

Sensitivity of tropospheric oxidants to biomass burning emissions: implications for radiative forcing

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[1] Biomass burning is one of the largest sources of trace gases and aerosols to the atmosphere and has profound influence on tropospheric oxidants and radiative forcing. Using a fully coupled chemistry-climate model (GFDL AM3), we find that co-emission of trace gases and aerosol from present-day biomass burning increases the global tropospheric ozone burden by 5.1% and decreases global mean OH by 6.3%. Gas and aerosol emissions combine to increase CH₄ lifetime nonlinearly. Heterogeneous processes are shown to contribute partly to the observed lower ΔO₃/ΔCO ratios in northern high latitudes versus tropical regions. The radiative forcing from biomass burning is shown to vary nonlinearly with biomass burning strength. At present-day emission levels, biomass burning produces a net radiative forcing of -0.19 W/m^2 (-0.29 from short-lived species, mostly aerosol direct and indirect effects, $+0.10$ from CH₄- and CH₄-induced changes in O₃ and stratospheric H₂O) but increases emissions to over 5 times present levels would result in a positive net forcing. **Citation:** Mao, J., L. W. Horowitz, V. Naik, S. Fan, J. Liu, and A. M. Fiore (2013), Sensitivity of tropospheric oxidants to biomass burning emissions: implications for radiative forcing, *Geophys. Res. Lett.*, 40, 1241–1246, doi:10.1002/grl.50210.

1. Introduction

[2] Biomass burning influences global oxidation capacity in complex ways, resulting from emissions of a mixture of trace gases and aerosols [Andreae and Merlet, 2001]. The trace gas emissions from wildfires produce ozone through the photochemical oxidation of carbon monoxide (CO) and hydrocarbons in the presence of nitrogen oxide (NO_x) [Mauzerall et al., 1998]. Meanwhile, emitted CO and hydrocarbons provide additional sinks for OH [Duncan et al., 2003a], extending the lifetime of methane (CH₄). Aerosol emissions

can influence photochemistry directly by changing photolysis rates [Martin et al., 2003] and heterogeneous interaction with gas-phase species [Jacob, 2000; Pozzoli et al., 2008] and indirectly by induced changes in clouds [Jiang et al., 2012; Liao et al., 2009]. As a result, beside the direct and indirect effects from biomass burning aerosols, its influence on radiative forcing can be further complicated by the changes in greenhouse gases, including ozone, CH₄ (via OH), and stratospheric water vapor (via CH₄ oxidation).

[3] An important feature of biomass burning is its significant inter-annual variability [Duncan et al., 2003a; Lamarque et al., 2010; Schultz et al., 2008; van der Werf et al., 2010], resulting from both natural and anthropogenic activities, such as El Niño, changes in land use, and population density [Field et al., 2009]. Akagi et al. [2011] estimate that the total fire emissions during El Niño years can be two times larger than other years. Meanwhile, considerable uncertainties remain in current estimates of biomass burning emission, with a range of a factor of 3 to 5 for individual species [Akagi et al., 2011; Bond et al., 2013]. We expect even greater uncertainties on constraining preindustrial biomass burning emissions [Mouillot et al., 2006] and predicting future emissions [Pechony and Shindell, 2010; Spracklen et al., 2009]. To take these into account, we conduct perturbation tests at different magnitude of emissions to quantify the impact of biomass burning on tropospheric oxidants. We then calculate the resulting changes in radiative forcing from aerosol effects and the change in tropospheric oxidant levels.

2. Methodology

[4] We use the Geophysical Fluid Dynamics Laboratory (GFDL) global chemistry-climate atmosphere model (AM3) for this study. Model physics and chemistry in AM3 are described in detail in Donner et al. [2011] and Naik et al. (V. Naik et al., Preindustrial to present day impact of changes in short-lived pollutant emissions on atmospheric composition and climate forcing, submitted to *Journal of Geophysical Research: Atmospheres*, 2012; hereinafter Naik et al., submitted). In particular, AM3 solves both tropospheric and stratospheric chemistry over the full domain, thus representing more realistic interactions between stratosphere and troposphere than many other global chemistry models. AM3 parameterizes aerosol activation into liquid cloud droplets [Ming et al., 2006] and computes the aerosol indirect effect from the increase of cloud albedo (first indirect effect), the increase of cloud lifetime (second indirect effect), and cloud evaporation from absorbing aerosols (semi-direct effect).

[5] We here incorporate recent updates of heterogeneous reactions, including HO₂, NO₂, N₂O₅, and NO₃, on the basis of work by Mao et al. [2013] and Jacob [2000] (Table S3 in

All Supporting Information may be found in the online version of this article.

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Supporting Information). In particular, the new HO₂ uptake scheme by aerosols (HO₂ → H₂O with $\gamma(\text{HO}_2) = 1$) provides a much stronger sink of HO₂ than previously thought, leading to a significant decrease of global OH. We also allow aerosol uptake on all types of aerosols, compared to only sulfate in standard version of AM3. As a result, these updates on heterogeneous reactions lead to significant improvements to simulated CO concentrations (Figure S2) and also decreases the N/S inter-hemispheric OH ratio from 1.16 [Naik *et al.*, submitted] to 1.06, in better agreement with the best estimate of 0.98 derived from methylchloroform observation [Krol and Lelieveld, 2003]. In order to take into account the effect of aerosols on actinic fluxes and photolysis rates, we calculate photolysis rates using the Fast-JX module with online cloud and aerosol fields [Bian and Prather, 2002].

[6] Biomass burning and anthropogenic emissions in this study follow the emission inventory for year 2000 provided by Lamarque *et al.* [2010]. In particular, biomass burning emissions in the model include CO (459 Tg/yr, 37% of global total emission), NO_x (5.5 Tg N/yr, 13%), NMVOC (37 Tg C/yr, 3.4%), SO₂ (1.9 Tg S/yr, 3.6%), BC (2.6 Tg/yr, 34%), and OC (23.3 Tg/yr, 65%). Biogenic emissions follow a climatology dataset (precursors of ozone and their effects in the troposphere) for 2000 (<http://accent.aero.jussieu.fr/POET.php>), to avoid the added complexity from feedbacks of model meteorology on biogenic emissions. We prescribe the concentrations of well-mixed greenhouse gases, including CO₂, N₂O, CH₄, and halocarbons, at their year 2000 values [Naik *et al.*, submitted]. In particular, CH₄ is held at 1751 ppb. We force the model with observed sea surface temperatures and sea-ice cover.

[7] We design five sets of simulations, multiplying the standard biomass burning emissions of aerosols and ozone precursors by a factor of 0, 1, 2, 5, and 10, respectively, retaining the same geographic distribution. We conduct three model experiments in each set of simulations: both aerosol and gas emissions (BB), aerosol emissions only (BB_AERO), and gaseous emissions only (BB_GAS). We also conduct additional sensitivity simulations with heterogeneous reactions turned off in our model. For each experiment, we conduct a continuous six-year run (2000–2005) with annually repeating emissions; the first year is discarded as model spin-up, and we analyze results from the final 5 years of each simulation in the following sections.

3. Perturbations to Tropospheric Oxidants

[8] The perturbations to annual mean tropospheric OH, O₃ burden, odd-oxygen (O_x) production rates, CO burden, methane lifetime, and methyl chloroform lifetime resulting from changes to biomass burning are shown in Figure 1. We focus on annual mean results because seasonal variations are found to be relatively small. We define the chemical troposphere as the atmospheric region where ozone is less than 150 ppb, following Prather *et al.* [2001]. The calculation of global OH, methane lifetime, and methyl chloroform lifetime follows the air mass-weighted approach [Lawrence *et al.*, 2001]. In the base run (1BB, naming is described in Table S4), mass-weighted global tropospheric OH is 9.8×10^5 molecules cm⁻³, lower than 10.5×10^5 molecules cm⁻³ calculated in Naik *et al.* (submitted), as a result of the updates described in previous section. The calculated methane lifetime and methyl chloroform lifetime against loss by tropospheric OH are 9.8

years and 5.8 years respectively in the base run (1BB), also longer than 8.5 years and 5.3 years in Naik *et al.* (submitted).

[9] Trace gas and aerosol emissions have opposite effects on global ozone burden and ozone production rate (Figures 1b and 1c). While trace gas emissions tend to increase global ozone burden and ozone production rate, aerosol emissions tend to decrease global ozone burden, mainly due to heterogeneous processes. The net effect on global ozone burden is an increase of 5.3% from 0BB to 1BB (Figure 1b). In contrast to the impact on ozone, both trace gas and aerosol emissions lead to a decrease of global OH (Figure 1a).

[10] Aerosol emissions influence global OH and ozone directly via photolysis and heterogeneous processes. The effect of biomass burning aerosols on photolysis rates is found to have only a minimal impact on global oxidant burdens. In separate runs with heterogeneous chemistry turned off (not shown), we find that global OH and ozone are not sensitive to aerosol emissions from biomass burning. This small global effect likely results from a cancellation between the suppression of ozone production in source regions and its enhancement downstream. Similar conclusions were found previously by Jonson *et al.* [2000] and Bian *et al.* [2003]. It should be noted that regional impacts, however, can still be significant [Martin *et al.*, 2003]. The indirect effect of aerosols on clouds appears to have minimal impacts on tropospheric oxidants, as it is limited by the small mass fraction of the atmosphere actually occupied by cloud [Jacob, 2000]. Therefore, the impact from aerosol emissions is mainly driven by heterogeneous processes.

[11] We now proceed to evaluate the effects of combined trace gas and aerosol emissions. Global mean OH decreases by 6.3% when biomass burning emissions are doubled from 1BB to 2BB. The magnitude of this perturbation is in general consistent with previous direct and indirect estimates for the 1997 Indonesia fire, as a typical El Niño year with biomass burning emissions doubled [Akagi *et al.*, 2011]. Duncan *et al.* [2003b] shows from a model study that global mean OH decreases by 6% in October and November of 1997. Prinn *et al.* [2005] shows from the inverse modeling of methyl chloroform that global OH dropped by 6% in 1997–1999 relative to 1978–2004 average values.

[12] CH₄ lifetime and CH₃CCl₃ lifetime are considerably enhanced by the co-emitted gas and aerosol emissions (Figures 1e and 1f). We find that CH₄ lifetime increases only by 0.18 year and 0.30 year from 0BB to 1BB_GAS and 1BB_AERO, respectively. However, the combination of both emissions leads to the increase of methane lifetime by 0.58 year (from 0BB to 1BB), greater than the arithmetic sum of impacts from separate aerosol and gas emissions. In fact, this nonlinear effect becomes more significant with increasing magnitude of biomass burning emissions. We find that the co-emission leads to an increase of CH₄ lifetime by 4.0 years from 0BB to 5BB, in contrast to 1.2 years from 0BB to 5BB_AERO and 1.6 years from 0BB to 5BB_GAS. Such non-linearity is mainly due to the extended lifetime of CO (and other VOCs) in biomass burning plumes, resulting from aerosol uptake. This leads to significant increase of global CO burden (Figure 1d), which in turn further reduces global OH.

[13] Another prominent feature in Figure 1 is the linearity of tropospheric ozone burden versus biomass burning emissions, reflecting NO_x-limited O₃ production on a global scale. This linearity is largely determined by the high VOC/NO_x ratio of wildfire emissions, as ozone production

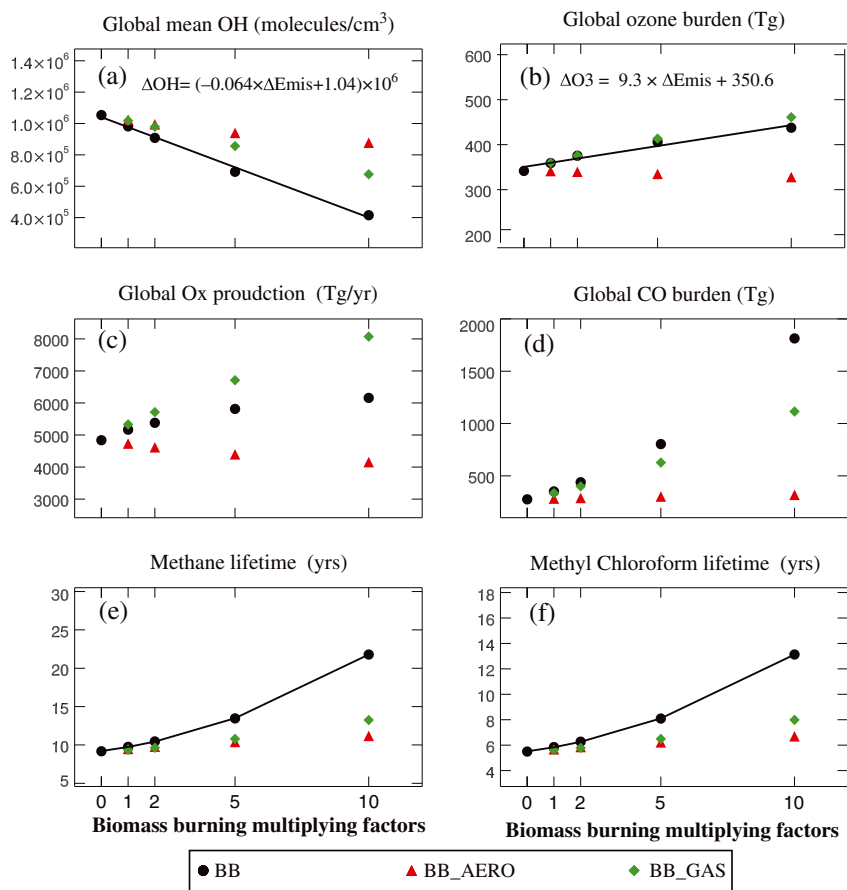


Figure 1. Perturbation to global oxidation capacity by the magnitude of biomass burning emissions, including (a) global mean OH (molecules/cm³), (b) global ozone burden (Tg), (c) global O_x production rate (Tg/yr), (d) global CO burden (Tg), (e) methane lifetime (years), and (f) methyl chloroform lifetime (years). Black circles are the runs with both gaseous and aerosol emissions (aerosol and gas). Red triangles are the runs with zero gas species emissions from biomass burning (aerosol only). Green diamonds are the runs with zero aerosol emissions from biomass burning (gas only). Solid line shows least square linear fit to “BB” simulation points in Figures 1a and 1b and connects estimated lifetimes in Figures 1e and 1f. Here O_x is defined as O₃ + NO₂ + 2*NO₃ + 3*N₂O₅. O_x production rate is then defined as the sum of HO₂ + NO and RO₂ + NO reaction rates.

efficiency is weakly dependent on VOC or CO levels [Wang and Jacob, 1998]. Similar linearity of ozone responses was found for perturbation tests of individual CO and NMVOC emissions [Wu *et al.*, 2009] and CH₄ emissions [Fiore *et al.*, 2008]. The approximate linearity justifies the additivity of the forcings from CH₄-induced O₃ as described below.

4. Ozone Production Efficiency ($\Delta O_3/\Delta CO$)

[14] The enhancement ratio $\Delta O_3/\Delta CO$ has been measured extensively as a metric to characterize O₃ production efficiency from fire plumes. Previous measurements indicate significantly higher $\Delta O_3/\Delta CO$ ratios in tropical regions (0.63 ± 0.19) than northern high latitudes (0.22 ± 0.23) for aged fire plumes (≥ 5 days) [Jaffe and Wigder, 2012]. Using global measurements of $\Delta O_3/\Delta CO$ ratios, Jaffe and Wigder [2012] estimated global ozone production rates to be 174 Tg/yr from biomass burning, using emissions from Global Fire Emissions Database 3 (GFED3) [van der Werf *et al.*, 2010]. In contrast, our model shows a higher increase of global ozone production (328 Tg/yr) from 0BB to 1BB (Figure 1c). However, this rate increases to 617 Tg/yr with all heterogeneous reactions turned off (not shown), suggesting

the importance of heterogeneous processes in suppressing ozone production.

[15] We here use the difference of annual mean burden between 0BB and 1BB, to examine modeled $\Delta O_3/\Delta CO$ in four latitude bands (90S–10S, 10S–30N, 30N–60N, and 60N–90N) as categorized in Jaffe and Wigder [2012]. In the annual mean, we assume that the long-range transport from low latitudes to high latitudes has little influence on the latitudinal gradient of $\Delta O_3/\Delta CO$, which is instead mainly driven by local chemical changes within each latitude band. This simplistic approach allows us to examine $\Delta O_3/\Delta CO$ from a model perspective. As shown in Figure 2, $\Delta O_3/\Delta CO$ decreases significantly in all latitude bands as a result of including aerosol uptake.

[16] Another remarkable feature in Figure 2 is that we see the opposite latitudinal gradient of $\Delta O_3/\Delta CO$ in northern hemisphere when heterogeneous chemistry is turned off. With heterogeneous chemistry turned on, however, $\Delta O_3/\Delta CO$ decreases from 0.25 to 0.16 from 10S–30N to 60N–90N band, suggesting that heterogeneous chemistry is at least partly responsible for this observed discrepancy of $\Delta O_3/\Delta CO$ between the tropics and northern high latitudes [Jaffe and Wigder, 2012]. We attribute this suppression of ozone production to two causes. First, HO₂ uptake by aerosols can be

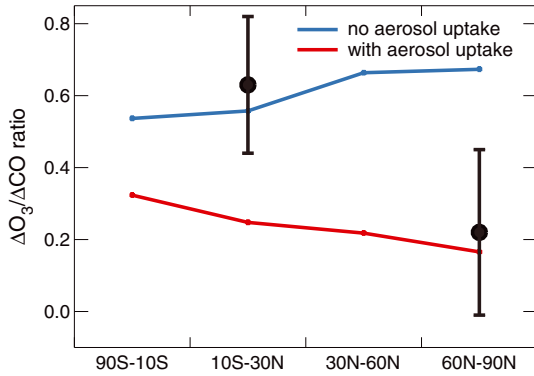


Figure 2. Comparison of observed and modeled $\Delta O_3/\Delta CO$ ratios. Observations (black solid circles with vertical bars) are from *Jaffe and Wigder* [2012]. Model ratios are calculated from the difference between 0BB and 1BB in annual mean burden of O_3 and CO when aerosol uptake is on (red solid lines) and off (blue solid lines).

increasingly important at high latitudes as a HO_x sink, because of the quadratic dependence of the gas-phase HO_x sink ($HO_2 + HO_2$) on HO_2 concentrations and the strong decrease of HO_2 concentrations towards high latitudes driven by solar zenith angle and water supply [*Logan et al.*, 1981]. Second, the sink of NO_x through heterogeneous uptake of N_2O_5 , NO_3 , and NO_2 becomes increasingly important relative to $OH + NO_2$ at high latitudes [*Dentener and Crutzen*, 1993; *Macintyre and Evans*, 2010]. We emphasize here that this impact is driven by the heterogeneous chemistry on all types of aerosols. This latitudinal gradient of $\Delta O_3/\Delta CO$ has important implications for understanding the ozone radiative forcing induced by biomass burning.

5. Radiative Forcing

[17] We now examine the radiative forcing, defined here as the change of top-of-the-atmosphere (TOA) net radiative flux (shortwave and longwave), due to biomass burning perturbations. Figure 3a shows the change of TOA net radiative forcing relative to a state without biomass burning (0BB). While trace gas emissions exert a slight positive forcing of $+0.03 \text{ W m}^{-2}$ (from 0BB to 1BB_GAS) as a result of the increase in ozone, aerosol emissions exert a negative forcing of -0.46 W m^{-2} (from 0BB to 1BB_AERO), due to both aerosol direct and indirect effects. Therefore, the combined emission of trace gases and aerosols (from 0BB to 1BB) is dominated by the effect of aerosols, producing a negative forcing of -0.29 W m^{-2} (Figure 3a). This forcing is consistent with a recent estimate of total forcing from open burning (-0.28 W m^{-2}) by *Bond et al.* [2013].

[18] Since our simulations hold CH_4 fixed, we cannot directly assess the radiative forcing that would result from the CH_4 changes induced by direct biomass burning emissions of CH_4 or by perturbations to tropospheric oxidants (and, thus, to CH_4 lifetime). After adjusting the changes in oxidants and to CH_4 emission changes from biomass burning, we estimate the steady state CH_4 mixing ratios to be 1506, 2050, and 3432 ppb for 0BB, 2BB, and 5BB, respectively (section S7 in Supporting Information), compared to a present-day value of 1751 ppb in 1BB. The radiative forcing due to the changes of CH_4 mixing ratios (CH_4_{ss} forcing) is calculated following the formulation of *Ramaswamy et al.* [2001]. We show in Figure 3b that the resulting change in net radiative forcing (versus 0BB) is 0.09, 0.20, and 0.60 W m^{-2} for 1BB, 2BB, and 5BB, respectively. This net radiative forcing (0.09 W m^{-2}) is comparable to a recent estimate ($0.05\text{--}0.13 \text{ W m}^{-2}$) by *Ward et al.* [2012].

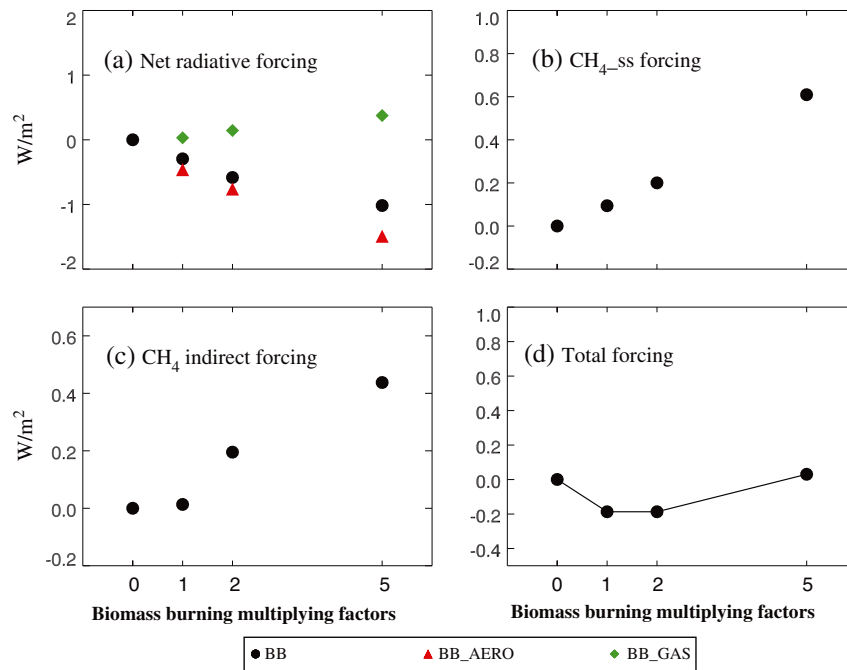


Figure 3. Response of net radiative forcing from perturbations of biomass burning emissions relative to a state without biomass burning (0BB), including (a) the combined rapid forcing from aerosols and ozone with fixed CH_4 concentrations, (b) the forcing from the estimated steady state change in CH_4 , and (c) the indirect forcing due to CH_4 -induced O_3 and stratospheric H_2O and (d) the total forcing. The simulation of 10BB is not shown.

[19] If the chemical system were allowed to respond to these estimated perturbations to CH₄ concentration, O₃ and stratospheric H₂O would change, leading to a further radiative forcing. To take into account this indirect forcing resulting from the CH₄-induced O₃ change and stratosphere H₂O change, we conduct additional AM3 simulations in the 1BB condition but with new prescribed CH₄ concentrations (1506, 2050, and 3432 ppb) as described above. The change of net radiative forcing from O₃ and H₂O is considered as the CH₄ indirect forcing resulting from the CH₄-induced O₃ change and stratosphere H₂O change (CH₄ indirect forcing). We show in Figure 3c that the CH₄ indirect radiative forcing is 0.01, 0.20, and 0.44 W m⁻² in the 1BB, 2BB, and 5BB simulations (versus 0BB), respectively. Our calculated radiative forcings from CH₄-induced changes are comparable to other model studies [Isaksen *et al.*, 2011].

[20] Figure 3d shows the sum of net radiative forcing from ozone and aerosols (Figure 3a), CH₄ perturbations (Figure 3b), and CH₄ indirect forcing (Figure 3c). We see that when the perturbation of biomass burning is larger than factor of 5, the indirect warming from CH₄ (Figures 3b and 3c) dominates over the direct cooling from emissions (Figure 3a), leading to a net warming effect from biomass burning. Despite the large uncertainties in current estimates of direct radiative forcing from biomass burning aerosols (+0.03 ± 0.12 W m⁻²) [Forster *et al.*, 2007], we show that the warming resulting from CH₄ and CH₄-induced changes in O₃ and stratospheric H₂O, which can be considered as an indirect chemical forcing, is comparable to the cooling from biomass burning aerosols with direct and indirect effect taken into account.

6. Conclusion and Discussion

[21] Trace gases and aerosols are intrinsically co-emitted by biomass burning. We show here that this co-emission leads to a net increase in tropospheric ozone production and ozone burden, a decrease in global mean OH, and a non-linear increase of CH₄ lifetime, exceeding the arithmetic sum of separate aerosol and gas effects by more than 20%, mainly due to heterogeneous processes. We also show that heterogeneous chemistry contributes to the observed lower ΔO₃/ΔCO ratios in northern high latitudes versus tropical regions. The implied radiative forcings from the change in steady state CH₄ concentrations, and from CH₄-induced change in O₃ and stratospheric H₂O, are comparable to the direct and indirect forcing from biomass burning aerosols.

[22] In fact, our estimate of warming effect from biomass burning emissions could be biased low. First, we have not considered CO₂ and N₂O emissions from biomass burning in this study, which would lead to further warming [Ward *et al.*, 2012]. Second, we may severely underestimate the NMVOC emissions as demonstrated by a recent study [Akagi *et al.*, 2011], which would lead to further increase of ozone and decrease of global OH. Further, the decrease of global OH will also lead to the increase of hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs), which would also contribute to warming. Therefore, the assessment of radiative forcing from biomass burning must consider the indirect chemical forcing from its perturbation to tropospheric oxidants. In particular, a better understanding of heterogeneous chemistry would be of great importance for assessing the role of aerosols in climate system.

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