REanalysis of the TROpospheric chemical composition over the past 40 years
A long-term modelling study of tropospheric chemistry

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Work Package 3

RETRO report

Report on Process Studies

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Deliverable D3.4

Contents

1. Introduction

This study complements the studies of model performance for seasonal variations reported in RETRO deliverable D3-2. This report focuses on model performance over shorter periods of time that those investigated in the D3-2. By looking at shorter timescale we can learn about the models' detailed treatment of the processes which are of importance for the case studies chosen.

The three studies here investigate three very different aspects of model performance: the models ability to reproduce a stratospheric intrusion, a long range transport event and the large East Asian fires of late 1997.

2. Stratospheric intrusion

On the 25th of February 1997, the surface ozone record from Mauna Loa, Hawaii (19° 31' 48" N, 155° 34' 48" W, 3397 m asl) exhibits a pronounced maximum of more than 120 ppb. This maximum lasted only for a few hours before the concentrations fell back to background values around 50 ppb. Such a signature is typical for a stratospheric intrusion event, and these are believed to be responsible for the majority of the net influx of ozone from the stratosphere into the troposphere. We chose this episode in February 1997 to test the models' ability to simulate the transport and the dilution of ozone in a large-scale weather pattern. Figure 1 shows the observed and modelled ozone concentrations at Mauna Loa. Also shown for all models except LMDz-INCA is the stratospheric ozone tracer O3S, i.e. a tracer which is set to the current stratospheric ozone concentration above the model tropopause and advected in the same manner as the normal ozone tracer. Within the troposphere, O3S experiences the same chemical loss as the normal ozone tracer, but it is not affected by photochemical production. Even though this cannot be a perfect measure, this diagnostics provides some quantitative estimate for the contribution of stratospheric air to the simulated ozone concentration.

As noted elsewhere (D3-2), LMDz-INCA shows the best skill in reproducing the observed background ozone levels followed by TM. p-TOMCAT exhibits the strongest high-bias with typically 20-30 ppb above the measurements. While the measurements show only one distinct ozone peak on the 25th of February around 03:00 h GMT, most models simulate a double peak with two intrusion events on the 24th of February between 00:00 h GMT and 06:00 h GMT and on the 25th of February between 06:00h GMT an d15:00 h GMT. The relationship of these events with stratospheric intrusions can clearly be seen from the O3S tracers. LMDz-INCA is the only model, which simulates a single peak, however, the amplitude of this peak is much smaller than in the other models or in the observations (about 10-15 ppb instead of more than 50 ppb). The other models exhibit peak amplitudes between 20 and 50 ppb. The observed peak has a symmetric shape and lasts only about 3 hours. Most of the modelled peaks are also symmetric, but their width is typically around 6 hours. It must be noted, however, that the temporal and spatial sampling of the time series was differing between models: some models (MOZECH, TM4) sampled the time series from archived 3-hourly "instantaneous" values, while other models used 6-hourly

averaged output (LMDz, p-TOMCAT). Also some models interpolated their results to the exact location of the Mauna Loa station (e.g. LMDz-INCA, TM4), while others sampled the nearest neighbouring grid box (e.g. MOZECH, p-TOMCAT, UiO).

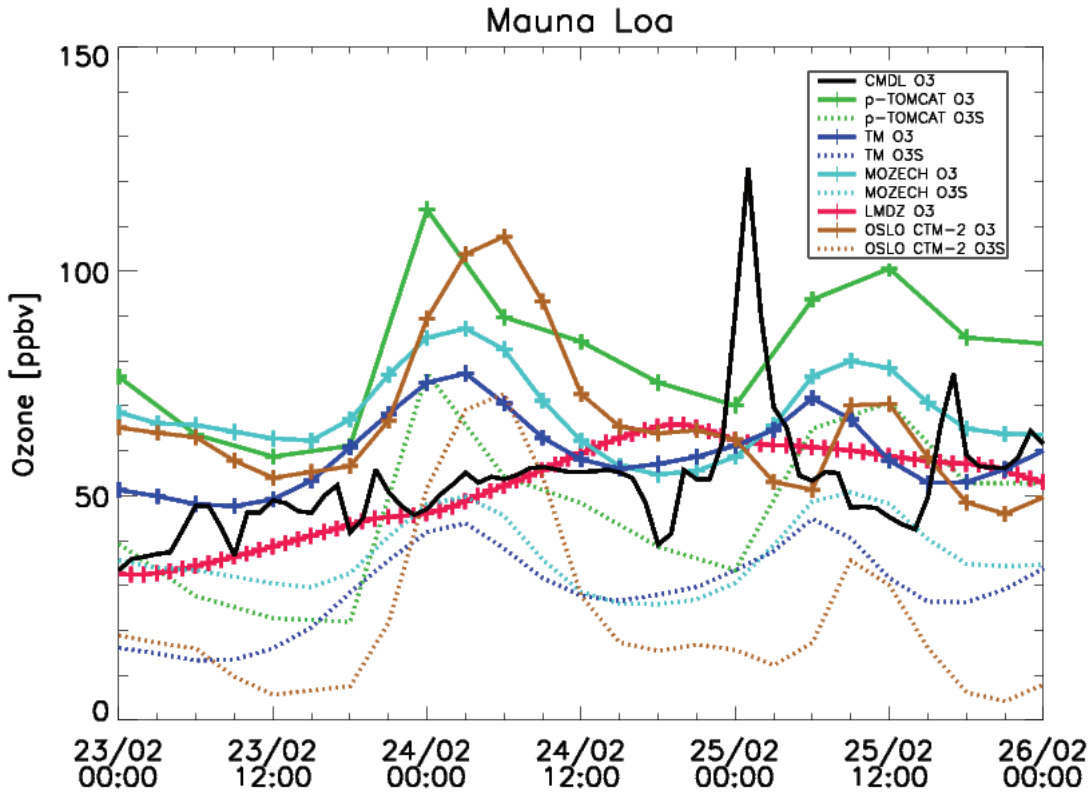

Figure 1. Ozone concentrations at Mauna Loa Observatory for 24-26/2/1997

In order to understand the reasons for the discrepancies between simulated and observed ozone concentrations, we interpolated 3-dimensional fields from all models onto a common vertical grid (1 km altitude) and plotted a sequence of horizontal and vertical cross sections (Figures 2 and 3). To obtain a clearer picture of the dynamical evolution of the intrusion event, we also generated animated 3D sequences. An example snap shot for MOZECH is shown in Figure 5.

As demonstrated by Figure 2, MOZECH and LMDz-INCA show qualitatively very similar features. Both models simulate an S-shaped intrusion which slowly rotates counter-clockwise north east of Hawaii. p-TOMCAT and UiO-CTM2 exhibit a pronounced double-peak with the stronger maximum north east of Hawaii and a weaker maximum to the south. In TM, one can only discern one elevated ridge in north-south direction. However, it should be noted that TM results are daily averages.

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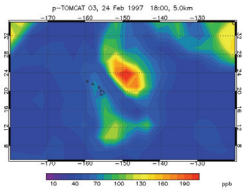
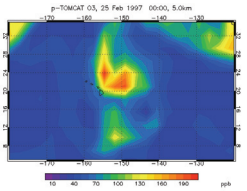
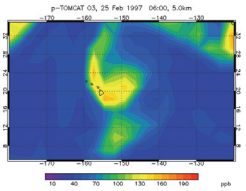
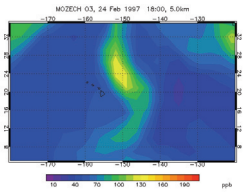
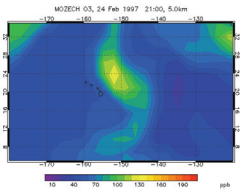
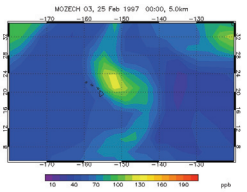
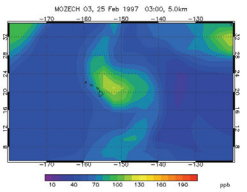
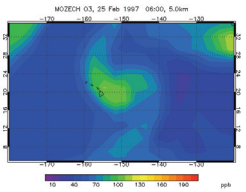
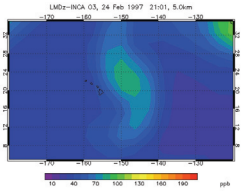
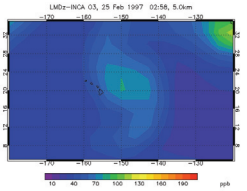
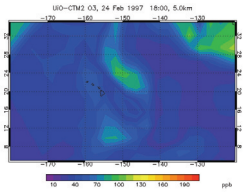
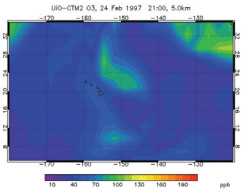
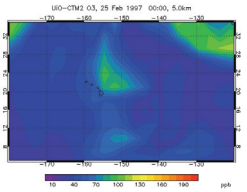
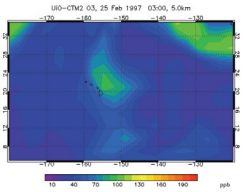
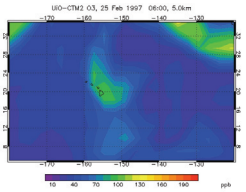
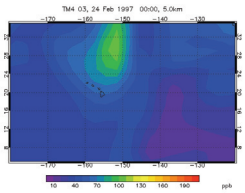
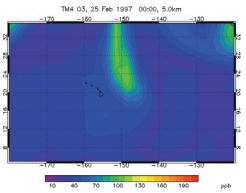
a					
b					
c					
d					
e					

Figure 2: Time series of the stratospheric intrusion event on February, 25th, 1997 as simulated by the five models (a) p-TOMCAT, (b) MOZECH, (c) LMDz-INCA, (d) UiO-CTM2, (e) TM. All model results were interpolated to a common altitude grid. Shown here is the 5 km level. Note that TM results are 24-hour averages.

With respect to the vertical structure of the intrusion (Figure 3), one first has to note that all models reproduce a strong feature near 150 W. The maximum ozone concentration inside the intrusion is above 200 ppb in all models. Furthermore, all models exhibit an ozone minimum between 10 and 15 km altitude and 140-180 E. With the exception of LMDz-INCA, all models predict ozone concentrations around 10 ppbv in this area. There is also a secondary maximum between 3 and 5 km altitude and 140-160 E, which is again weakest in LMDz-INCA and strongest in p-TOMCAT. The shape of the main intrusion differs somewhat between the models, although the agreement is remarkably good. p-TOMCAT and MOZECH show a minimum to the east of the main feature, which is absent in UiO-CTM2 and TM. In LMDz-INCA, ozone concentrations above 12 km altitude and east of 150 E are generally lower than in the other models, presumably due to a higher tropopause altitude.

Even though the shape of the intrusion is similar, the vertical and horizontal gradients of the ozone concentrations are rather different. It is therefore not surprising that the time series of ozone concentrations at one specific location (below the main intrusion) varies strongly between the models.

As shown in Figure 4, the contribution of stratospheric ozone to the simulated ozone concentrations differs markedly between the models. While the maximum contribution at 5 km altitude is always at least 80%, the influence on the region outside the intrusion varies from below 10% for UiO-CTM2 to more than 40% for MOZECH. This kind of discrepancy has not been demonstrated before to our knowledge.

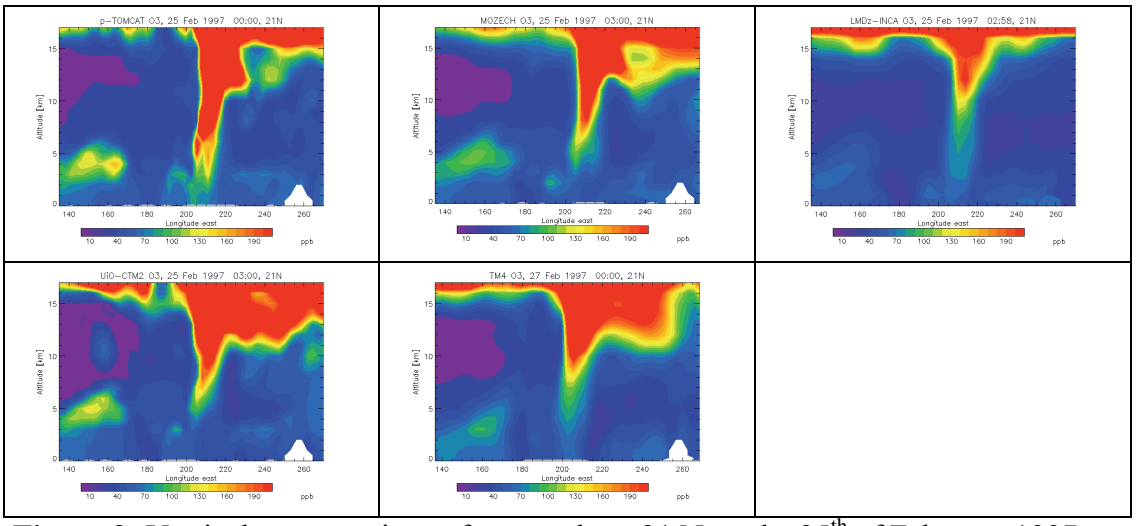


Figure 3: Vertical cross sections of ozone along 21 N on the 25th of February 1997, 03:00 h GMT. Top row from left to right: p-TOMCAT, MOZECH, LMDz-INCA, bottom row: UiO-CTM2 and TM. Note that p-TOMCAT results are for 00:00 h GMT and TM results are a 24-hour average.

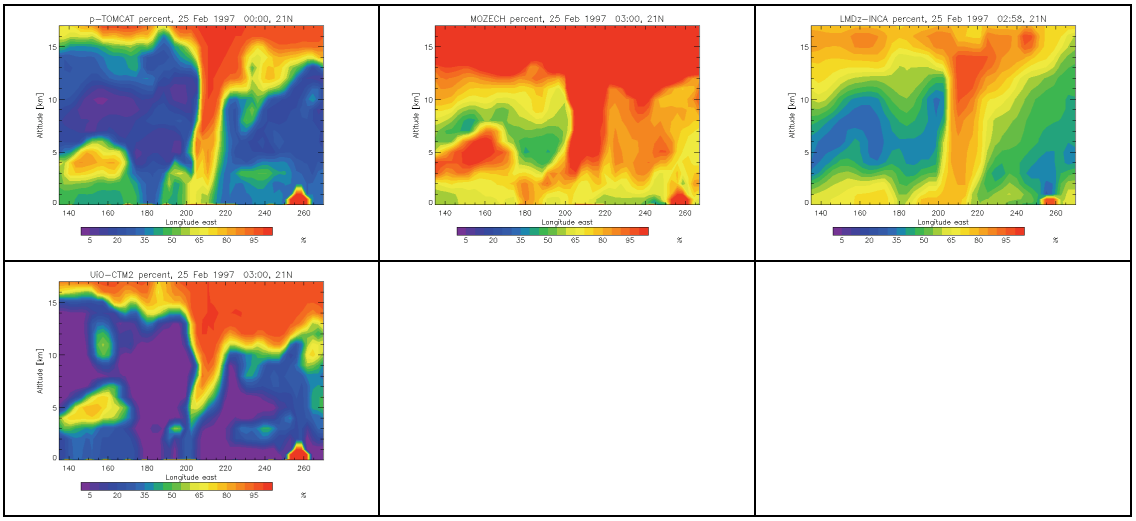


Figure 4: Vertical cross sections of the percent contribution of stratospheric ozone to the simulated ozone concentrations. Same location and time and same order of models as in Figure 3. There was no O3S output available for the TM model

It remains unclear why most models generate two peaks when the observations show only one. Since this is a common feature among the models, this may be an artefact of the ERA-40 data. Potentially, a closer inspection of ERA-40 fields for this episode could help reveal the causes for this discrepancy, but this is beyond the scope of this study.

In summary it must be stated, that all models are able to qualitatively capture an isolated short-term intrusion event and that most models also capture the observed concentration enhancements at Mauna Loa with a lag time of ±1 day and about 50% accuracy. However, the details of the intrusion and its impact on the regional ozone budget can be quite different.

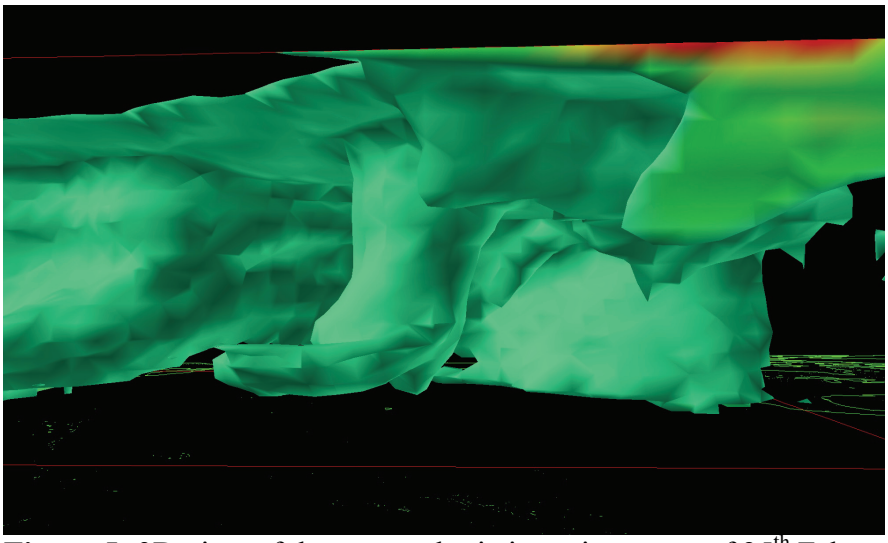


Figure 5: 3D view of the stratospheric intrusion event of 25th February 1997 over Hawaii as simulated by the MOZECH model. The greenish surface depicts the 60 ppb ozone contour.

3. Long Range Transport

Wenig et al. (2003) presents a case study of a plume of NO_x from the Highveld region which was advected across the Indian Ocean towards Australia. Using the tracer model FLEXPART (Stohl et al., 2001) they were able to gain good qualitative agreement with the satellite observations of the plume. Here we reproduce this study but using the retrieval from the University of Bremen (Richter et al., 2005) and comparing the results to the models in the RETRO study.

This is a challenging study for two sets of reasons. Firstly there are the limitations of the GOME satellite data: in particular the low frequency with which any particular part of the earth surface is observed (approximately every three days); also the issue of clouds – normally pixels with a cloud cover greater than 0.2 are ignored in the Bremen retrieval but for this study we need the maximum possible coverage so both cloud free and cloudy pixels have been used, which implies that in some areas the NO_2 columns will be underestimated – and finally the large uncertainties associated with these retrievals (see e.g. van Noije et al., 2006) . The second set of reasons why it

is a significant challenge lie with the models: all models in this study have a far lower resolution than the observations (320 km x 40 km) and it is difficult to represent the detailed transport at these resolutions; the meteorological situation itself was a complex one and as NO_2 is a short lived species it is critical to model all its sources and sinks correctly. However, the same reasons that make the case a challenging episode for models to simulate make it an excellent test case.

Figure 2 shows both the GOME and model columns from the period 8-14th May 1998. As in the retrieval presented by Wenig et al. plumes can be seen starting from the heavily industrialised Highveld region in South Africa and reaching out over the Indian Ocean. There are several features of the meteorology and type of emissions in this region which favour the satellite observation of this plume. Emissions from this region are very high – in the GEIA inventory this region has the second highest emissions worldwide. The region is elevated and many of the emissions come from high stacks which helps prevent the removal of the emissions by dry deposition and also improves the sensitivity of the satellite measurements. An anti-cyclonic system dominated until the 6th of May which allowed emissions to accumulate after which time a low pressure system caused large pressure gradients to develop and the emissions were rapidly carried over the Indian ocean in a south-easterly direction.

In order to improve the spatial coverage of the satellite for this period, the normal cloud screening algorithm is not used in this retrieval. This means that the NO_2 columns from the satellite may be underestimated in regions of high cloud cover and this must be taken account of when the model results are being compared to GOME data. Therefore Figure 2 also shows the GOME columns only for pixels with a cloud fraction of 0.2 or less. The plot also shows the GOME data regridded to 2.5 x 2.5 degrees resolution to be more comparable with the model resolution.

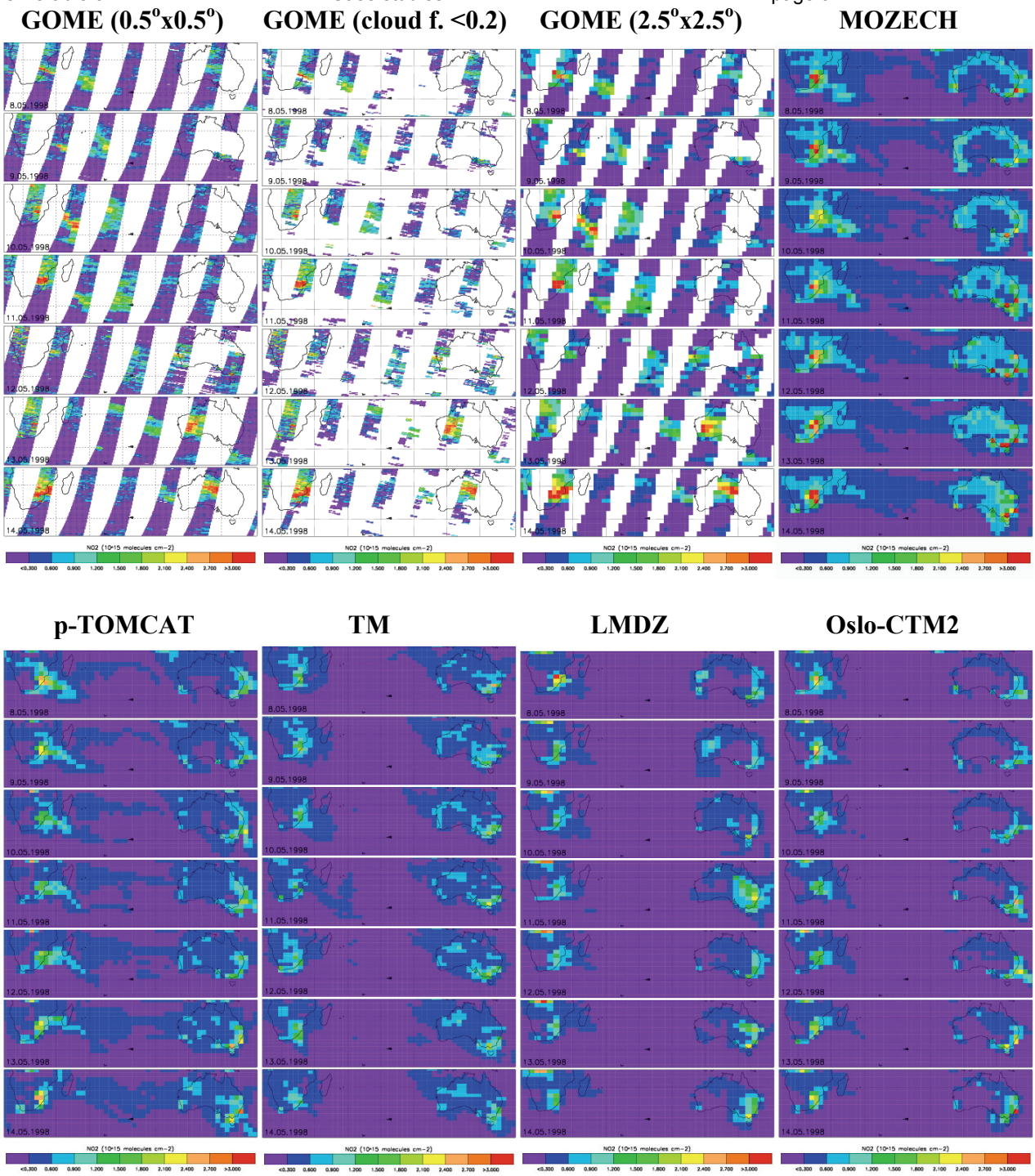


Figure 2. Top: GOME retrievals of NO_2 columns for 8-14/5/1998; MOZECH; Bottom: p-TOMCAT, TM, LMDZ, Oslo-CTM2

The columns of NO_2 over the Highveld in GOME are typically around 3 x 10^{15} molecules cm^{-2}. The only model which has such high values over this region is MOZECH while all other models have lower columns. Given that all the models use the same emissions for this study these differences are the result of other processes in the models such as the treatment of deposition, chemistry, convection and the boundary layer parameterisation. Another key factor is the height at which these emissions are introduced into the models.

In general, none of the models have NO_2 columns over the ocean as high as seen in the GOME retrieval. The maximum columns in the region of the plumes in the middle of the Indian Ocean are seen in the p-TOMCAT and MOZECH models and are between 0.3-0.6 x 10^{15} molecules cm^{-2}. In contrast the retrieval shows plumes with concentrations >2 x 10^{15} molecules cm^{-2}. In the p-TOMCAT and MOZECH models the plumes are located in approximately the correct locations but appear to be more disperse. These problems could indicate that the modelled transport is incorrect. However the reasonable location of the plumes in p-TOMCAT seems to suggest that either the modelled emissions from the Highveld region are too low, in agreement with the study by van Noije et al. (2006), or the modelled lifetime of NO_2 is too low.

4. 1997 Wildfires Event

From September to November 1997 massive wildfires burned in Indonesia, Malaysia and Papua New Guinea. These fires were a result of land clearance, arson and accidental fires and were made worse by a prolonged drought, land drainage and logging (Duncan et al., 2003 and reference therein). There are large uncertainties in the amount of pollution emitted by these fires. For example Yokelson et al. (1997) estimated the total carbon emissions from the fires in Indonesia at between 80 and 2570 Tg C. Levine et al. (1999) estimated that emissions of CO from Sumatra and Borneo during the period September-October 1997 were 38-114 Tg (CO), while Duncan et al. used emissions of 124 Tg (CO) for the period September to November for Borneo, Sumatra and New Guinea. In the RETRO fire emissions inventory (Schultz et al., 2006), CO emissions for this episode are estimated as 370 Tg (CO). This large value is due to the inclusion of peat fires which have been neglected in most earlier studies (cf. Page et al., 2002). Figure 3 below compares the RETRO CO emissions inventory for September 1997 and 1998. It can be seen that there are far higher emissions of CO in 1997 than in 1998.

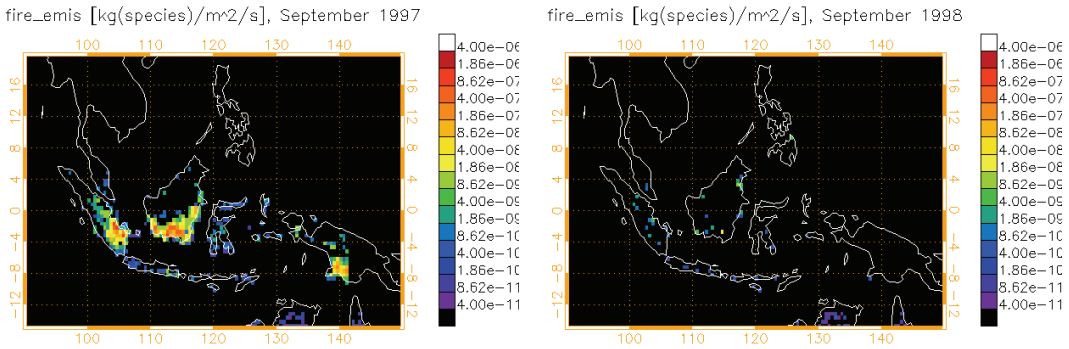


Figure 3. RETRO emission inventory for fire emissions of CO, September 1997 and 1998.

Figure 4 below shows the surface CO from all five RETRO models for 1997 and 1998. All models show an increase in surface CO over Indonesia for 1997 compared to 1998. This increase is smallest for MOZECH[§] and largest for the TM model. Differences in model chemistry and the horizontal/vertical transport schemes are the most likely explanation of the differences, given that the models are all using the same emissions of CO for this model run. The difference may also be related to the emission height distribution for biomass burning. The recommended height profile for wildfire emissions (not used by all models) in some places releases 12.8% of the fire emissions between 6 and 7 km, and lower fractions even higher up in the troposphere (up to 12 km).

Duncan et al. (2003) also used tagged tracers to show that increases in CO seen in the CMDL flask network observations at Mahe Island in the Seychelles were due to the wildfires in Indonesia. Increases in CO at Samoa of 50 to 100 ppb due to emissions of

[§] After this analysis had been completed, an error was found in the MOZECH simulations from 1992-1999. Due to a file transfer problem, a wrong fire inventory with far lower emissions was used and renders the MOZECH results for this study useless.

CO from Indonesia were modelled. Figure 5 below shows measurements of CO from Samoa and the Seychelles compared to the results from the models.

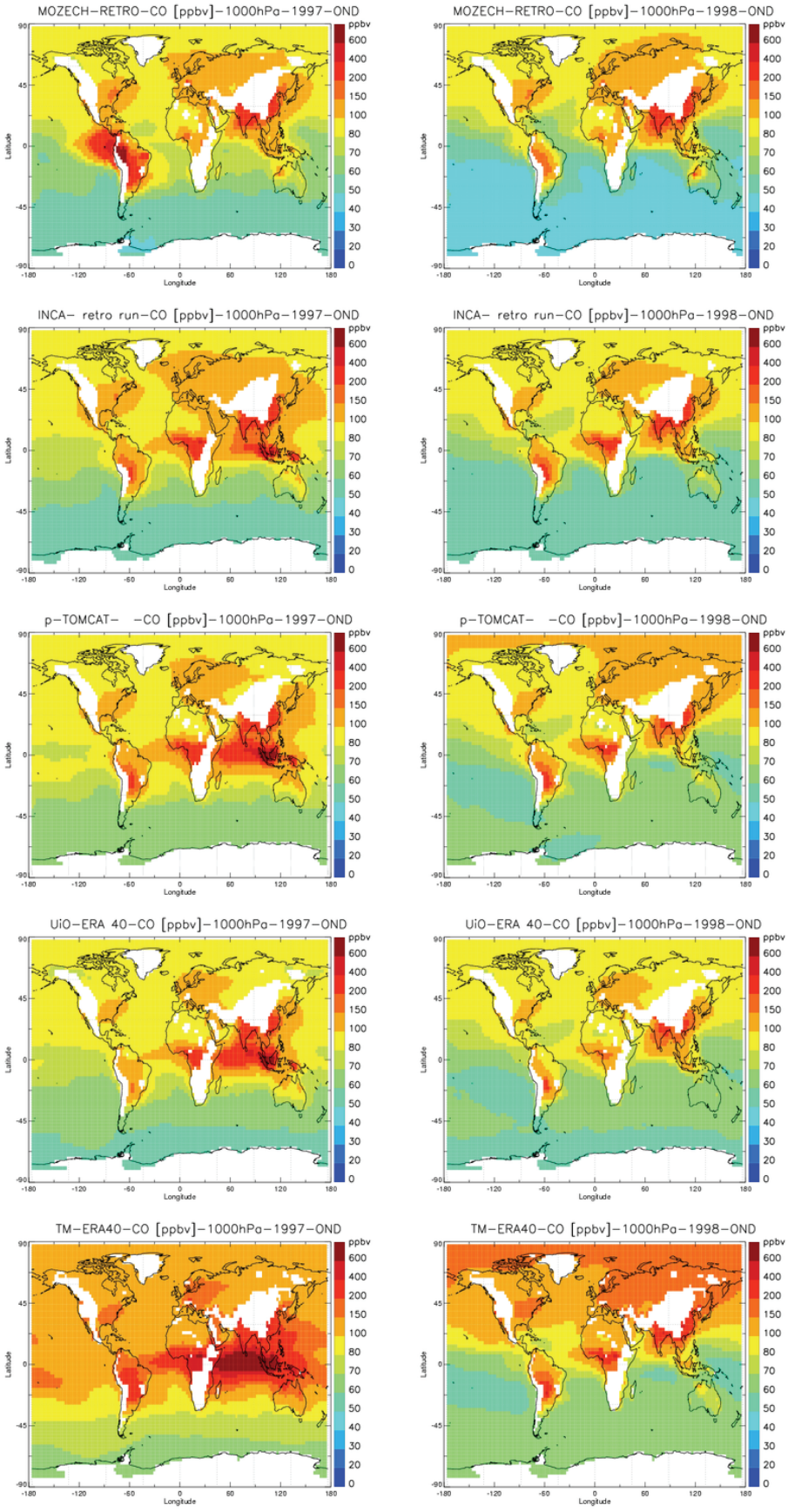


Figure 4. Modelled surface CO for October–December 1997 and 1998. Note that MOZECH results are based on erroneous fire emissions

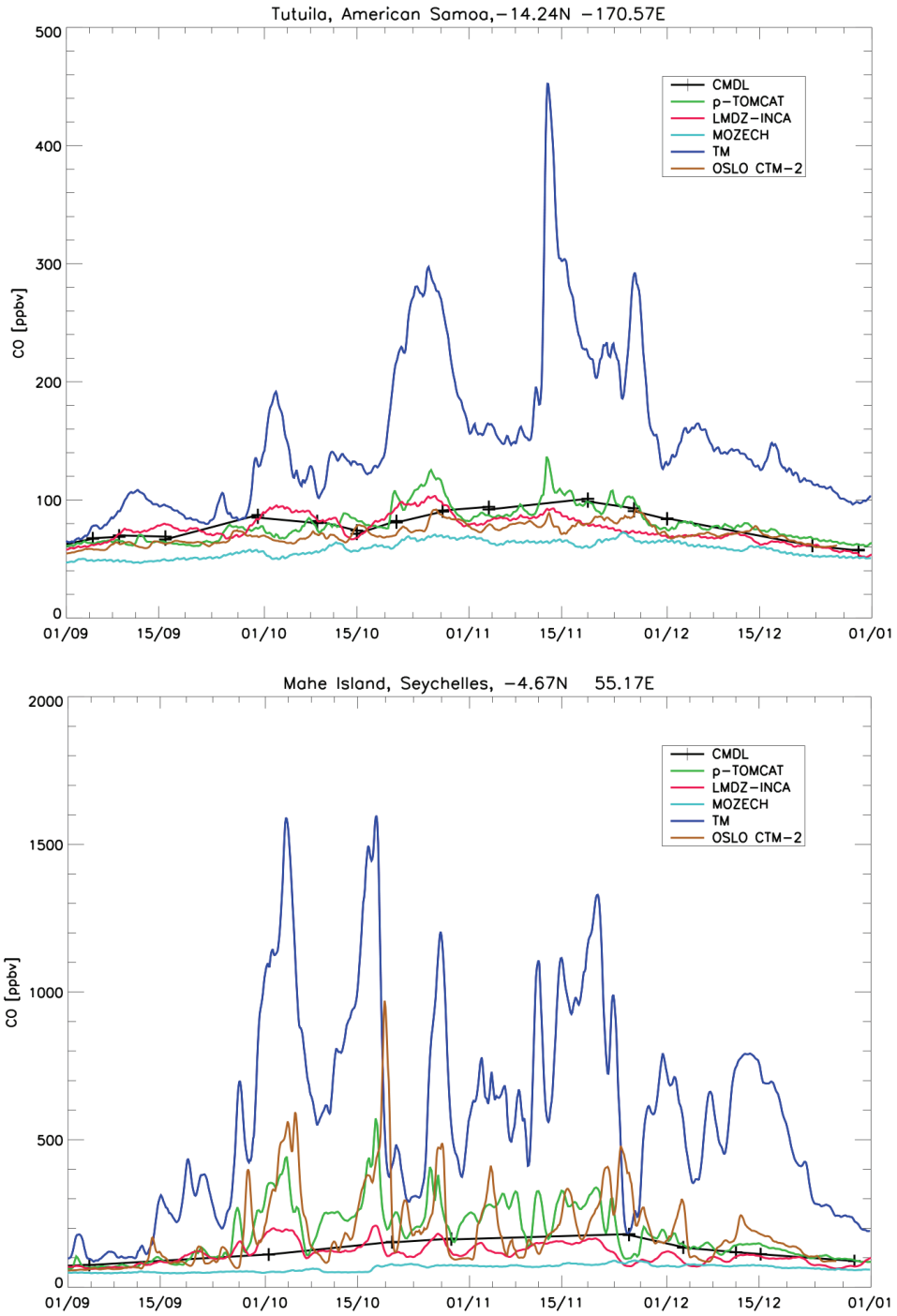


Figure 5. Observations and modelled surface CO at American Samoa and Seychelles. Note that MOZECH results are based on erroneous fire emissions

The amplitude of CO event in LMDZ agrees well with the CMDL data at both sites; p-TOMCAT and Oslo-CTM2 both represent the increase in CO at Samoa well but are somewhat too high in the Seychelles during the first half of the episode. MOZECH is too low for both, with hardly any increase in CO seen for in the Seychelles in this time although there is a clear increase in the concentrations at Samoa and given that the concentrations before the start of the period are lower than the observations the

amplitude of the increase is similar to that of the observations which suggests that the impact of the fires at Samoa is modelled reasonably well and it is the background CO which is too low at this site in MOZECH. In contrast TM is too high for both episodes. The high CO values in TM are mainly caused by the applied emission height distribution for biomass burning. This is demonstrated in Figure 6, which shows the corresponding surface CO fields calculated by TM in case all fire emissions are released at the surface. Figure 7 shows the corresponding time series for American Samoa and Seychelles. With surface emissions the agreement between the TM model and the observations is much improved.

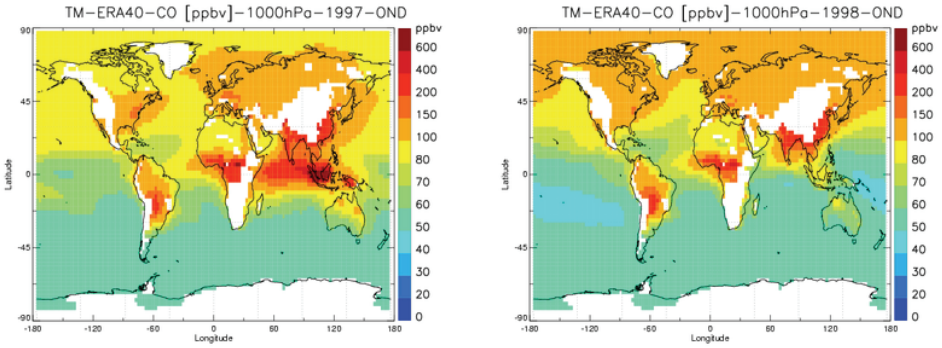
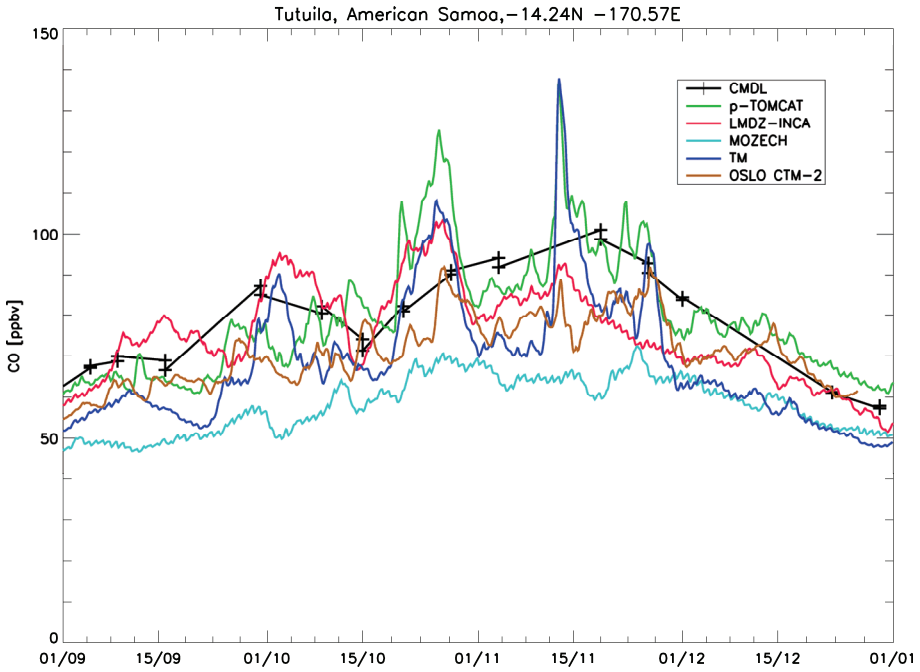


Figure 6. Surface CO for October–December 1997 and 1998 calculated by TM in case the wildfire emissions are emitted at the surface.



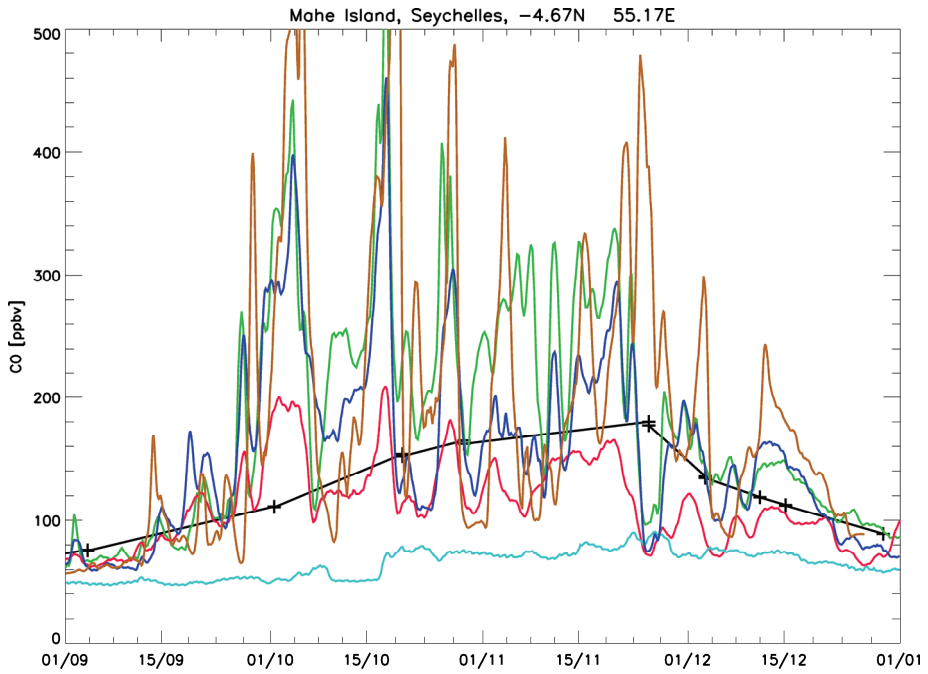


Figure 7. Observations and modelled surface CO at American Samoa and Seychelles using TM results from a simulation with wildfires emissions released at the surface. Note that MOZECH results are based on erroneous fire emissions

Given that the model results bracket the observations with one model consistently lower and four close to the observations the CO emissions in the RETRO inventory for this event appear to be reasonable.

5. Conclusions

We have used three very different data sets to evaluate the models' performance on shorter timescales than for D3-2. Three different species and process have been examined giving a challenging set of conditions for the models to reproduce.

A stratospheric intrusion at Mauna Loa is seen in the models but due to limitations of model resolution the timing and intensity compares badly to observations

For the plume from the Highveld region which was observed in the GOME NO_2 columns, there is some evidence of the models capturing the transport correctly but all lose the NO_2 too rapidly. The best correspondence here is for the MOECH model but even this model seems to have NO_2 columns which are lower in the middle of the Indian Ocean than the GOME retrieval.

For the biomass burning event in Indonesia in September 1997, the emission in the RETRO database clearly capture the large increase compared to other years and they seem to be of reasonable magnitude as demonstrated by the comparison of the model results with CMDL station data downwind of the emission regions. As demonstrated involuntarily by the MOZECH model, the correct representation of fire emissions is critical for simulating this episode correctly.

6. References

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