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Quantifying Magnitudes and Distributions of CO Sources

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Abstract

We use a simplified version of the GRANTOUR chemical tracer model to analyze emissions of CO from various sources. The GRANTOUR model has been simplified to include the eigenvalue chemistry solution method of Prather (1994) The analysis includes fossil fuel combustion, biomass burning, hydrocarbon oxidation, ocean, and terrestrial sources of CO. We also present a 1°×1° emissions inventory of CO from fossil fuel combustion.

Introduction

Carbon monoxide (CO) has a variety of important roles in the atmosphere. In the non-urban troposphere, the importance of CO is its significant role in affecting, and possibly controlling, the oxidizing capacity of the earth's atmosphere. Reaction with CO is one of the principal processes by which OH is removed from the atmosphere. If the CO abundance of the atmosphere increases, the net effect is a decrease of OH. Since reaction with OH is the primary removal mechanism of most atmospheric chemical trace species, CO is important in determining the future concentrations of many environmentally important trace gases.

The major sources of CO in the atmosphere, including fossil fuel combustion, biomass burning, oceanic emissions, methane and non-methane hydrocarbon (NMHC) oxidation, and terrestrial biogenic emissions. These sources are not well understood, in particular their magnitudes and distributions are not well quantified. This study uses a simplified version of the GRANTOUR 3-D chemical tracer model (Walton et al. 1988; Penner et al., 1991) to analyze the cycle of CO in the atmosphere, and to better quantify these sources. The simplified chemistry is based on the eigenvalue solution of Prather (1994). The model simulates the cycle of CO using prescribed CH_4 fields, and OH and NMHC parameterizations from the full GRANTOUR chemistry (Penner, et al. 1994).

We run seven different model scenarios varying the source magnitudes and distributions of CO. These scenarios labeled (a) through (g) are described in Table 1. Carbon monoxide concentration results from the model scenarios are compared to

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Table 1. Description of the model input scenarios referred to in the text. Scenarios were run to constrain the magnitude of CO sources.

Scenario	Input sources for model runs (Tg CO yr^{-1})
(a)	Fossil fuel, biomass burning, 16 Tg ocean, NMHC oxidation
(b)	Fossil fuel, biomass burning, 16 Tg ocean, NMHC oxidation, 500 Tg winter source evenly distributed over all land masses
(c)	Fossil fuel, biomass burning, 165 Tg ocean, NMHC oxidation
(d)	Fossil fuel, biomass burning, 16 Tg ocean, NHMC oxidation, 380 Tg winter source evenly distributed over NH land masses
(e)	Fossil fuel, biomass burning, 16 Tg ocean, NMHC oxidation, 380 Tg winter source distributed according to NH biomass burning practices.
(f)	Fossil Fuel, biomass burning, 16 Tg ocean, 380 Tg distributed according to biogenic activity (Dignon et al., 1992)
(g)	Fossil fuel, biomass burning, 16 Tg ocean

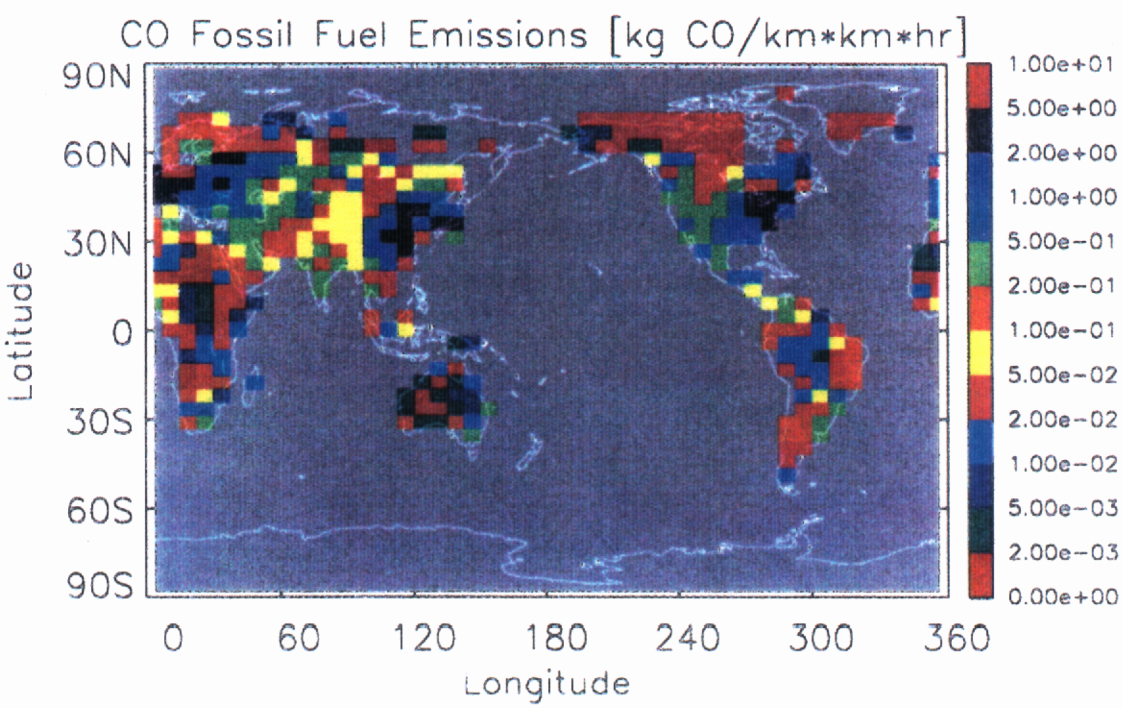


Figure 1. Global emissions inventory of CO emissions from Fossil Fuel Combustion (kg CO km^{-2} hr^{-1}), shown at the 4.5° latitude × 7.5° longitude GRANTOUR grid. (1°× 1° is available from the authors).

available observations published in the literature. The biomass burning source is based on Dignon and Penner (1991) for the boreal emissions and Liousse et al. (1996) for the tropics. In addition, we present a 1 degree latitude by 1 degree longitude emission inventory of CO from fossil fuel combustion. We believe analysis of these model scenarios using our new fossil fuel inventory has provided constraints on the magnitudes and distribution the other atmospheric sources of carbon monoxide.

Discussion

Generating source inventories are challenging and demanding projects that require constant updates, more accurate estimates of emission factors and source data in remote areas. Here we describe how we estimated CO emissions resulting from technological sources of fossil fuel consumption, industrial processes and waste disposal. CO emission for the base year 1987 were calculated to be approximately 525 Tg CO yr^{-1}, with the largest sector attributed to gasoline powered transportation (116 Tg CO yr^{-1}). Emission factors were those recommended by Logan et al. (1981), while fuel usage categories were from UN (1989), UN (1992), and IEA (1989). The emissions were derived for each country then distributed by population onto a $1° \times 1°$ degree grid within each country (Dignon, 1988; Logan, manuscript in preparation, 1995). For this project the emissions were then converted to the approximate $4.5° \times 7.5°$ grid of the GRANTOUR chemical transport model shown in Figure 1. The areas where emissions are concentrated, principally Northeastern USA, Central Europe, Eastern China, and Japan.

In our sensitivity study, scenario run (a) is our baseline case which includes fossil fuel combustion, biomass burning, NMHC oxidation and ocean sources (see Table 1.) Fossil fuel emissions were prescribed as indicated above. The biomass burning source the same as that used in Penner et al. (1994) and is based on Dignon and Penner (1991) for the boreal emissions and Liouse et al. (1996) for the tropics. This provides a Northern Hemisphere and Southern Hemisphere biomass burning source that is roughly equal. This distribution is unlikely since there is a larger land area and more population in tropical NH than the SH, however we begin with this scenario. From Figures 2 through 4, it is obvious that the CO concentration is low at most stations. Thus indicating that there is a CO source missing or under-represented.

In our comparison between runs (a) and (c), we are experimenting with the oceanic CO source. Run (c) uses the 165 Tg CO yr^{-1} ocean CO source estimate of Erickson and Taylor (1992). From Figures 2 and 3 we can see that at every remote site, where oceanic influence is greatest, the CO concentration is too high at least part of the year. In fact at the Southern Hemisphere sites of American Samoa (SMO) and Cape Grim (CGO) the concentrations are out of range for the plot. This suggest that a value of 165 Tg CO yr^{-1} is too large. Run (a) uses a source distributed geographically similar Erickson and Taylor (1992) but 10 times less in magnitude (16.5 Tg CO yr^{-1}). A recent report by Johnson and Bates (1996) supports this lower estimate. They report a global ocean source of 13 Tg. This also suggests that any additional CO source must be land based.

Southern Hemisphere Remote

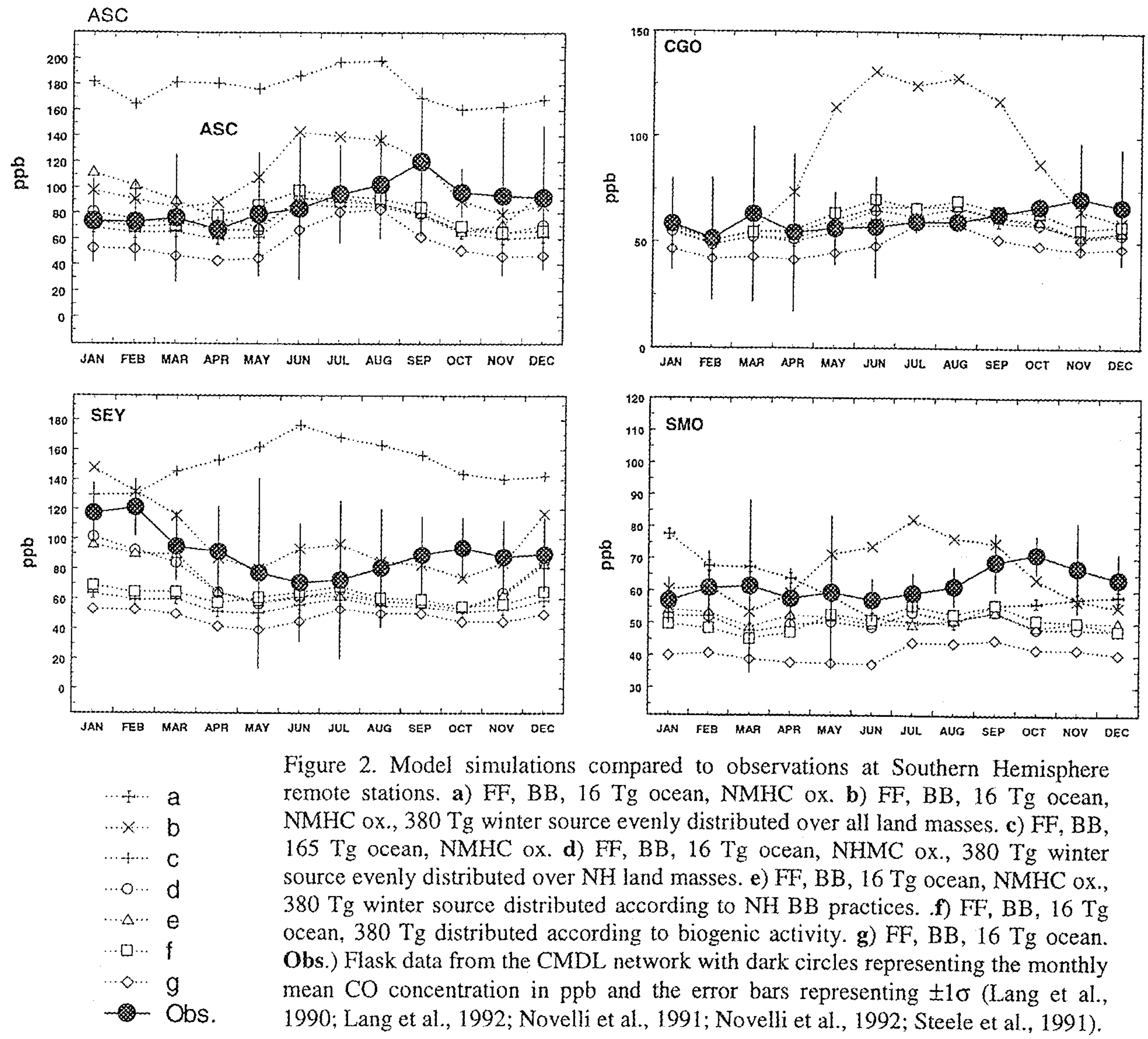


Figure 2. Model simulations compared to observations at Southern Hemisphere remote stations. **a**) FF, BB, 16 Tg ocean, NMHC ox. **b**) FF, BB, 16 Tg ocean, NMHC ox., 380 Tg winter source evenly distributed over all land masses. **c**) FF, BB, 165 Tg ocean, NMHC ox. **d**) FF, BB, 16 Tg ocean, NHMC ox., 380 Tg winter source evenly distributed over NH land masses. **e**) FF, BB, 16 Tg ocean, NMHC ox., 380 Tg winter source distributed according to NH BB practices. **.f**) FF, BB, 16 Tg ocean, 380 Tg distributed according to biogenic activity. **g**) FF, BB, 16 Tg ocean. **Obs.**) Flask data from the CMDL network with dark circles representing the monthly mean CO concentration in ppb and the error bars representing $\pm 1\sigma$ (Lang et al., 1990; Lang et al., 1992; Novelli et al., 1991; Novelli et al., 1992; Steele et al., 1991).

To test the land theory, scenario (b) adds an additional 500 Tg evenly to all land masses. In Figure 2, this run illustrates that there is no need to enhance the Southern Hemisphere source and that the underrepresented source is predominantly in the Northern Hemisphere. Analysis of figures 3 and 4 suggest that the source is not as large as 500 Tg, probably more localized at lower latitudes, and it is primarlly in the boreal winter season. Additional emissions in the winter season therefore suggest that there is not a large additional CO source from biogenic emissions of CO or chemical production from additional natural hydrocarbons. Run (d) adds the 380 Tg to all Northern Hemisphere land masses in the winter season only. looking particularly at the Barrow,

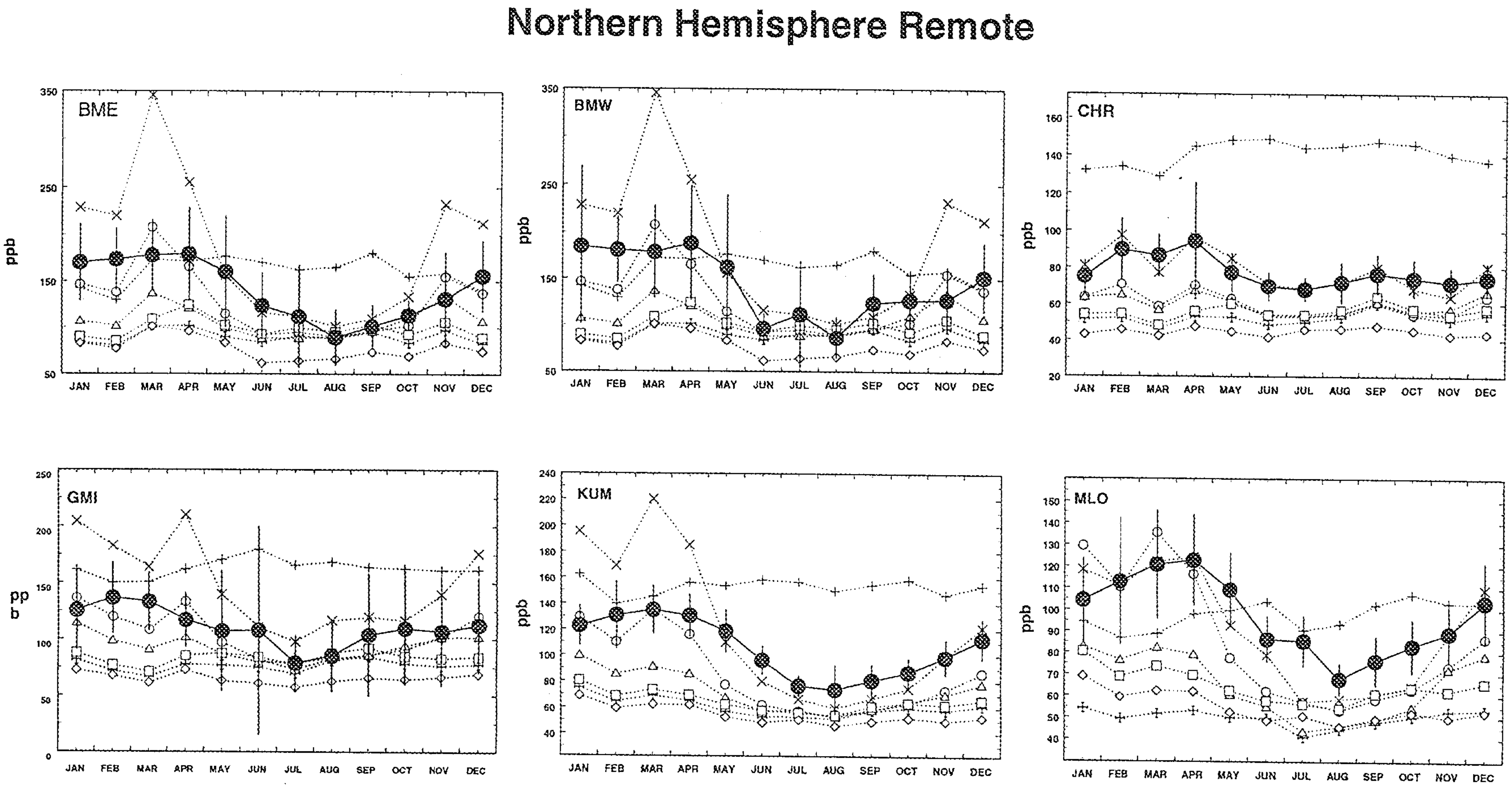
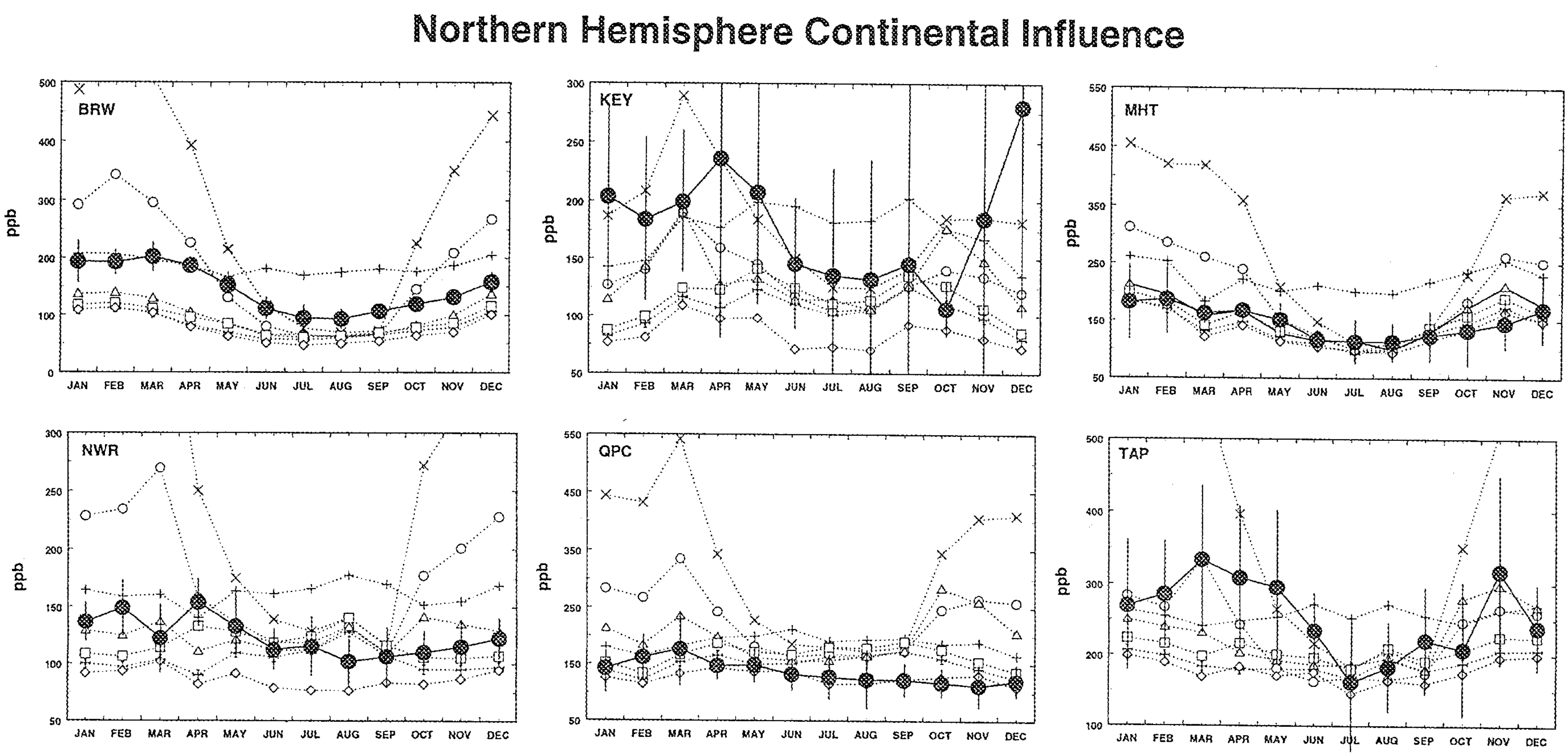


Figure 3. Model simulations compared to observations at Northern Hemisphere remote stations. **a)** Fossil fuel, biomass burning, 16 Tg ocean, NMHC oxidation. **b)** Fossil fuel, biomass burning, 16 Tg ocean, NMHC oxidation, 380 Tg winter source evenly distributed over all land masses. **c)** Fossil fuel, biomass burning, 165 Tg ocean, NMHC oxidation. **d)** Fossil fuel, biomass burning, 16 Tg ocean, NHMC oxidation, 380 Tg winter source evenly distributed over NH land masses. **e)** Fossil fuel, biomass burning, 16 Tg ocean, NMHC oxidation, 380 Tg winter source distributed according to NH biomass burning practices. **f)** Fossil Fuel, biomass burning, 16 Tg ocean, 380 Tg distributed according to biogenic activity **g)** Fossil fuel, biomass burning, 16 Tg ocean. **Obs.)** Flask data from the CMDL network with dark circles representing the monthly mean CO concentration in ppb and the error bars representing ±1σ (Lang et al., 1990; Lang et al., 1992; Novelli et al., 1991; Novelli et al., 1992; Steele et al., 1991).



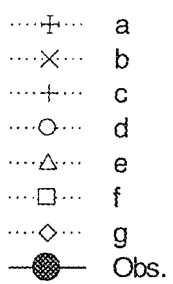


Figure 4. Model simulations compared to observations at Northern Hemisphere stations with continental influence. **a**) FF, BB, 16 Tg ocean, NMHC ox.. **b**) FF, BB, 16 Tg ocean, NMHC ox., 380 Tg winter source evenly distributed over all land masses. **c**) FF, BB, 165 Tg ocean, NMHC ox.. **d**) FF, BB, 16 Tg ocean, NHMC ox., 380 Tg winter source evenly distributed over NH land masses. e) FF, BB, 16 Tg ocean, NMHC ox., 380 Tg winter source distributed according to NH BB practices. **f**) FF, BB, 16 Tg ocean, 380 Tg distributed according to biogenic activity. **g**) FF, BB, 16 Tg ocean. **Obs.**) Flask data from the CMDL network with dark circles representing the monthly mean CO concentration in ppb and the error bars representing $\pm 1\sigma$ (Lang et al., 1990; Lang et al., 1992; Novelli et al., 1991; Novelli et al., 1992; Steele et al., 1991).

AK (BRW), Mace Head Ireland (MHT), and Niwot Ridge, CO (NWR) stations again this supports the idea that the source may be more tropically located.

Most of the biomass burning in the Northern Hemisphere tropics is between October and March, therefore a predominantly Northern Hemisphere winter source. Run (e) adds 380 Tg to the Northern Hemisphere, while scaling this total to our existing biomass burning source distribution. Thus, giving a total biomass burning source of 830 Tg CO yr^{-1} with roughly 230 Tg CO from the Southern Hemisphere and 500 Tg CO from the Northern Hemisphere. Recent estimates of the biomass burning source seem to support this higher value as well (Levine and Pinto, 1997; Chatfield, 1997).

Run (f) was used to constrain the biogenic source of CO. A 380 Tg CO yr^{-1} biogenic source was scaled according to the biogenic source of NO_x developed by Dignon et al. (1992). This brings the CO concentrations closer to the observations when compared to run (a) however the seasonal cycle is reversed in the Northern Hemisphere. Again supporting the idea that it is predominantly a NH winter source. However, the Southern Hemisphere has improved. Therefore a 380 Tg CO yr^{-1} biogenic source is most likely an upper limit.

The eigenvalue chemical solution method (Prather, 1994) used in the GRANTOUR model allows us to determine an upper limit of CH_4 oxidation Therefore, we have calculated the upper limit of CO produced from CH_4 oxidation to be 950 Tg CO yr^{-1}. Table 2. illustrates our model estimates of all of the CO sources discussed.

Conclusions

An increase in atmospheric CO concentration could trigger a profound change in the chemistry of the atmosphere that would require many years for the system to recover. This idea has also been recently been suggested in terms of chemical eigenstates of the atmosphere by Prather, 1994. It is necessary further study of the CO cycle in order to understanding the future of our atmosphere. This begins with quantifying the magnitudes and distributions of the various CO sources.

Table 2. Estimate of CO sources in Tg CO yr^{-1}.

Source	**Tg CO yr^{-1}**
Fossil Fuel	525
Biomass Burning	450-830
NMHC Oxidation*	580
Ocean Emissions	16
Biogenic Emissions	<380
Methane Oxidation	<950

We have presented a CO emissions inventory on a 1°×1° grid from fossil fuel combustion for use in 2 and 3-dimensional model studies. These results were used in the GRANTOUR chemical transport model. GRANTOUR model results suggest that in order to balance the CO budget an additional Northern Hemisphere winter source is required. Some or all of this additional source may be attributable to unaccounted biomass burning.

Oceanic source of CO has been estimates between 10 and 1200 Tg CO yr^{-1} (Johnson and Bates, 1997; Erickson and Taylor, 1992; Zuo and Jones, 1995). Sensitivity studies with the GRANTOUR model suggest that the oceanic source is on the order of 10^1 which is supported by the most measurements. In addition we have provided upper limits to the biogenic and methane oxidation sources. Results from this work will be included in future global change research incorporating full chemistry 3-D simulations and have already been used in CH_4 perturbation global warming calculations (Grossman et al., 1998).

Acknowledgements

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