EMISSIONS OF SOME TRACE GASES FROM BIOMASS FIRES

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Abstract. Airborne measurements of 13 trace gases from seven forest fires in North America are used to determine their average emission factors. The emission factors are then used to estimate the contributions of biomass burning to the worldwide fluxes of these gases. The estimate for NH$_3$ (~7 Tg N yr$^{-1}$) is about 50% of the global emissions of this gas. Combined NH$_3$ and NH$_4^+$ emissions from biomass burning could be the most important component of the NH$_3$ cycle. N$_2$O from biomass burning (~2 Tg N yr$^{-1}$) is also significant worldwide. The estimate for NO$_x$ from biomass burning worldwide (~19 Tg N yr$^{-1}$), which is greater than previous estimates, is comparable to emissions from fossil fuel combustion. The estimate of the global flux of F$_{12}$ (CF$_2$Cl$_2$) from biomass burning based on the complete data set (~0.2 Tg yr$^{-1}$) is ~50% of the total global emission of F$_{12}$. However, this estimate is strongly influenced by a very high emission of F$_{12}$ from a fire in the Los Angeles Basin. Disregarding this fire yields a global flux of 0.06 Tg yr$^{-1}$ (~15% of total global emissions). The high emissions of NO$_x$ and F$_{12}$ are due in whole or part to the resuspension of previously deposited pollutants. Since this can be the only source of F$_{12}$ in the smoke from fires, deposition may be a significant sink for F$_{12}$. Our estimate for NO$_x$ emissions from biomass burning in the South Coast Air Basin of California is much greater than previous estimates.

Introduction

Emissions of trace gases from biomass burning are known to be an important source of several trace gases (e.g., CO$_2$, CO and CH$_4$) in the atmosphere [e.g., Crutzen et al., 1985]. Also, the emissions from fires can have a marked effect on local and regional air quality [e.g., Radke et al., 1978]. However, quantitative estimates of the contributions of such emissions to the global budgets of various trace species are difficult to make. For example, Logan et al. [1981] estimated that CO emissions from global biomass burning ranged from 310 to 1250 Tg CO yr$^{-1}$. Estimates are even more uncertain for species that are more difficult to measure (e.g., NO$_x$).

In this paper, we present measurements of some trace gas emissions from seven biomass fires in North America.

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Emission factors are calculated for some trace gases and estimates are made of the significance of such emissions on global and local scales.

Fires Studied and Measurement Techniques

The locations, times, sizes and fuel types for the seven fires examined in this study are listed in Table 1. The fires encompass a wide range of woody biomass fuel types, including the rarely studied chaparral of the Southwest United States.

With the exception of the data shown in Figure 1, which are based on steel canister samples collected on or near the ground (from towers) in the combustion zone, all measurements were obtained from the University of Washington's C-131A research aircraft. The aircraft instrumentation package and general sampling procedures have been described by Hegg and Hobbs [1980], Hegg et al. [1987] and Radke et al. [1988]. Many of the trace gases measured from the aircraft and from the ground (e.g., hydrocarbons, N$_2$O) were analyzed by a combination of gas chromatography and a variety of detection techniques including mass spectroscopy and electron capture using the methodologies described by Rasmussen et al. [1974] and Rasmussen and Khalil [1980, 1981]. The samples themselves were "grab" samples obtained in stainless steel electroplated canisters and analyzed within two weeks of collection.

Several points regarding sampling strategy are particularly important to our analysis. Firstly, considerable care was taken to characterize the ambient air not only at plume level but near the base of the fire column to permit evaluation of the effects of low-level entrainment of ambient air on plume concentrations. This was especially critical for the measurements in the Los Angeles Basin. Secondly, where possible, measurements were obtained above the plume to test for the presence of elevated polluted haze layers. Both of these possible influences were found to have an insignificant effect on plume concentrations.

Results

Emission Factors

Average emission factors for thirteen trace gases from the fires are shown in Table 2. The emission factors were derived using the carbon balance method [Radke et al.,...
TABLE 1. Fires Examined in This Study

<table>
<thead>
<tr>
<th>Name</th>
<th>Location</th>
<th>Dates</th>
<th>Size (ha)</th>
<th>Fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lodi I</td>
<td>Los Angeles Basin</td>
<td>December 12, 1986</td>
<td>40</td>
<td>Chaparral, Chamise</td>
</tr>
<tr>
<td>Lodi II</td>
<td>Los Angeles Basin</td>
<td>June, 22 1987</td>
<td>150</td>
<td>Chaparral, Chamise</td>
</tr>
<tr>
<td>Myrtle/Fall Creek Silver</td>
<td>Roseburg, Oregon</td>
<td>September 2, 1987</td>
<td>2,000</td>
<td>Pine, brush, Douglas Fir, True Fir, Hemlock</td>
</tr>
<tr>
<td>Hardiman</td>
<td>Grants Pass, Oregon</td>
<td>September 17-19, 1987</td>
<td>20,000</td>
<td>Jack Pine, Aspen, Birch</td>
</tr>
<tr>
<td>Eagle</td>
<td>Chapleau, Ontario (Canada)</td>
<td>August 28, 1987</td>
<td>325</td>
<td>Black Sage, Sumac, Chamishe</td>
</tr>
<tr>
<td>Battersby</td>
<td>Timmins, Ontario (Canada)</td>
<td>August 12, 1988</td>
<td>718</td>
<td>Jack pine, White &amp; Black Spruce</td>
</tr>
</tbody>
</table>

Fig. 1. Concentration ratio of CO/CO₂ (circles) and H₂ concentration (triangles) versus O₂ in the plume from the Battersby fire. The data were obtained at or within 100 m of the ground. The lines shown are linear regressions (see text).

The intra-fire variability in the emission factors commonly exceeds the inter-fire variability. This illustrates the importance of temporal variabilities in the fires (e.g., flame temperature) on emissions. We have noted this previously with regard to NH₃ emissions [Hegg et al., 1988]. This variability can obscure differences in emissions between fires due to fuel type.

The average emission factors of the trace gases, based on the measurements from all seven fires, have quite high standard deviations (Table 2). For example, the mean emission factor for CO is 91 ± 21 g kg⁻¹. Of the variance of 21 g kg⁻¹, 23% is due to intra-fire variability and the remainder to inter-fire variability. The uncertainties in the mean emission factors propagate through our estimates of the fluxes of the gases and our extrapolations to the global scale.

Global Significance of Emissions From Biomass Burning

If we had good estimates of the amount of biomass burned each year around the globe, we could multiply this quantity by the emission factor for each of the gases given in Table 2 to obtain estimates of the contribution of biomass burning to the global emission fluxes of the gases. Unfortunately, biomass burning worldwide is not well quantified. However, estimates are available of the amounts of CO and CO₂ produced annually from biomass burning [e.g. Crutzen et al., 1985; Mooney et al., 1987]. To utilize these estimates, the relative emission ratio of each trace gas to CO (or CO₂) was determined from the data listed in Table 2 and then this number was multiplied by the estimated global emission flux of CO (or CO₂) from biomass burning to obtain estimates of the contribution of biomass burning to the global emission flux of the trace gas. The emission factors for CO and CO₂ listed in Table 2 are in reasonable agreement with the values used by Logan et al. [1981] and Andreae et al. [1988]. However, since our estimates for CO₂ emissions are somewhat higher than normally assumed, and therefore may not be as typical of global emissions as our CO data, we have used CO as our "ratio species" for estimating global fluxes.

Listed in Table 3 are the derived values for the mean ratios of the emissions factors for the various trace gas species to the emission factors for CO, together with our estimates of the global fluxes into the atmosphere from biomass burning based on these ratios. The value for global CO emissions from biomass burning utilized in the flux.
calculation was 800 Tg yr\(^{-1}\) [Crutzen et al., 1985]. This flux is given by the authors as uncertain by a factor of two, an uncertainty that will propagate through all of our subsequent analysis. In comparison, using Radke's [1989] estimate for the amount of biomass burned globally (\(-10^4\) Tg yr\(^{-1}\)), and our emission factor for CO, we obtain 910 Tg yr\(^{-1}\) of CO worldwide from biomass burning.

The first species shown in Table 3 (O\(_3\)) is not emitted directly by fires. Rather it is created in the fire plume by the chemical interaction of reactive hydrocarbons and NO\(_x\) [Evans et al., 1974; Radke et al., 1978]. A regression of the O\(_3\) emission factor onto the NO\(_x\) emission factor for our data yields an intercept of \(-2.2\) Tg yr\(^{-1}\), a slope of 1.4 and a correlation coefficient (r) of 0.8 significant at the 98% level. This suggests an O\(_3\)-production mechanism analogous to that in photochemical smog formation. Furthermore, the mechanism appears to be limited by the amount of NO\(_x\) (i.e., there is insufficient NO\(_x\) to react with the available reactive hydrocarbons) and thus the O\(_3\) production increases with increasing NO\(_x\) [Dismitriodes and Dodge, 1983]. We can assess the global impact of biomass burning on ozone as follows. Based on a tropospheric volume of \(-6 \times 10^{18}\) m\(^3\) (derived from the global surface area and typical tropopause heights [Hegg, 1985]) and troposphere O\(_3\) concentrations of \(-30-50\) ppbv, we obtain a tropospheric O\(_3\) reservoir of \(-150-200\) Tg. This reservoir has a variable turnover time, depending on season and latitude, but is certainly no more than two weeks [Logan, 1985]. Total tropospheric O\(_3\) production must therefore be on the order of \(-4000\) Tg yr\(^{-1}\). Therefore, the global flux of O\(_3\) shown in Table 3 (48 Tg yr\(^{-1}\)) constitutes a very minor source of tropospheric O\(_3\).

Our estimate of the flux of NH\(_3\) from biomass burning (\(-8\) Tg yr\(^{-1}\)) suggests that it contributes significantly to the atmospheric reservoir of NH\(_3\). Since we have addressed this point previously [Hegg et al., 1988] we will not dwell on it here except to note that the present measurements support our contention that biomass burning is an important source of NH\(_3\).

Andreae et al. [1988] measured significant fluxes of particulate NH\(_4\)\(^+\) from biomass fires in the Amazon Basin and suggested that if (as seems likely) these emissions also contained substantial amounts of gaseous NH\(_3\) (which they did not measure), biomass burning would have a very substantial impact on the global NH\(_3\) cycle. Our worldwide flux estimate for NH\(_3\), (equivalent to \(-7\) Tg N yr\(^{-1}\) of NH\(_3\)) is twice the flux of NH\(_4\)\(^+\) from biomass fires estimated by Andreae et al. and clearly substantiates their conjecture. Indeed, if we assume that the fluxes of NH\(_4\)\(^+\) and NH\(_3\) from biomass burning are additive, the combined flux of \(-10\) Tg N yr\(^{-1}\) would be the most important input to the atmospheric NH\(_3\) cycle [c.f. Galbally, 1985].

The global flux of CH\(_4\) shown in Table 3 is in reasonable agreement with previous estimates. The measurements of CH\(_4\) also reveal an interesting facet of fire chemistry. A regression of the CH\(_4\) emission factor on to the ratio of CO to CO\(_2\) emission factors yields a linear correlation coefficient (r) of 0.92 significant at >99% confidence level. Clearly, the ratio of CO to CO\(_2\) in a plume is, in a broad sense, indicative of the extent of oxidation in a plume, with high ratios suggesting limited oxygen (i.e., the combustion is fuel rich) or possibly quenching of the oxidation reactions prior to
TABLE 3. Mean Values of $EF_x/EF_{CO}$ for Biomass Burning Calculated From the Data Listed in Table 2. Also Shown Are Estimates of the Global Fluxes of Various Trace Gases From Biomass Burning Based on the $EF_x/EF_{CO}$ Ratios, and Estimates of Worldwide CO Emissions From Biomass Burning From Crutzen et al. [1985]

<table>
<thead>
<tr>
<th>Species</th>
<th>$EF_x/EF_{CO}$</th>
<th>Estimated Flux From Biomass Burning Worldwide (Tg yr$^{-1}$)</th>
<th>Estimated Contribution of Biomass Burning to Worldwide Flux of Species (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$O_3$</td>
<td>0.06 ± 0.05</td>
<td>48</td>
<td>1</td>
</tr>
<tr>
<td>$NH_3$</td>
<td>0.01 ± 0.008</td>
<td>8</td>
<td>50</td>
</tr>
<tr>
<td>$CH_4$</td>
<td>0.03 ± 0.003</td>
<td>24</td>
<td>~5</td>
</tr>
<tr>
<td>$C_3H_6$</td>
<td>0.006 ± 0.001</td>
<td>5</td>
<td>?</td>
</tr>
<tr>
<td>$C_2H_6$</td>
<td>0.005 ± 0.002</td>
<td>4</td>
<td>?</td>
</tr>
<tr>
<td>$C_3H_8$</td>
<td>0.003 ± 0.001</td>
<td>2.4</td>
<td>?</td>
</tr>
<tr>
<td>$C_2H_2$</td>
<td>0.003 ± 0.001</td>
<td>2.4</td>
<td>?</td>
</tr>
<tr>
<td>$N_2C_4$</td>
<td>0.002 ± 0.002</td>
<td>1.6</td>
<td>?</td>
</tr>
<tr>
<td>$N_2O$</td>
<td>0.004 ± 0.001</td>
<td>3</td>
<td>20</td>
</tr>
<tr>
<td>$F_12$</td>
<td>0.0002 ± 0.0001</td>
<td>0.2</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>(0.00008 ± 0.00005)$^*$</td>
<td>(0.06)$^{***}$</td>
<td>(1.5)$^{***}$</td>
</tr>
<tr>
<td>$NO_X$</td>
<td>0.07 ± 0.04</td>
<td>56</td>
<td>40</td>
</tr>
</tbody>
</table>

* Where $EF_x$ is the emission factor of trace gas species $x$ and $EF_{CO}$ the emission factor of CO.

** The mean values of $EF_x/EF_{CO}$ were obtained by first ratioing the values of $EF_x$ to $EF_{CO}$ for each of the fires listed in Table 2 and then averaging these seven ratios.

*** A more conservative estimate obtained by eliminating results from the Lodi I fire (see text).

completion as suggested by Cofer et al. [1989]. For oxygen-limited conditions one would expect enhanced production of $CH_4$ (the most saturated hydrocarbon) due to anomalously high $H_2$ levels. A relatively extreme example of this phenomenon can be seen in the comparatively intense Battersby fire. Plotted in Figure 1 are the $CO/CO_2$ and $H_2$ concentrations as a function of $O_2$ concentration in the plume from the Battersby fire at or near ground level. (These samples were collected by automated steel canister samplers placed on the ground and in towers in the fire zone prior to fire ignition). The $H_2$ will only be present in substantial quantities under at least mildly oxygen-limited conditions, and thus supports the low $O_2$ hypothesis. Figure 1 confirms the utility of the $CO/CO_2$ ratio as a indicator of oxygen-limited combustion. The lines shown in the Figure are linear regression lines for $CO/CO_2$ and $H_2$ against $O_2$. The correlation coefficients ($r$) are -0.78 and -0.94 for $CO/CO_2$ and $O_2$, respectively. These data, together with the correlation between $CH_4$ and $CO/CO_2$, suggest that biomass fires are commonly oxygen limited. A similar conclusion has been reached by Cofer et al. [1985].

An interesting sidelight of the data on $H_2$ shown in Figure 1 is the very high $H_2$ levels. Indeed, $H_2/CO_2$ ratios calculated from these data produce numbers as high as 0.01-0.04. These are similar to values obtained by Crutzen et al. [1985] and support the view of these authors that biomass burning is an important source of atmosphere $H_2$.

Another interesting consequence of the fuel/air mixture, as revealed by the $CO/CO_2$ ratio, is the production of particulate smoke. Many of the fires studied showed pronounced increases in particulate mass emission factors as the fire became increasingly oxygen-limited (see, for example, Figure 2 which shows the particulate mass emission factor versus $CO/CO_2$ for the Battersby fire). Particulate mass used in these calculations was determined by weighing teflon filters through which samples from the plume were drawn.

![Fig. 2. Emission factor for particulates as a function of the CO/CO$_2$ concentration ratio in the plume from the Battersby fire.](image)

Our estimate for the worldwide flux of non-methane hydrocarbons (NMHC) from biomass burning shown in Table 3 (~15 Tg yr$^{-1}$) is consistent with previous estimates [e.g. Crutzen et al, 1985] of ~30 Tg yr$^{-1}$, since only a fraction of the NMHC's are included in our flux estimates.
Our estimate for the N2O emission flux (~2 Tg N yr⁻¹) is also consistent with previous estimates (e.g., 1.6 Tg N yr⁻¹) by Crutzen et al. [1985]. This confirms that biomass burning is a significant source of atmospheric N₂O.

The only sink for F12 that is generally considered in atmospheric budget calculations is loss by photodissociation in the stratosphere [National Research Council, 1983]. However, the global flux of F12 from biomass burning shown in Table 3 (~0.2 Tg yr⁻¹) is ~50% of the estimated yearly global emission of F12 (~0.4 Tg yr⁻¹) [National Research Council, 1983]. Since F12 cannot be produced by fires, it must have been previously deposited onto the fuel bed and revolatilized in the fires. Hence, our results suggest that deposition of F12 may be important on the global scale, contrary to current understanding of this issue. However, extrapolation of our average emission factor for F12 to global scales may not be warranted. This is because the mean value of the F12/CO ratio used in our calculation of the global flux of F12 from biomass burning is strongly influenced by the very high emissions of F12 that we measured from the Lodi I fire in the Los Angeles Basin (see Table 2). The F12 emissions from Lodi I are five times greater than those from the Lodi II fire, which was at the same location as Lodi I. The reason for the high emissions of F12 from Lodi I is not clear, although several speculations are possible. For example, the Lodi I fire, in contrast to Lodi II, took place in the relatively wet winter season and, in fact, occurred only six days after significant rainfall. This rainfall could have produced high deposition of organic particulates in which the F12 could have been dissolved at relatively high levels. Concentrations of various organic species in both aerosols and rain are known to be quite high in Los Angeles [Seinfeld, 1989; Pankow et al., 1983]. Furthermore, particle scavenging by hydrometeors is a significant pathway for organic aerosol removal [Pankow et al., 1983]. However, neither laboratory chemical data nor field data are available to permit a quantitative appraisal of the high F12 values measured in the smoke from Lodi I.

It is also important to note that the F12 data from Lodi I do not show the same excellent internal consistency as those from Lodi II or the other fires. For example, the F12 emissions for Lodi I do not correlate with the CO emissions. This casts some suspicion on the Lodi I data. It is conceivable that the Lodi I samples were contaminated by F12 aboard the aircraft (which was used to calibrate the nephelometers). However, it is difficult to see how only the plume and not the ambient samples were contaminated if this were the case. We therefore feel there is no compelling reason to disregard the Lodi I data. F12 was not aboard on any of the other flights.

Nevertheless, if the high emissions from Lodi I are not included in our calculations, the reduced average F12 to CO emission ratio of ~7.6 x 10⁻⁵ yields a global flux of ~0.06 Tg yr⁻¹ of F12 from biomass burning worldwide. This is still quite significant (15% of the worldwide flux of F12). Thus, the deposition of F12 onto the earth's surface could be the tropospheric sink postulated as a possible explanation for discrepancies in F12 atmospheric lifetime estimates [Cunnold et al., 1983].

Our estimate for the global flux of NOₓ from biomass burning (~56 Tg N yr⁻¹) converts to ~19 Tg N yr⁻¹ (based on our measurements which show the NOₓ to be ~70% NO₂). Logan [1983] estimated a global flux of NOₓ from biomass burning of 12 Tg N yr⁻¹, with a possible range of 4-24 Tg N yr⁻¹. Our flux estimate, while compatible with Logan's wide range, suggests that NOₓ from biomass burning may be more significant than previously assumed. For example, our estimate for the global flux of NOₓ from biomass burning is ~40% of total NOₓ emission to the atmosphere, which makes it comparable to that generally given for NOₓ emissions from fossil fuel combustion [Crutzen et al., 1979; Logan, 1983].

One possible explanation for the high emission factor for NOₓ from biomass burning that we have estimated is the revolatilization of NOₓ previously deposited on the vegetation and ground. We have discussed this possibility in an earlier study [Hegg et al., 1988]. The data presented here provide additional support for this hypothesis. A regression of the NOₓ emission factor onto the F12 emission factors yields an intercept of 2.7 g kg⁻¹, a slope of 145 and a linear correlation coefficient of 0.8 significant at >95% confidence level (Figure 3). Because the source of F12 must be deposition of pollutants, its correlation with NOₓ suggests that a substantial fraction of the emitted NOₓ is also due to deposition. Indeed, the regression intercept, which can be interpreted as the emission factor for NOₓ after discounting revolatilization, suggests that ~30% of the average NOₓ emitted from the seven fires examined was due to the deposition of pollution. For the fires in Southern California, the percentage would clearly be much higher but is difficult to quantify because only two fires are available for regression analysis. It is also noteworthy that a calculation of global NOₓ flux from biomass burning using the intercept NOₓ emission factor ratioed to the average emission factor for CO shown in Table 2, yields ~8 Tg N yr⁻¹, which is more in line with earlier estimates [Crutzen et al., 1979; Logan, 1983]. Clearly, resuspension by biomass burning of NOₓ previously deposited from air pollution should be taken into account in global flux estimates.

Local Effects of Emissions from Biomass Burning

Most urban, photochemical air pollution is generally attributed to emissions from combustion sources, such as power plants and automobiles. In particular, hydrocarbon and NOₓ emissions from motor vehicles are generally considered to be the major sources of air pollution in Los Angeles [Finlayson-Pitts and Pitts, 1985]. However, the relatively high emissions of NOₓ from biomass burning that we have estimated in this paper make it prudent to reassess this view.

Emissions of CO and NOₓ are available for the South Coast Air Basin of California (which encompasses the counties of Los Angeles, Orange, Riverside and San Bernardino). Relevant data are shown in Table 4. Utilizing the data in Table 2 for NOₓ and CO emissions from the two Lodi fires in the Los Angeles Basin, the ratio of the emission factor of NOₓ to CO is found to be ~0.082. The product of this quantity with the emissions of CO from wildfires shown in Table 4 yield the estimates shown in the last column of Table 4 for NOₓ emissions from fires in the South Coast Air Basin. Although our emission factors yield far more NOₓ,...
from fires in the South Coast Air Basin than do the values used by the California Air Resources Board, the estimates of total NO$_X$ emissions in the South Coast Air Basin are not significantly perturbed by this revision, since biomass burning in the South Coast Air Basin is small compared to emissions from motor vehicles. However, the seasonal and episodic nature of biomass burning could make emissions from this source important at various times of the year.

Conclusions

Measurements of the emissions of a number of trace gases from biomass fires in North America have been used to calculate emission factors for these gases. While most of the emission factors are in reasonable agreement with previous estimates, several of them are not. In particular, our estimates of the emissions of NH$_3$, NO$_X$ and F12 are considerably higher than previous estimates. The enhanced NO$_X$ and F12 emissions were probably due to resuspension of previously deposited pollutants. This suggests that re-evaluations of the global budgets of NH$_3$, NO$_X$ and F12 may be in order.

Another interesting facet of our emissions data is the suggestion that biomass fires may, to some extent, be oxygen limited. This leads, for example, to a more prolific production of saturated hydrocarbon species (e.g. CH$_4$) than might be expected.

Our data also suggests that NO$_X$ emissions from biomass burning in the South Coast Air Basin of California may be much greater than previously thought.
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