

Interactive comment on “Model analysis of the factors regulating the trends and variability of carbon monoxide between 1988 and 1997” by B. N. Duncan and J. A. Logan

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We appreciate the reviewers comments, which are italicized, and have carefully addressed each of them.

General Comment: This paper examines possible reasons for a decrease in boundary layer carbon monoxide observed during the late 1980s and 1990s. Understanding changes in CO is important due to the reaction of CO and OH, which in the remote atmosphere is the major sink for both species. The few measurements made before the mid-1980s suggested that in the Northern Hemisphere CO was increasing, causing concern that OH would subsequently decrease. Soon after reports of an increase, global measurements indicated a decrease in CO. This was assumed to primarily re-

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flect decreasing emissions in the industrialized north. Measurements in South Africa showed no long-term trend. The reasons for these changes have not been examined quantitatively.

This manuscript provides a detailed investigation of why CO decreased in the Northern Hemisphere during the 1990s. The modeling allows for the separation of changes in sources, chemistry and the effects of transport. While the approach used is not unique the application details described here can provide a model for future investigations of CO over other periods of time.

This is a very good paper and suitable for ACP. The manuscript is well organized and very clearly written.

We appreciate the complement!

Specific Comments: P.3, I.20-28; P.5 Fossil fuel emissions. National emission statistics from China are known to be low. Was this corrected for?

Our original inventory for fossil fuel emissions was developed for 1985, and we then scaled forward in time using fossil fuel statistics, as described in Duncan et al. (2007). We did not have the Chinese provincial energy statistics when we developed the CO inventory in the 1990s. We performed a sensitivity simulation of model CO to a 25 percent increase in fossil fuel emissions, which we discuss in Section 6.2 in Duncan et al. (2007). We found that the higher emissions did indeed improve the model bias at the stations downwind of China in the north Pacific.

The fossil fuel emissions inventory, including uncertainties for Chinese emissions, is discussed in detail in Section 3.1 of Duncan et al. (2007), and is compared to the Streets et al. (2006) inventory.

It would be quite useful to this reader if a table of the annual emissions from all sources was provided in Table 1.

We summarize and discuss the sources of CO in detail in Duncan et al. (2007). To

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address this comment, we changed the 2nd paragraph in Section 2.1 to:

The sources of CO include direct emissions from biomass burning, fossil fuel and bio-fuel combustion, and chemical production from the oxidation of methane and biogenic and anthropogenic non-methane hydrocarbons (NMHC). These sources are presented in detail in Section 3 of Duncan et al. (2007) and are summarized in Table 1 of that paper.

P.5, Fossil Fuel emissions. During 1988-1997 did CO emissions change in other parts of the world? CO₂ emissions from India increased by 75 percent between 1988 and 1997 (CDIAC) and we might expect CO emissions also increased dramatically.

The fossil fuel emissions inventory and trends from 1988 to 1997 are described in detail in Section 3.1 of Duncan et al. (2007). For most countries for which we did not have national inventories, we scaled CO emissions to liquid fuel use from CDIAC; for China we scaled to coal use. Indian CO emissions increased by about the same factor as total CO₂ emissions with our approach, simply because liquid fuel use grew by about the same factor as total fossil fuel use in India from 1988 to 1997. It is important to note that this was not the case in all countries. Since CO emissions are a product of inefficient combustion, and CO₂ a product of total fossil fuel combustion, one cannot assume that CO emissions will necessarily scale with CO₂ emissions, particularly if CO emissions are controlled (e.g., by catalytic convertors on vehicles). We caution in our 2007 paper that our emission estimates for developing countries such as India may be too low, so the absolute increase will be too low.

Figure 2 of Duncan et al. (2007) shows trends in CO emissions for various regions. Indias emissions are split between East Asia and Other totals. Our work implies that East Asian emissions increased from 102 Tg to 155 Tg. Other fossil fuel emissions increased from 50 to 66 Tg CO from 1988-1997. The increase in Other occurred primarily from increases in Latin America (7 Tg) and Africa (6 Tg). While this increase is small relative to changes in Europe and East Asia, it is similar to the decrease in

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North America during the period. We added the following sentence to Section 3.1:

Relatively small increases (16 Tg CO) occurred elsewhere, primarily in Latin America (7 Tg) and Africa (6 Tg).

We also added emissions for the rest of world to Table 1.

P.5-6. Overhead ozone column. The description of the ozone trend calculation is not clear. The text states effects of the solar cycle and QBO are not included, yet it also seems to conclude that much of the trend in the tropics is due to the solar cycle. Please clarify.

The important point is that we are not trying to compute a trend in tropical ozone caused by changes in chlorine and bromine in the stratosphere, which is what most trend studies of column ozone do; thus they remove the effects of known causes of variability, such as the solar cycle and quasi-biennial oscillation, QBO, by including terms for the dependence of ozone on these factors in their regression models. We compute the trend in column ozone, as observed, over our period of interest, 1988-1997, and it then does depend on the solar cycle. If you want to extract the trend in ozone due to chlorine and bromine, you would want to remove the effects of the solar cycle variation, so our way of computing the trend would not be appropriate. As noted in Section 3.2 and Figure 1, we do not remove the effects of the solar cycle and QBO on column ozone, as we are computing the trend in column ozone caused by any and all factors, as this is what impacts tropospheric OH, and subsequently, CO. We have clarified the text as follows in Section 3.2:

We calculate trends in ozone with the multivariate linear regression model of Ziemke et al. (1997), except that we do not include terms to remove the dependence of ozone on the solar cycle and the QBO. Such regression models usually include these terms to remove the effects of natural variability on column ozone, in order to compute trends attributable to changes in chlorine and bromine. Our purpose is different however. We are interested in the trend in ozone for 1988-1997, whatever the cause, as this is what

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impacts tropospheric OH and, subsequently, CO.

P.8. Trends in Model CO. Can model trends be compared to measured trends? Some comment/discussion of this point is needed.

We compared model and observed trends for the GMD and column stations in detail in Tables 11-12 and Section 5 of Duncan et al. (2007). We already say in Section 2.3 of the present manuscript that the model does indeed capture the 10 percent decrease observed during the study period in the North Pacific and the 20 percent decrease observed at high northern latitudes.

P. 12, I.23-29. Episodic Events. The model used in this study did not fully account for changes in OH relating to the Mt. Pinatubo eruption in 1991 because it did not include the scattering effects of volcanic aerosols. The authors state the eruption had little effect on their trend calculations in the HNH. However the impact would have been greater in the mid- and tropical latitudes. This should be presented more quantitatively (for example, how would CO respond if model OH was overestimated by 5-8 percent (Dlugokencky et al., 1996)).

The impact of the stratospheric aerosols on the model trends is complicated by the fact that significant fires occurred in Indonesia in late 1991, shortly after the eruption of Mt. Pinatubo. The high and prolonged CO emissions from the fires decreased tropical OH for more than 3 months. Therefore, the CO and methane anomalies reported in Figure 2 of Dlugokencky et al. are likely a combination of influences of the fires and the stratospheric aerosols after August 1991.

Dlugokencky et al (1996) estimated that a 10 percent decrease in OH over a 6 month period would lead to about a 8 ppbv increase in tropical CO. For perspective, the anomalies due to the Indonesian fires in late 1991 were similar, 5-10 ppbv for 3+ months (Figure 5). We recalculated the trend assuming a 8 ppbv increase in model CO for the last 6 months of 1991. The overall impact was to make the tropical trend less positive. For example, the trend in the southern tropics decreased from 1.13 Tg/y

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(Figure 4) to 0.96 Tg/y from September–November and the trend in the northern tropics from June–August decreased from -0.65 Tg/y to -0.75 Tg/y. So, from this back of the envelope calculation, neglecting stratospheric aerosols in our simulation does have an impact, albeit small, that does not change any of our conclusions in this study.

To address this comment, we changed the last paragraph of Section 5.3 to:

There is not a major response of the model CO burden to the aftermath of the Mt. Pinatubo eruption in June 1991 (Figure 5). Fuglestad et al. (1994) found the response of methane was damped as the largest changes in the column occurred outside of the tropics and in winter (Figure 1). Model calculations in the literature indicate that OH initially decreased during the first year following the eruption because of the scattering effects of sulfate aerosols, which reduced the amount of UV radiation in the troposphere (Dlugokencky et al., 1996), and began to increase in 1992 and 1993 as the ozone column decreased in 1992/1993 (Figure 1; Bekki et al., 1994). We do not include the scattering effects of stratospheric sulfate aerosols on photolysis rates. Dlugokencky et al. (1996) estimated that tropical OH decreased by about 10 percent because of the stratospheric aerosols which would cause CO to increase by about 8 ppbv. This estimated increase would cause a minor change in the trend in tropical CO (Figure 5) in our model, which would not affect any of our conclusions.

P.13. Transport. Does the simulation provide some information about the possible effects of ENSO phases since you have a run without ENSO meteorology and another with it?

To answer this question adequately, would require a careful analysis of the transport variations between ENSO phases and the strength of a given phase, as well as consideration of the changes in biomass burning emissions during El Niño. This is beyond the scope of this manuscript, though it is an interesting question. It is not as simple as comparing a run with interannually varying meteorology and one with the same year of meteorology, repeated for each year.

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P.14: Summary. Although the paper focuses on changes in the High Northern Hemisphere, trends from the three other zonal averages are presented. A concluding statement about these should be included.

We do, in fact, discuss the tropics and southern extra-tropics in the Summary. We say in the 2nd paragraph:

A monotonic increase in methane between 1988 and 1997 contributed positively to CO, but the concurrent decrease in the ozone column and concomitant increase in OH caused a compensating decrease in CO. Consequently, the annual trends in CO in the tropics and southern extra-tropics were small (0.25 Tg/y) or insignificant.

And in the 3rd paragraph, we say:

The Indonesian wildfires in late 1997 released a tremendous amount of CO over about 3 months. Consequently, the trends in tropical CO were high (0.7-1.1 Tg/y) in September-November. The trend in the southern extra-tropics (0.6 Tg/y) was also affected by transport of CO from this event.

We only devote the 1st paragraph to trends in the northern extra-tropics.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9099, 2008.

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