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Interannual variability of tropospheric composition: the influence of changes in emissions, meteorology and clouds

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Abstract

We have run a chemistry transport model (CTM) to systematically examine the drivers of interannual variability of tropospheric composition. On a global scale, changing meteorology (winds, temperatures, humidity and clouds) is found to be the most important factor driving interannual variability of NO_2 and ozone on the timescales considered. The strong influence of emissions is largely confined to areas where intense biomass burning events occur. For CO, interannual variability is almost solely driven by emission changes, while for OH meteorology dominates, with the radiative influence of clouds being a very strong contributor. Through a simple attribution analysis we conclude that changing cloudiness drives 25% of the interannual variability of OH over Europe by affecting shortwave radiation. Over Indonesia this figure is as high as 71%. Changes in cloudiness contribute a small but non-negligible amount (up to 6%) to the interannual variability of ozone over Europe and Indonesia. This suggests that future assessments of trends in tropospheric oxidizing capacity should account for interannual variability in cloudiness, a factor neglected in many previous studies. The approach followed in the current study can help explain observed tropospheric variability, such as the increases in ozone concentrations over Europe in 1998.

1 Introduction

Tropospheric trace gases play an important role in the Earth system, influencing climate (CH_4, ozone) and determining air quality (ozone, NO_x, volatile organic compounds). Their concentrations vary on a vast range of timescales, and characterising this variability is a major challenge. In particular, understanding what drives the interannual variability (IAV) of annual and seasonal mean concentrations of oxidants is important for explaining the observed trends of tropospheric composition.

Recent model studies have examined the drivers of interannual variability of tropospheric trace gases such as OH (e.g. Dentener et al., 2003; Dalsoren and Isaksen,

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2006), CO (e.g. Szopa et al., 2007) and NO_2 (e.g. Uno et al., 2007; Savage et al., 2008), reaching to a variety of useful conclusions. However, a systematic study a) involving more than one interdependent species, b) focusing on global as well as regional scales, and c) investigating the role of clouds separately in addition to meteorology and emissions, has not been presented so far. Especially the radiative effect of clouds on chemistry (through photolysis) has commonly been neglected as a driver of either IAV or trends in previous studies.

One factor that can affect tropospheric composition significantly is changing emissions from anthropogenic sources. These have changed significantly during recent times in many parts of the globe: in East Asia, rapid industrial development has caused a dramatic increase of pollution (Richter et al., 2005), while in Western Europe, industrial/transport emissions have slowly decreased during the 1990s due to control strategies (Fowler et al., 2001). Changes in biomass burning emissions (NO_x, CO, aerosols) from year to year can also affect tropospheric composition. An example is the fluctuations in wildfire-driven emissions over the Maritime Continent, depending on whether a year is characterized by El Niño or La Niña conditions (Hauglustaine et al., 1999).

Meteorology is also known to be very important in driving tropospheric composition IAV, but the extent of this influence is debated. Extraordinary meteorological conditions, like strong El Niño events (e.g. in 1997–1998) can lead to large year-to-year changes, with O_3 increases over the Western Pacific and decreases over its eastern parts (Chandra et al., 1998; Sudo and Takahashi, 2001; Doherty et al., 2006). There is evidence that El Niño also has an impact on the amount of stratospheric ozone entering the troposphere through stratosphere-troposphere exchange (STE) (Zeng and Pyle, 2005).

Apart from the strongly anomalous situations related to El Niño, meteorology can be important in modifying the distribution of pollutants in the troposphere via regular long-range transport events which coincide with weather systems like fronts or strong convection. Transport of pollution from America and Asia can cause high amplitude changes in the concentrations of ozone and related species over Europe (Creilson

et al., 2003; Pfister et al., 2004; Derwent et al., 2004; Auvray and Bey, 2005). On the other hand, European pollution can impact either cleaner areas like the Atlantic and Northern Africa (Duncan and Bey, 2004) or North America during anomalous transport events (Li et al., 2002). Transpacific transport can also be very important, especially during El Niño years when Asian emissions can impact clean oceanic regions significantly, or even reach North America depending on the local meteorology (Liu et al., 2005). All the above midlatitude processes show significant interannual variability.

We focus here on the 1996–2000 period to investigate what the most important drivers of IAV are. Similarly short periods have been used to examine trends in recent studies (e.g. Richter et al., 2005). However, the focus of the current study is not specifically to assess the trends during 1996–2000, but to extract useful conclusions on the factors affecting IAV by applying a method based on Savage et al. (2008).

In Sect. 2 we present the basic model features and the experimental set-up, as well as show that the model reasonably captures the regions of significant variability in pollution. Section 3 analyzes the drivers of IAV of global NO_2 and ozone columns, while Sect. 4 quantitatively assesses the influence of emissions, meteorology and clouds on ozone, CO and OH. Finally, the conclusions are included in Sect. 5.

2 Model set-up and validation of its ability to capture interannual variability

The model used for the experiments is the updated version of the p-TOMCAT tropospheric CTM described in Voulgarakis et al. (2009a). A detailed description of the ozone budget as calculated in the model is described in Voulgarakis et al. (2009b). The horizontal resolution is 2.8°×2.8° and there are 31 vertical levels extending from the surface to 10 hPa. Tropospheric chemistry for 63 trace species is simulated with the ASAD chemistry package (Carver et al., 1997), and photolysis rates are calculated using the Fast-JX photolysis scheme (Wild et al., 2000). Six-hourly meteorological data (winds, temperatures, humidities, cloud water contents) from the ECMWF analyses are used to drive the model.

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Annually and monthly-varying emissions for industry, transport, shipping and biomass burning come from the RETRO emissions database (Schultz, 2007). Biogenic emissions are taken from Müller (1992) and Lathière et al. (2006). Lightning emissions of NO_x are based on the parameterization of Price and Rind (1994) as implemented by Stockwell et al. (1999). The average lightning emission for the 1996–2000 period is 3.9 Tg(N)yr^{-1}. We use a fixed global annual 3-D field for methane produced from an earlier long-term integration (global burden: 4760 Tg methane). Year-to-year variations in global annual total emissions for each species are shown in Table 1. For most, the highest global emission rates occurred in 1997–1998. As noted in Sect. 1, these two years were characterized by intense wildfire events, which influenced global tropospheric chemistry for many months.

The analysis is based on the results of three model integrations from 1996 to 2000: a) BASE, in which all variables vary from year to year, b) EmFix, in which the surface emissions of all species vary seasonally but not interannually (fixed at 1996 values) and c) MetFix, in which the 1996 meteorology (temperatures, winds, humidity and clouds) is repeated for each year (still varying 6-hourly). The model was run from June 1995 to December 1995 for spin-up. In Table 2 we summarise these runs. CldFix, a run like BASE but with clouds only fixed at 1996 values, is analyzed in Sect. 4.

A detailed evaluation, presented in Voulgarakis et al. (2009a), showed that the model is capable of capturing ozone and CO seasonal cycles well, and that the concentrations of these tracers at various sites compare well with observations. Notable underestimates were found in the Northern Hemisphere for surface CO and overestimates in the upper troposphere for ozone. These features are relatively common in present-day global models (Shindell et al., 2006; Stevenson et al., 2006).

In Fig. 1 we present a comparison of global modelled NO_2 to observations from the GOME satellite instrument for 1996–2000. We compare 5-year mean columns (a, b) and standard deviations (c, d) to provide an assessment of how well the model captures the IAV of global pollution. Sampling of model output data (at 10:45 LT, the GOME overpass time) and post-processing of satellite retrievals is performed as described in

Savage et al. (2008), and the annual averaging helps to remove random errors in the measurements.

The model generally captures the distribution of NO_2 tropospheric column maxima around the globe quite well, and this is also true on a seasonal basis (not shown). The model underestimates total NO_2 in the Northern Hemisphere, especially over industrialized regions (Northeast America, Western Europe, East Asia), suggesting that the NO_2 lifetime may be too short in the model. However, there is an overestimation of the maxima related to biomass burning in the tropics (Indonesia, Central Africa). Over South America and Central/Southern Africa the observed columns are more spatially widespread than those in the model runs, where maxima occur closer to source regions. This negative model bias over industrialized regions and positive bias over biomass burning regions are also evident for most of the models involved in the intercomparison reported by van Noije et al. (2006). They did not reach to a clear conclusion as to what causes these discrepancies.

The absolute standard deviations of the NO_2 column (Fig. 1c and d) show that regions of strong IAV are captured reasonably well, but the amplitude of year-to-year variations is underestimated by the model over northern hemispheric industrialized regions. For the areas where strong biomass burning events occur IAV is overestimated. These differences between measured and modelled standard deviations could be related to uncertainties in the column retrievals or to uncertainties in the modelled IAV. Examining the standard deviation normalized to the 5-year mean (not shown), we find that underestimated variability over industrialized regions shown in Fig. 1 is not caused by the underestimation of NO_2 columns. Possible reasons for these discrepancies include a) lack of IAV in aerosol concentrations driving NO_2 loss in the model and b) no representation of year-to-year variations in methane and stratospheric ozone in the runs analyzed for the current study.

We conclude that many of the general features of pollution variability are captured well by the CTM, and we next analyze the results of sensitivity experiments examining the drivers of IAV on global and regional scales.

3 Emissions and meteorology: their influence on NO_2 and ozone interannual variability

3.1 Tropospheric NO_2 column interannual variability

Figure 2 shows the 1996–2000 differences from the 5-year mean of tropospheric NO_2 columns for all individual years. The BASE run describes the overall IAV. In the idealized case that these differences were entirely driven by emission (meteorology) changes, the values on the plots for the EmFix (MetFix) run would all be zero, while the ones for the MetFix (EmFix) run would look identical to the ones for BASE. We focus on percentage differences, as they better highlight the effect of emissions and meteorology both over polluted and un-polluted regions. Recall that in Fig. 1 we had shown the average NO_2 columns, and its standard deviations, in concentration units.

It is clear from Fig. 2 that the BASE and EmFix differences look very similar for corresponding years, while the ones for MetFix differ significantly, with deviations from the 5-year mean being generally closer to 0%. This indicates that IAV in meteorology is far more important as a driver for changes in NO_2 abundances than emissions over most of the globe on these timescales. The only regions where IAV is captured with fixed meteorology are associated with important biomass burning events. The 1997 and 1998 events in Indonesia (Hauglustaine et al., 1999) and Siberia/Canada (Spichtinger et al., 2004) correspondingly cause large differences from the 5-year mean (up to +240% in Indonesia and +100% in Siberia) which dominate the variability over these regions. To a lesser extent, fires over Central Africa/Amazonia (mainly 1998) and the Iberian peninsula (2000) appear to drive much of NO_2 IAV over these areas. As discussed in Sect. 2, the effect of fires over tropical regions may be overestimated in the model, but the dominant role of emissions in controlling the variability is expected to be represented well.

Changes in meteorology drive the IAV of NO_2 in a number of regions around the globe. Without large changes in meteorology in the tropics during 1997, the NO_2 pollution caused by the wildfires would have been mainly confined to the Indonesian region

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(MetFix plot for 1997). When these emissions are not taken into account, a reduction in the NO_2 maximum over Borneo and Sumatra is seen. Apart from the biomass burning effect, the decrease is also enhanced by the reduction in lightning NO_x due to suppressed convection. But there are still large increases in the columns in the surrounding areas (Indian Ocean, Central West Pacific) captured mainly by meteorological IAV. The export of pollution from Indonesia during late 1997 occurred primarily towards these regions (Duncan et al., 2003). NO_2 has a short lifetime, so not much of it is transported far, but peroxyacetyl nitrate (PAN), a major reservoir of NO_x, can be transported and release NO_2 at a large distance from the source region.

Figure 3 shows PAN tropospheric column differences from the 5-year mean for 1997 and 1996. From the BASE run, it is clear that PAN increases in 1997 over the Maritime Continent when compared to 1996. When emissions are fixed at 1996 values (EmFix) (1996 was not an anomalous year for biomass burning in Indonesia) PAN still increases over surrounding regions. The anomalous circulation patterns caused by El Niño (mainly the strong surface-level divergence centred around Indonesia) increases the export of pollution significantly and explains the large increases of PAN, and subsequently NO_2, over the Indian Ocean and the Central Western Pacific.

El Niño-associated dryness, another meteorological factor (Chandra et al., 1998), could also have played a role in increasing NO_2 columns, as a reduced abundance of OH would increase the lifetime of NO_x. However, as shown later, 1997 was a year with high OH levels over Indonesia due to reduced cloud cover, so dryness cannot have been the main driving factor. Decreased wet deposition of HNO_3, a loss process for NO_x, will also play some role in boosting the positive signals over the Indian Ocean and Western Pacific.

The opposite effect during 1997 is seen for the NO_2 column over the Central/Eastern Pacific and is almost entirely driven by meteorology. This is consistent with Sudo and Takahashi (2001). Higher humidity and HNO_3 wet deposition are expected to be the main reasons for this feature.

Other cases where meteorological processes appear to have driven significant NO_2

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column deviations from the 5-year mean include: a) large increases in NO_2 columns over the British Isles in 1996, as also seen by Savage et al. (2008), b) low NO_2 columns off the east coast of North America in 1996, and c) increased NO_2 columns off the west coast of Central Africa in 1999–2000.

In all these cases, the deviations from the 5-year mean seen in the EmFix case are associated only with changes in meteorology. They can be related to increased lightning activity (especially in the tropics), humidity, cloud optical depths or anomalous transport processes in the corresponding years. The influence of transport can be isolated by looking at the differences from the 5-year mean of the radon tracer, based on the method described by Jacob et al. (1997). Radon has a short lifetime of around 5.5 days, is emitted predominantly over land and its tropospheric distribution depends strongly on horizontal and vertical transport. Figure 4 shows the difference from the 5-year mean of its abundances over the British Isles and Northwest Atlantic (1996) and over West Central Africa (1999). Both for the midlatitude and the tropical areas it is clear that the minimum and maximum radon deviations are strongly correlated with the features found for NO_2 over the same areas. Transport of NO_2 itself or of reservoir species (PAN, HNO_3) away from sources is the most important factor driving these features. The deviations over the British Isles and off the east coast of North America in 1996 are associated with the negative phase of the NAO leading to a) less storm/frontal activity and thus less smearing of NO_2 features and b) less effective long-range transport of plumes (Creilson et al., 2003) across the Atlantic.

Note that the changes in meteorology are almost entirely responsible for year-to-year NO_2 variability over the oceans, while over land the emission changes also appear to play some role (though in general smaller than that of meteorological changes). The fact that the oceans are areas with low NO_2 abundance (see Fig. 1) does not diminish the importance of this conclusion, since in NO_x-limited environments even small changes in NO_2 concentrations can have a significant effect on other tracer budgets (e.g. for ozone). IAV driven by shipping emissions is not detectable, but it is likely that any related variability is masked by the effect of biomass burning plumes transported

over the oceans.

3.2 Tropospheric ozone column interannual variability

Figure 5 shows the differences between each year's tropospheric ozone columns and the 5-year mean. The maximum differences for ozone are smaller than for NO_2 (maximum +36% over Indonesia in 1997 and around +20% over northern high latitudes in 1998). However, the various meteorological and emission effects are still important and dominate large parts of the globe due to the longer lifetime of ozone compared to NO_2. A common feature of Fig. 2 and Fig. 5 is that the BASE and EmFix plots again look similar, implying that changing meteorology was the most important driver of tropospheric ozone IAV during this 5-year period.

The ozone increases in the tropics in 1997 are driven both by the intense biomass burning events over Indonesia (increase of precursors) and by the favourable meteorological situation related to El Niño: suppressed convection, downward motion and dryness favour both the downward transport of ozone-rich upper tropospheric air throughout the troposphere and decreased ozone loss by the O^1D+H_2O reaction, over a large area centred around Indonesia. The effects on NO_x mentioned in the previous section are also important, since NO and NO_2 are major ozone precursors. The ozone pollution resulting from the wildfires was transported to large distances from the sources. The only tropical area where ozone decreased in 1997 is the Central/Eastern Pacific where conditions are opposite to those found over the Western Pacific. When fixing meteorology to 1996 values (MetFix), this feature disappears completely and increases are found in almost all tropical areas, excluding Central Africa, possibly due to lower than average biomass burning emissions in 1997. However, the positive differences with MetFix are smaller in magnitude than with the other two runs; the only region where they are larger than +5% is over and around Indonesia. Thus, while the IAV in emissions alone would cause ozone increases in tropical areas, it is the meteorology, related to El Niño, which makes this more than a regional feature.

In 1998, large positive differences from the 5-year mean (up to 20%) are seen in all

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the extratropical areas, almost symmetrically centred around the tropics, where negative differences occur. The large boreal fires which occurred during that year and, to a smaller extent, the high fire activity in South America caused large increases in the amount of ozone precursors in the troposphere, partly causing the increases in extratropical ozone in this year. Also, the impact of tropical pollution produced in 1997 is expected to have a signal in the extratropics for several months, especially through the long-range transport of long-lived precursors (e.g., CO, see Duncan et al. (2003)). However, with emissions at 1996 values (EmFix), all the features of the BASE plot for 1998 are preserved, with just a small reduction in the deviations. In contrast, with fixed meteorology (MetFix), smaller, almost globally distributed deviations from the 5-year mean are found for this year, with maxima of 2–5% located around the main biomass burning regions. Anomalous emissions only cause a small fraction of this year's anomaly seen in the extratropics.

The fact that the large extratropical positive deviations from the 5-year mean in 1998 are ubiquitous could indicate that long-range transport from other mid-latitude anthropogenically polluted areas may not be the cause of the extratropical ozone increases. High horizontal wind speeds, transporting ozone and precursors to unpolluted regions, would also have caused reductions of tracer concentrations over the polluted areas. But large increases are seen in the BASE and EmFix runs even for the southern extratropics where pollution sources are much less than at northern mid/high latitudes, suggesting that long-range transport within the troposphere is not the main reason for ozone increases. Zeng and Pyle (2005) found that 1998 was a year of very high STE in the extratropics associated with high SST anomalies in the Pacific (El Niño) a few months earlier. This transport-related process which is affected directly when fixing meteorology to 1996 is more likely to have caused the large positive signal.

By examining the IAV of tropopause heights around the globe for the 1996–2000 period we note that the main features of Fig. 5 are not driven by changes in the total mass of air included in the calculated tropospheric columns.

We have assumed so far that the main effect on extratropical IAV of fixing meteorol-

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ogy is through changes in horizontal and vertical transport. However, other meteorological variables may have an influence, and the most important of these are humidity and cloud optical depth. Water vapor can increase loss of ozone (via O^1D+H_2O) but can also enhance the production of peroxy radicals which drive ozone production. However, there were no large hemispheric-scale increases in water vapour in 1998 in the model. We therefore examine the role of clouds in determining IAV of ozone in the next section.

4 Quantitative analysis of the role of emissions, meteorology and clouds

Clouds play an important role in altering photochemical processes by modifying solar radiation throughout the tropospheric column. To our knowledge, there have been no previous studies assessing the role of clouds in the IAV of global tropospheric composition. Here we separate the shortwave radiative effect of clouds from other aspects of meteorological variability by conducting an additional model run. In this run, CldFix, 6-hourly varying 3-D cloud optical depths for 1996 are used throughout the 1996–2000 period; everything else varies as in BASE. Note that "clouds" are a subset of "meteorology" in the current approach, meaning that in the case that meteorology is fixed to 1996 values (MetFix), clouds are kept fixed as well. Deviations of the tracer abundances from the 5-year mean are compared to those seen when fixing the meteorology or the emissions to 1996 conditions.

4.1 Analysis of global ozone IAV drivers

Figure 6 shows the difference in each year's ozone burden from the 5-year mean. It also shows the percentage of variability which is explained by each one of the factors separately, calculated using a simplified attribution approach based on Szopa et al. (2007). The 5-year anomalies for each run were averaged and then divided by the average anomaly for the BASE run in order to determine by how much the variability is

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reduced when each field is fixed at 1996 values. By subtracting this percentage from 100, the variability explained by each individual factor can be quantified. The equation used is the following:

$$P = \left[1 - \frac{\frac{1}{N}\sum_{i=0}^{N} \left| X(i)_S - \overline{X(i)_S} \right|}{\frac{1}{N}\sum_{i=0}^{N} \left| X(i)_B - \overline{X(i)_B} \right|} \right] \times 100 \quad (1)$$

where P is the percentage shown on the plots, $X(i)_S$ represents the anomalies from the 5-year mean for each year in the sensitivity runs (EmFix, MetFix, CldFix), and $X(i)_B$ is the same variable but for the BASE run. N is the number of years considered (5 in this study).

Figure 6 shows a strong peak in the global ozone burden (+7% in 1998) associated with the 1997–1998 El Niño event. Table 3 shows the global annual budget terms for 1996–2000 as calculated in the BASE run. It is clear that the increased global tropospheric ozone abundances found in the 1997–1999 period are strongly related to increases in STE. Net chemical production is lower than average during these years and the deposition rate is larger, so changes in these terms are unlikely to have driven the ozone burden increases we find here.

Changes in meteorology have a stronger impact on global ozone than changes in emissions. The variability in the MetFix run is very small, and shows only a small increase in 1998 reflecting the intense wildfires. Meteorology drives almost 80% percent of the IAV of ozone on a global scale.

Clouds exert a smaller but non-negligible influence in the IAV of the ozone burdens. Globally they are responsible for 6% of the variability, which is around 8% of the total influence of changes in meteorology.

An examination of the importance of clouds and the rest of the factors in regional scales is presented next.

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4.2 Regional scale analysis: Europe and Indonesia

We focus here on Europe and Indonesia as important examples of the northern extratropics and the tropics in order to examine how similar the responses of ozone, CO and OH are in these regions to year-to-year changes in emissions, meteorology and clouds.

4.2.1 Tropospheric ozone IAV

Figure 7 shows that there is a maximum in O_3 burden over Europe in 1998 followed by a relative decline in 1999 and, especially, 2000. Meteorology is responsible for 86% of this variability while emissions changes are less important (5% compared to 8% generally in the northern extratropics). Cloud changes drive 4% of the European O_3 IAV. Over Indonesia, emissions drive a significant part of O_3 IAV (32%).

Table 4 shows the regional ozone budget terms for the BASE run over Europe and Indonesia. Europe is a net exporter of ozone for all the years of study (−8 to −18 Tg/yr), and and we find a decrease in the net export of ozone from Europe in 1998–1999 (around 30% less than average) which may reflect increased import or decreased export. Net chemistry (production minus loss) was lower than average during the same years and deposition was higher due to higher ozone concentrations, so the ozone peak is attributed to the decreased net export term. Decreased net export may be a result of both/either decreased net chemistry and/or the dominance of transport regimes which do not favor export. Similarly, the ozone burden decrease over Europe in 2000 (La Niña period since 1999) can be attributed to changes in net transport of ozone from the European region.

With the approach taken in this study, we are able to attribute the features of the tropospheric ozone distribution to a variety of influences. Confidence in the approach is given by the fact that the BASE model is able to capture major features of the observed ozone changes. In Fig. 8 we show a 12-month running mean of ozone monthly differences from the 5-year mean for the Zugspitze research station (47° N, 11° E), as

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revealed by measurements and as calculated by the model. Zugspitze is considered to be representative of Central Europe for background atmospheric studies (e.g. Kaiser et al., 2007). Clearly the model captures the important ozone increases from late 1997 to mid 1999, and the much lower than average values in 2000.

Over Indonesia, also a net exporter of ozone, the 1997 ozone burden maximum is caused by increased net chemistry but also by slightly lower than average net export from the region. This is most likely caused by subsidence of stratospheric ozone-rich air causing increased import of ozone. Horizontal transport to Indonesia within the troposphere was lower than average in 1997 due to strong low-level divergence. In 2000, the minimum ozone burden relates to the transport term being around 20% lower than average, meaning higher than average net export.

4.2.2 Tropospheric CO and OH IAV

Figure 9 shows the CO and OH results of the four runs (BASE, EmFix, MetFix and CldFix) for Europe and Indonesia. For CO, clouds make a minor contribution, especially over Indonesia where only 1% of the IAV is explained by changes in cloudiness. Changes in meteorology are far less important than they are for ozone. Year-to-year variations in emissions explain 92% of the IAV of CO over Indonesia and even over Europe, where biomass burning is not nearly so intense, this figure is as high as 82%. These results contrast with those of Szopa et al. (2007), who found that in the tropics meteorology is the main reason driving CO IAV, while in the extratropics changes in emissions and meteorology are equally important. Differences in chemical schemes, such as in the secondary production of CO from hydrocarbon oxidation, may partly explain these different results. As noted in Sect. 2, p-TOMCAT overestimates pollution over Indonesia during the period of this study, suggesting that the effects on tropical IAV may be exaggerated. However, we do not expect that the main IAV signal would change with the use of lower emissions.

The IAV of mass-weighted OH concentrations is examined in the bottom panels of Fig. 9. The analysis focuses on the boundary layer, where the impact of clouds is ex-

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pected to be greatest. As shown by Voulgarakis et al. (2009b) and other previous studies, above and below-cloud modification of radiation is more important than in-cloud effects for OH. For Europe, we present the results for summer (June-July-August), as this is also the season with the greatest solar radiation, when the impact of clouds on OH concentrations is highest. The figures show that when meteorology does not vary, almost none of the OH IAV is reproduced. This is consistent with Dentener et al. (2003), who found that IAV in OH is driven more strongly by meteorology than by surface emissions changes. It is clear that clouds are a very important component of this OH variability in the lower troposphere. 71% of the year-to-year variations in OH over Indonesia are caused by changes in cloudiness. Over Europe the figure is smaller (25%) but still important. Thus, radiative cloud effects should not be ignored when studying the IAV of the tropospheric oxidizing capacity, especially since OH changes affect methane, a gas of major climate importance.

An OH minimum in 1998 (−4%) over Europe can be partly explained by changes in cloudiness: it is less pronounced when fixing clouds and cloud optical depths are indeed lower than average in 1998 (not shown). The decrease of OH in 1998 could have been one of the causes of decreased net ozone chemistry over Europe in the same year shown in Table 4. By examining production and loss separately, we find increased ozone destruction in 1998 driven by higher ozone abundances, but also decreased ozone production, possibly due to lower OH and thus reduced peroxy radical concentrations. The effect of this is also boosted by the lower NO_2 abundances in 1998 over much of Europe in the model (see Fig. 2).

Over Indonesia, cloud optical depths are mostly lower than average in 1997 (not shown), when the maximum OH concentrations are seen. This is related to the El Niño anomaly with less deep convection over a normally highly convective region. Although the area was dry during 1997, not favouring OH production, there was clearly a stronger influence by the higher photolysis rates due to fewer clouds. Higher ozone values (see Fig. 5) also contribute to the OH increase.

5 Conclusions

We have presented an assessment of how meteorology, emissions and clouds drive the interannual variability of important tropospheric tracers based on CTM calculations for the period 1996–2000. For NO_2 and ozone, meteorology is the most important factor driving this variability for this period. On a global scale, around 80% of the ozone variability can be explained by changes in meteorological conditions (winds, humidity, clouds, temperatures). A strong contribution from emissions variability is confined to areas where intense biomass burning occurs (e.g., Indonesia, Siberia). In contrast, emissions variability makes the largest contributions for CO, both in the tropics and in the extratropics. For OH, interannual variability is strongly driven by changes in meteorology and a particularly important component of this influence is the radiative effect of the variability in cloudiness.

A regional analysis reveals that the impact of meteorological variations on the observed interannual variability in ozone is stronger than that of emission variations in both extratropical and tropical regions (Europe and Indonesia), but is more dominant in the extratropics (86% over Europe). Clouds make a small but non-negligible contribution to the interannual variability of ozone. The interannual variability of CO shows no significant sensitivity to changes in clouds or meteorology over Indonesia. Over Europe, meteorology has a slightly greater effect, but remains a less important factor compared to emissions. However, the short lifetime of OH makes it susceptible to changes in clouds and to meteorology as a whole. Over Europe, cloud variability drives 25% of the interannual variability of OH, while over Indonesia this figure is as high as 71%. This suggests that future assessments of trends in tropospheric oxidizing capacity need to account for the interannual variation in cloudiness.

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Table 1. Global annual total emissions for all species emitted in p-TOMCAT (in Tg yr^{-1}). Lightning and aircraft NO_x emissions are not included here.

Species /Year	1996	1997	1998	1999	2000
NO_x	136.2	139.9	140.4	134.8	137.9
CO	1032.5	1221.9	1168.2	998.8	1013.2
Ethane	9.5	11.9	10.7	9.4	9.6
Propane and higher alkanes	36.1	37.6	35.4	34.9	35.1
Isoprene	522.2	522.2	522.2	522.2	522.2
Formaldehyde	30.9	32.9	32.8	30.5	30.8
Acetaldehyde	32.3	33.7	33.4	32.2	32.3
Acetone	76.9	78.3	77.9	76.7	76.8

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Table 2. Runs conducted for this study and how they differ. Interannually-varying fields are denoted with "1996–2000" and fields without interannual variation are labelled "1996". Note that "Clouds" are a subset of "Meteorology" in the current approach, meaning that in the case that meteorology is fixed to 1996 values (MetFix), clouds are kept fixed as well.

	Emissions	Meteorology	Clouds
BASE	1996–2000	1996–2000	1996–2000
EmFix	1996	1996–2000	1996–2000
MetFix	1996–2000	1996	1996
CldFix	1996–2000	1996–2000 (except clouds)	1996

Table 3. Global annual ozone budget terms for all the years of study (BASE run). The transport term represents the STE for this global case.

	1996	1997	1998	1999	2000
Net Chemistry (Tg/year)	658	581	380	466	662
Transport (Tg/year)	674	789	1012	859	627
Deposition (Tg/year)	−1348	−1374	−1410	−1364	−1325

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Table 4. Regional annual ozone budget terms for the BASE run over Europe and Indonesia. The transport term relates both to STE and to transport processes within the troposphere to and from the regions.

	1996	1997	1998	1999	2000
EUROPE					
Net Chemistry (Tg/year)	76	74	66	67	75
Transport (Tg/year)	−13	−15	−9	−8	−18
Deposition (Tg/year)	−63	−63	−65	−65	−63
INDONESIA					
Net Chemistry (Tg/year)	56	77	68	63	73
Transport (Tg/year)	−29	−32	−45	−32	−42
Deposition (Tg/year)	−37	−47	−37	−35	−35

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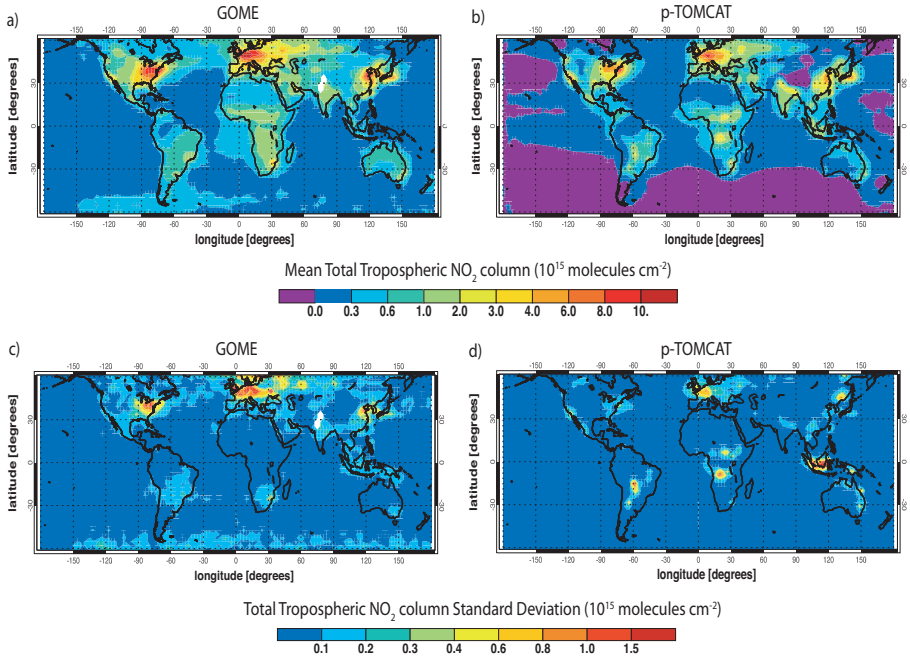

Fig. 1. Comparison of **(a, b)** annual 5-year mean p-TOMCAT tropospheric NO_2 columns and **(c, d)** their standard deviation, with GOME observations. The white area over North India denotes total lack of observations during the 1996–2000 period. The purple (negative) areas relate to an artifact caused by the method used for the total column calculation (clean air subtraction – see Savage et al. (2004) for further details). They appear only over remote areas where NO_2 concentrations are low.

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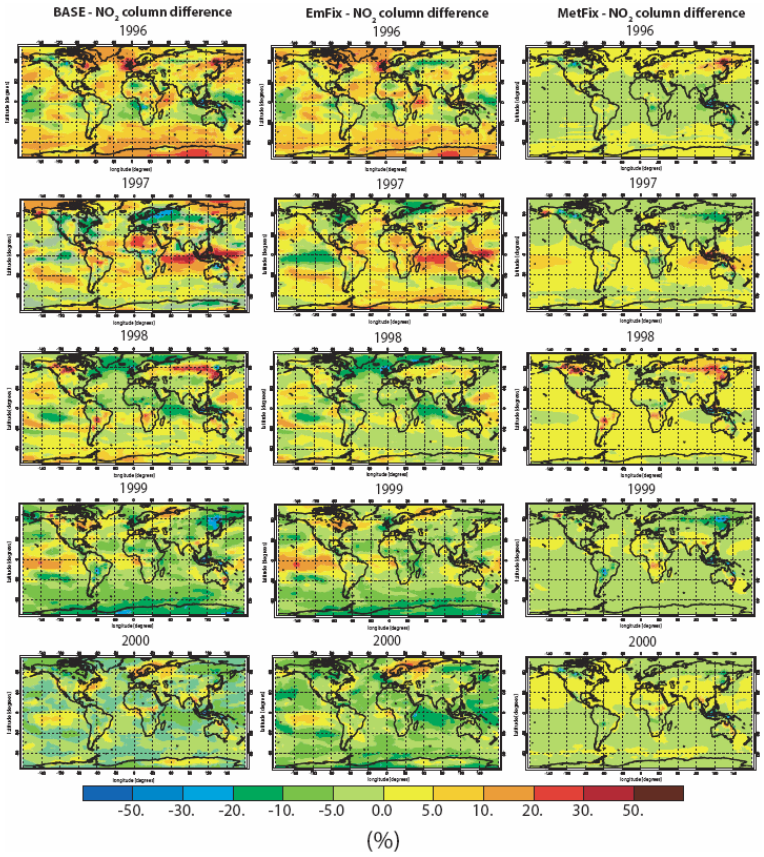

Fig. 2. Percentage differences between tropospheric NO_2 columns for each year and the 5-year (1996–2000) mean. Each column corresponds to one sensitivity run (BASE, EmFix, MetFix). The tropopause was assumed to follow the 380 K isentropic surface in the tropics and the 3.5 PVU surface in the extratropics.

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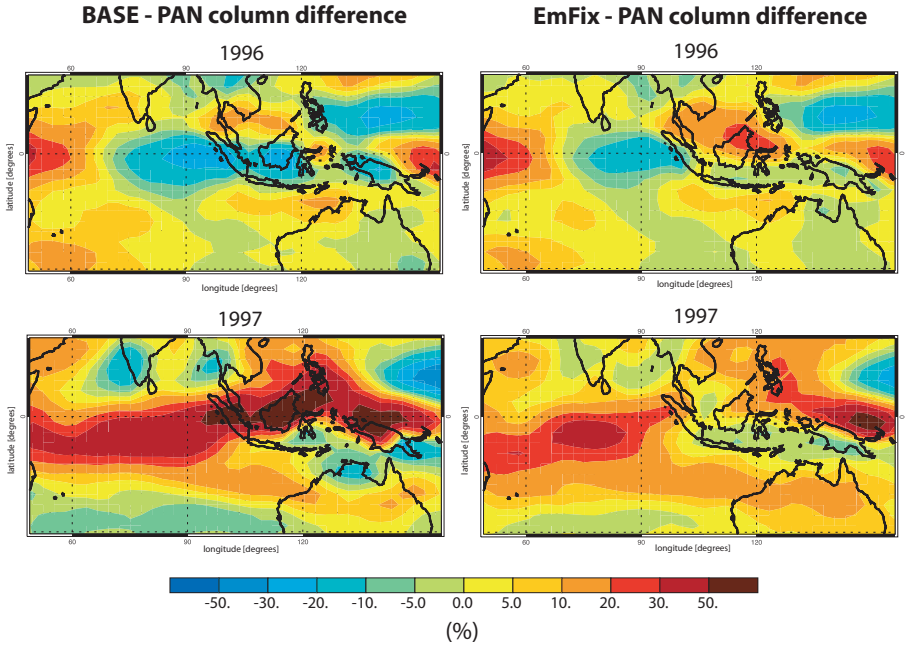

Fig. 3. 1996 and 1997 percentage differences of PAN tropospheric columns from the 5-year mean for BASE and EmFix. The tropopause is the same as for Fig. 2.

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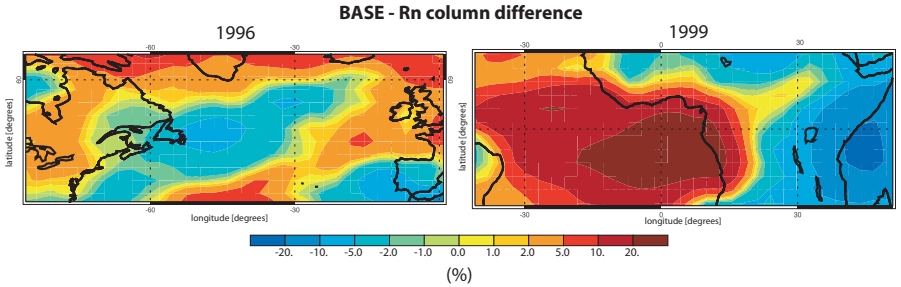

Fig. 4. 1996 and 1999 percentage differences of radon tropospheric columns from the 5-year mean for three areas with high amplitude NO_2 deviations from the 5-year mean driven by meteorology: the British Isles (1996), Northwest Atlantic (1996) and West Central Africa (1999).

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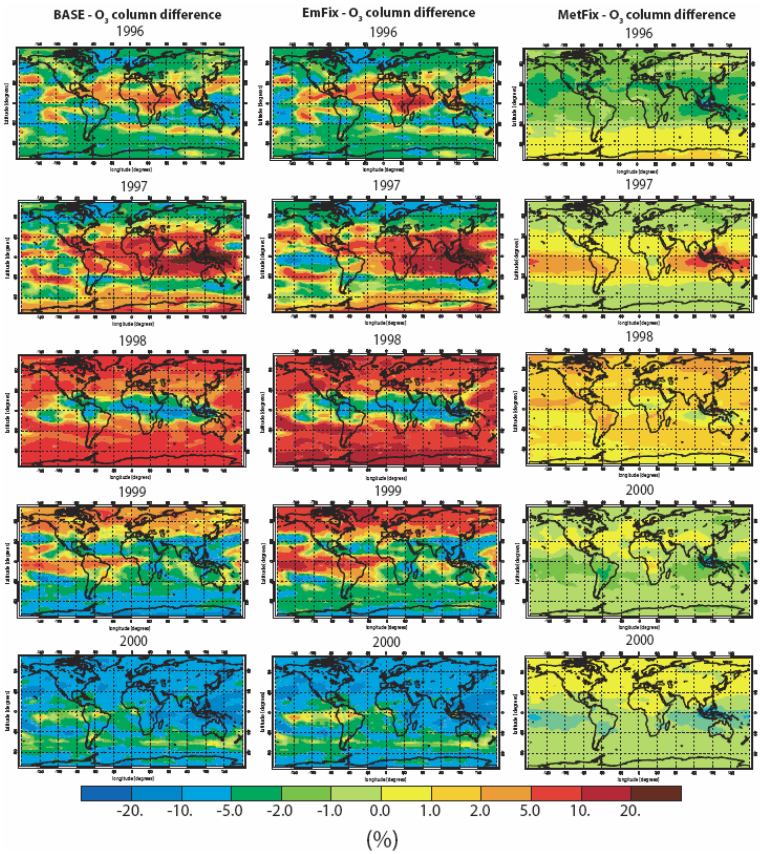

Fig. 5. Percentage differences between tropospheric ozone columns for each year and the 5-year (1996–2000) mean. Each column corresponds to one sensitivity run (BASE, EmFix, MetFix). The tropopause was assumed to follow the 380 K isentropic surface in the tropics and the 3.5 PVU surface in the extratropics.

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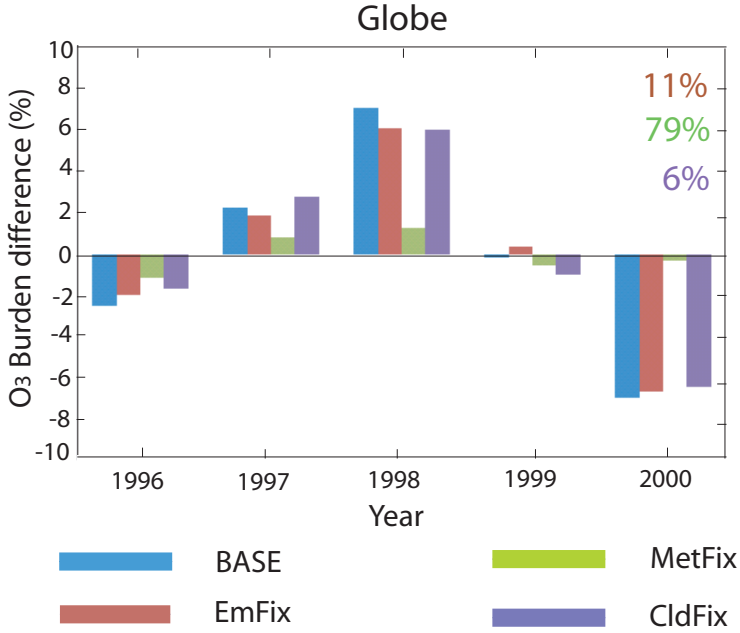

Fig. 6. Percentage global annual ozone burden differences from the 5-year mean as calculated from the four sensitivity runs: BASE (blue), EmFix (red), MetFix (green) and CldFix (purple). The numbers at the upper right parts of the plots represent the amount of variability explained by each of the individual drivers: changing emissions (red), changing meteorology (green) and changing cloudiness (purple).

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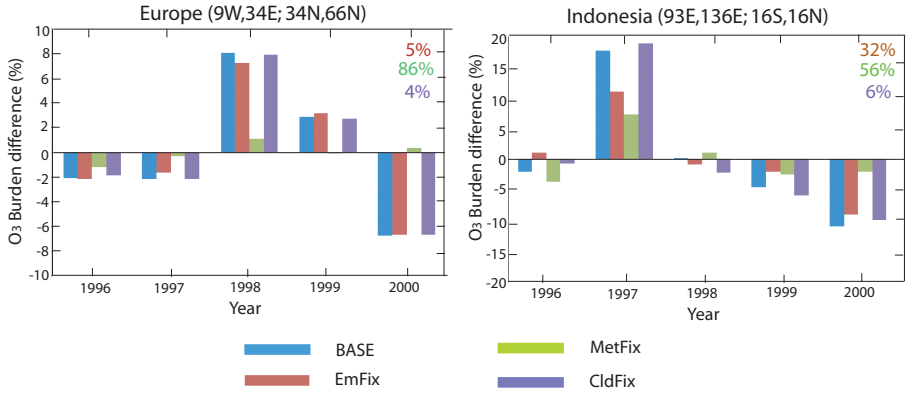

Fig. 7. Same as Fig. 6 but for the European and Indonesian boxes. Note the difference in the scale used for Indonesia (−20 to 20%) due to the higher variability over this area.

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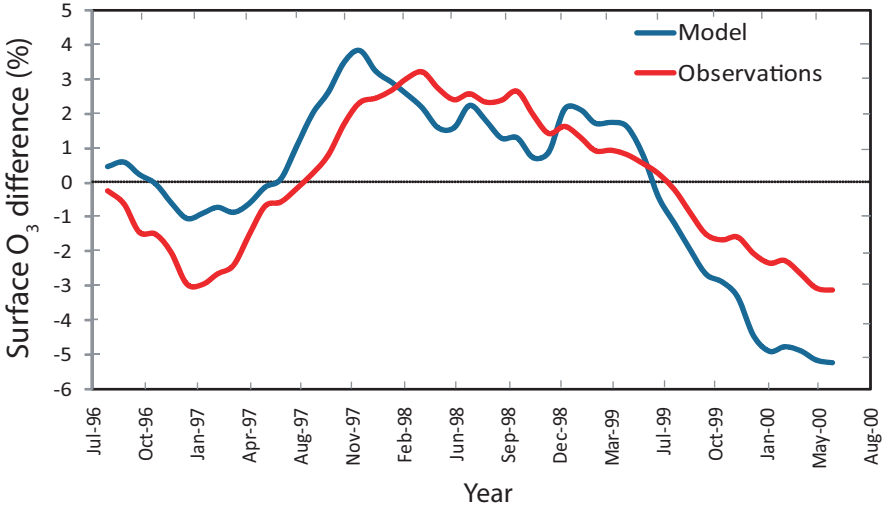

Fig. 8. 12-month running means of ozone monthly differences from the 5-year (1996–2000) mean for the Zugspitze research station (47° N, 11° E), as revealed by measurements (red) and as calculated by the model (blue).

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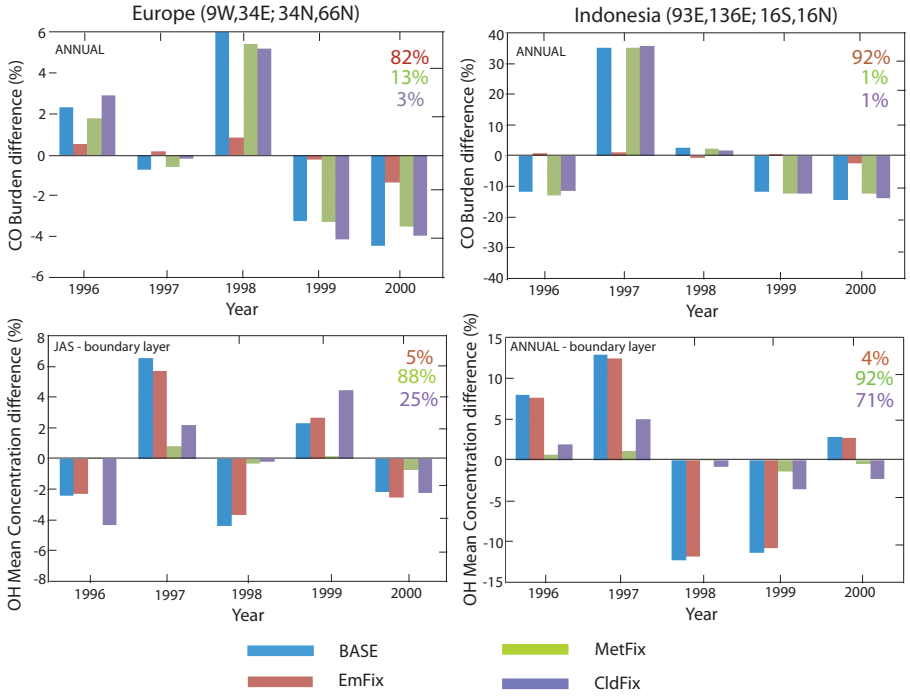

Fig. 9. Same as Fig. 7 but for CO and boundary layer OH. For OH, July-August-September (JAS) means are examined over Europe while annual means are examined for Indonesia.

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