. $6^{\circ}x4^{\circ}$ (TM5)), on the European zoom domain the correlation for TM5 is higher ($1^{\circ}x1^{\circ}$ (TM5) vs. $3^{\circ}x2^{\circ}$ (TM4)).



surface

December



Figure 3.1: ²²²Rn intercomparison: surface (top: June 2001; bottom: December 2001).



500.000hPa

December



Figure 3.2: ²²²Rn intercomparison: free troposphere (500 hPa) (top: June 2001; bottom: December 2001).







December



Figure 3.3: ²²²Rn intercomparison: vertical distribution (latitudinal averages) (top: June 2001; bottom: December 2001).



Figure 3.4: ²²²Rn intercomparison: Surface monitoring stations.



Figure 3.4: continued.



Figure 3.4: continued.

Figure 3.4: continued.

Figure 3.4: continued.

Figure 3.4: continued.

Figure 3.4: continued.

Figure 3.4: continued.

Figure 3.4: continued.

Figure 3.5: Correlation between observations and model simulations as function of latitude. Data points are separated for different TM5 domains (global $6^{\circ}x4^{\circ}$, European $3^{\circ}x2^{\circ}$, and European $1^{\circ}x1^{\circ}$. Within the European $1^{\circ}x1^{\circ}$ higher correlation is achieved by the TM5 model compared to TM4 (with globally uniform resolution of $3^{\circ}x2^{\circ}$), while outside the European zoom the TM4 achieves higher correlation (i.e. $3^{\circ}x2^{\circ}$ (TM4) vs. $6^{\circ}x4^{\circ}$ (TM5)).

4 SF₆ intercomparison

4.1 SF₆ Modelling protocol

SF₆ emissions

The EDGAR V3.2 SF₆ inventory for 1995 is used for the *whole* simulation period (1994-2001) (see Fig. 4.1).

SF₆ sink

the SF_6 sink is assumed to be zero.

Initialization and simulation period

simulation period: 2001 (1 full year, spin-up 1994-2000) initialization: Initial fields of SF₆ have been provided by MPI Jena (S. Körner), based on TRANSCOM simulations and representing atmospheric mixing ratios at 01.01.1994.

4.2 SF₆ results

The 3D distributions of simulated SF₆ mixing ratios are illustrated in Fig. 4.2 (surface), Fig 4.3 (free troposphere at 500 hPa), and Fig 4.4 (vertical distribution). Surface mixing ratios are significantly elevated close to the main SF₆ emission regions of North America, Europe, and Southeast Asia. Consistent with the ²²²Rn simulations, these enhancements are somewhat stronger in TM5 and TM4 than in the other 3 models.

Furthermore, significant differences in the NS gradients of SF₆ mixing ratios are visible. This gradient is weakest for the LMDZ model, indicating faster interhemispheric mixing than in other models.

We calculate the 3D interhemispheric exchange time T_{NS} [Denning et al., 1999] as:

$$T_{NS} = \frac{C_N - C_S}{\frac{d}{dt}(C_N - C_S) - \left(\frac{S_N}{M_N} - \frac{S_S}{M_S}\right)}$$
(eq 4.1)

where C, S, and M represent the (hemispheric) SF_6 mixing ratios, SF_6 emissions, and air masses, and the subscripts N and S refer to the northern and southern hemisphere. Monthly values of T_{NS} are shown in Fig. 4.6, yearly mean values in Table 4.1. This compilation confirms that LMDZ has a much faster interhemispheric exchange time (6.3 months) than the other models (TM5, TM4, TM3: 9.9-10.4 months; IMAGES: 12.5 months). This overall range is similar to the range for the models of the TransCom2 intercomparison (6.6-15.1 months) [*Denning et al.*, 1999]. Fig 4.4 also shows large differences in the vertical SF_6 distribution, in particular in the upper troposphere and stratosphere, indicating significant differences in the stratospheric-tropospheric exchange time (T_{STE}). We calculate T_{STE} as:

$$T_{STE} = \frac{C_T - C_S}{\frac{d}{dt}C_S}$$
(eq 4.2)

from the mixing ratios C in the troposphere (T) and stratosphere (S), and the stratospheric increase (and assuming a constant tropopause at 150 hPa). STE is weaker (15.1–16.6 months) in TM5, TM4, and LMDZ than in TM3 and IMAGES (7.9-8.7 months). The difference in STE between TM3 vs. TM5/TM4 is probably largely due to the different vertical resolution of the applied model versions.

Measurements of SF₆ by balloons [*Patra et al.*, 1997] and satellites [*Rinsland et al.*, 2005] showed typical gradients between the middle stratosphere and the troposphere in the order of 0.8 ppt (0.7-1.0 ppt). This is similar to the gradient simulated by TM5, TM4, and LMDZ (~0.8 ppt), while TM3 (~0.4 ppt) and IMAGES (~0.2 ppt) have much smaller gradients (Figure 4.4). Therefore, the greater T_{STE} of TM5, TM4, and LMDZ seems more realistic. However, a more detailed analysis will have to take into account the variation of the vertical stratospheric SF₆ gradient with latitude, and also with time.

Comparison of model simulations with surface measurements are shown in Fig. 4.5. The difference of model simulations between NH sites and SH sites (e.g. compare Barrow with South Pole) is reflecting the different T_{NS} (Table 4.1) of the models, leading to the smallest gradient between these two stations for the LMDZ model. It should be emphasized, however, that the surface mixing ratios (in particular in the NH) are also significantly influenced by the vertical mixing (see also [*Denning et al.*, 1999] for comparison of 1D, 2D, and 3D based calculations of T_{NS}).

Furthermore, it is interesting to note that at most background sites synoptic variations of SF_6 are very small, or not present at all (consistent between observations and simulations). An exception is station Schauinsland where significantly elevated SF_6 mixing ratios are observed during certain synoptic conditions. These are very well captured by the TM5, TM4 and TM3 model (with correlation coefficients of 0.41-0.51).

model	TM5 (JRC)	TM4 (KNMI)	TM3 (MPI)	IMAGES (BIRA-IASB)	LMDZ (CNRS-LSCE) external partner
T _{NS} [months]	10.4	9.9	10.0	12.5	6.3
T _{STF} [months]	16.6	16.3	8.7	7.9	15.1

Table 4.1: Derived interhemispheric exchange time (T_{NS}) and stratospheric-tropospheric exchange time (T_{STE})

Figure 4.1: SF₆ emissions

surface

December

Figure 4.2: SF_6 intercomparison: surface (top: June 2001; bottom: December 2001). Note change in color scale (to account for atmospheric increase).

500.000hPa

December

Figure 4.3: SF_6 intercomparison: free troposphere (500 hPa) (top: June 2001; bottom: December 2001). Note change in color scale (to account for atmospheric increase).

vertical distribution

December

₆ intercomparison: vertical distribution (latitudinal averages) (top: June 2001; bottom: December 2001). Note change in color scale (to account for atmospheric increase).

Figure 4.5: SF₆ intercomparison: Surface monitoring stations.

Figure 4.5: continued.

Figure 4.5: continued.

Figure 4.5: continued.

Figure 4.5: continued.

Figure 4.5: continued.

Figure 4.6: SF₆ intercomparison: SF₆ intercomparison: Upper panel: Interhemispheric exchange time T_{NS} (eq. 4.1); lower panel: stratospheric-tropospheric exchange time T_{STE} (eq. 4.2).

5 CH₄ intercomparison

5.1 CH₄ Modelling protocol

CH₄ emissions

CH₄ emission were taken from [*Bergamaschi et al.*, 2005] (a priori emissions for year 2001; see Fig. 5.1).

CH_4 sink

 CH_4 + OH sink was used as described in [*Bergamaschi et al.*, 2005]. This OH distribution is also illustrated in section 6 ("TM5_JRC_offline"). In the stratosphere, also CH₄ destruction by O(1D), CI is taken into account [*Bergamaschi et al.*, 2005; *Brühl and Crutzen*, 1993].

Initialization and simulation period

simulation period: 2001-2003 (3 full years + spin-up 1 year), i.e. model start: 01/01/2000 model end: 31/12/2003 initialization: Initial fields of CH₄ (for 01.01.2000) have been provided by JRC (P. Bergamaschi)

5.2 CH₄ results

 CH_4 simulations were performed with the TM5, TM4 and IMAGES model. Despite the prescribed CH_4 sinks, the different 3D distibutions of CH_4 in the models lead to a small drift in CH_4 mixing ratios of IMAGES vs. TM5/TM4 of about 20 ppb within 3 years. To facilitate comparison of plots with have subtracted this offset of 20 ppb for the IMAGES results in all plots (for year 2003).

The 3D distributions of simulated CH₄ mixing ratios are illustrated in Fig. 5.2 (surface), Fig 5.3 (free troposphere at 500 hPa), and Fig 5.4 (vertical distribution). The CH₄ distribution reflects the major differences between the models as seen from the ²²²Rn and SF₆ simulations:

- TM5 and TM4 have higher surface CH₄ mixing ratios than IMAGES over CH₄ source regions (due to the weaker vertical mixing).
- TM5 and TM4 have a slightly smaller inter-hemispheric gradient than IMAGES due to the somewhat higher inter-hemispheric exchange (i.e. smaller T_{NS}).
- TM5 and TM4 have a much more pronounced vertical gradient in the upper troposphere and stratosphere due to the significantly higher T_{STE}.
- TM5 and TM4 simulations are very similar (confirming the consistency between the two models) but also show the effect of the different horizontal resolution (resolution 3°x2° (TM4) vs. 6°x4° (TM5)), in particular over source regions.

Fig. 5.5 shows comparison with flask measurements from the NOAA network, and Fig. 5.6 comparison with continuous measurements from different networks (NOAA, AGAGE, UBA). TM5 and TM4 generally simulate the synoptic variability rather well

and achieve overall very good correlations with observations (r=0.57-0.59; Fig. 5.7). As for ²²²Rn a clear improvement is visible with increasing model resolution. On the global domain TM4 achieves higher correlation with measurements than TM5 (resolution $3^{\circ}x2^{\circ}$ (TM4) vs. $6^{\circ}x4^{\circ}$ (TM5)), on the European zoom domain the correlation for TM5 is higher ($1^{\circ}x1^{\circ}$ (TM5) vs. $3^{\circ}x2^{\circ}$ (TM4)).

Figure 5.1: CH₄ emissions

TM4_KNMI

IMG_BIRA

surface

Figure 5.2: CH₄ intercomparison: surface (top: June 2003; bottom: December 2003).

June

TM5_JRC

35

16.4E2 16.2E2 CH, [ppb]

IMG_BIRA

December

bottom: December 2003).

16.5E2 16.4E2 16.2E2

TM4_KNMI

200

400

600

800

1000

200

400

600

800

1000

-90

-60

-30

p [hPa]

-90

-60

IMG_BIRA

-30

0 latitude

0 latitude

30

30

60

90

90

60

p [hPa]

December

Figure 5.4: CH₄ intercomparison: vertical distribution (latitudinal averages) (top: June 2003; bottom: December 2003).

June

⁴ intercomparison: surface monitoring sites (NOAA flask sampling sites).

Figure 5.5: continued.

Figure 5.5: continued.

Figure 5.5: continued.

Figure 5.6: CH₄ intercomparison: surface monitoring sites: high frequency measurements from various networks (NOAA, AGAGE, UBA).

Figure 5.6: continued.

Figure 5.6: continued.

Figure 5.6: continued.

Figure 5.7: Correlation between observations and model simulations as function of latitude. Data points are separated for different TM5 domains (global $6^{\circ}x4^{\circ}$, European $3^{\circ}x2^{\circ}$, and European $1^{\circ}x1^{\circ}$. Within the European $1^{\circ}x1^{\circ}$ higher correlation is achieved by the TM5 model compared to TM4 (with globally uniform resolution of $3^{\circ}x2^{\circ}$), while outside the European zoom the TM4 achieves higher correlation (i.e. $3^{\circ}x2^{\circ}$ (TM4) vs. $6^{\circ}x4^{\circ}$ (TM5)).

6 OH intercomparison

6.1 OH Modelling protocol

For this comparison, no specific modelling protocol was defined. Instead models have been applied in their typical configuration. Simulation period was year 2001.

6.2 OH results

The 3D distributions of simulated OH concentrations are illustrated in Fig. 6.1 (surface), Fig 6.2 (free troposphere at 500 hPa), and Fig 6.3 (vertical distribution). The OH distribution named "TM5_JRC_offline" has been used for the CH₄ intercomparison (section 5) and has been used in recent CH₄ inversion studies [*Bergamaschi et al.*, 2006; *Bergamaschi et al.*, 2005]. It has been calibrated with methyl cloroform, resulting in a mean tropospheric CH₄ lifetime vs. OH of 9.4 years, very close to the TAR recommended value of 9.6 years [*IPCC*, 2001]. For all other OH fields, no calibration has been performed. The resulting CH₄+OH lifetimes (turnover time for whole atmospher) are compiled in Table 6.1 and are in the range of 8.3 - 11.4 years. All models suggest 20-40% higher CH₄ lifetime in the SH, compared to the NH.

Simulated OH fields show significant differences near the surface (Fig. 6.1), probably largely due to the applied different emission inventories (CO, NMHC, NO_x). In the free troposphere, however, the spatial OH distribution are relatively similar (Fig. 6.2 and 6.3). Furthermore, the seasonal OH variation is very consistent for all model runs.

stratosphere) and ratio of lifetimes	stratosphere) and both hemispheres, T_{NH+SH} , for the individual hemispheres (T_{NH} , T_{SH}) and ratio of lifetimes for both hemispheres (T_{SH}/T_{NH})						
model	IMAGES	TM4	TM5	TM5_offline			

Table 6.1: Derived CH4+OH lifetimes for the whole atmosphere (i.e. troposphere +

model	IMAGES (BIRA-IASB)	TM4 (KNMI)	TM5 (JRC)	TM5_offline (JRC)
T _{NH+SH}	9.9	11.4	8.3	10.2 ¹
T _{NH}	8.5	10.6	7.7	9.4
T _{SH}	12.1	12.2	9.0	11.2
T _{SH} /T _{NH}	1.4	1.2	1.2	1.2

tropospheric CH₄+OH lifetime: 9.4 years

surface

Figure 6.1: OH intercomparison: surface (top: June; bottom: December).

Smo

¥

December

500.000hPa

Figure 6.2: OH intercomparison: free troposphere (500 hPa) (top: June; bottom: December).

vertical distribution

December

Figure 6.3: OH intercomparison: vertical distribution (latitudinal averages) (top: June; bottom: December).

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Abstract

A comprehensive inter-comparison of 5 atmospheric chemistry transport models (TM5, TM4, TM3, IMAGES, and LMDZ) has been performed. The main objective was to analyze differences in model transport, in particular vertical mixing (boundary layer and convective transport), synoptic variations, and large scale global circulation (including inter-hemispheric exchange and stratospheric tropospheric exchange (STE)). For this purpose simulations of various tracers with very different atmospheric lifetimes τ have been carried out: ²²²Rn ($\tau = 3.8$ days), SF₆ ($\tau = ~3000$ years), and CH₄ ($\tau = ~9$ years), using prescribed boundary conditions for all models. Furthermore, OH fields from various model simulations with full chemistry have been compared.

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