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Trace Gas Emissions From a Mid-Latitude Prescribed Chaparral Fire

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Gas samples were collected in smoke plumes over the San Dimas Experimental Forest during a 400-acre prescribed chaparral fire on December 12, 1986. A helicopter was used to collect gas samples over areas of vigorous flaming combustion and over areas of mixed stages (vigorous/transitional/smoldering) of combustion. Sampling was conducted at altitudes as low as 35 m and as high as 670 m above ground level. The samples, collected in 20-L Tedlar bags, were returned by helicopter to our field laboratory and immediately transferred to electropolished stainless steel canisters. Analyses for the trace gases carbon dioxide (CO_2), carbon monoxide (CO), hydrogen (H_2), methane (CH_4), total nonmethane hydrocarbons (TNMHC), and nitrous oxide (N_2O) were performed within hours after collection. Samples of gas were also collected upwind of the burn and analyzed to determine ambient background levels. Mean emission ratios ($\Delta X/\Delta CO_2$, where X is equal to each species, vol/vol) determined for these gases relative to CO_2 were generally lower (except for N_2O) than mean emission ratios previously reported for large biomass-burning field experiments. No substantial differences in CO_2 normalized emission ratios for these gases (except for N_2O) were determined when samples from vigorously flaming and mixed stages of combustion were compared. Emissions from flaming portions of chaparral, however, would be expected to release disporportionately larger volumes of trace gases into the sampled smoke plumes. This, coupled with the very high surface-to-volume fuel ratio typical of a chaparral fire (which minimizes the smoldering stage), may have accounted for the consistency in the emission ratios.

INTRODUCTION

It has become increasingly apparent over the last decade that the burning of biomass, e.g., forests, vegetation, and grasslands, is a significant global source of environmentally important atmospheric trace gases, including carbon dioxide (CO_2), carbon monoxide (CO), nitric oxide (NO), nitrous oxide (N_2O), methane (CH_4), and other hydrocarbons. Many diverse and widespread burning practices combine to yield a large total global amount of surface biomass burned each year. Biomass burning includes the burning of forests for clearing, the burning of vegetative stubble after harvesting, and lightning- and human-induced wildfires. Recent estimates of the global surface area involved in biomass burning each year range from 3 to 7 × 10^6 km^2, which represents between 2–5% of the total land area of our planet [*National Academy of Sciences* (*NAS*), 1984]. Estimates for the total biomass burned range from 4400 to 7000 Tg/yr [*NAS*, 1984].

During biomass burning, large amounts of living and dead material are chemically transformed, mobilized, and volatilized. This may occur quickly during the very active stages of fires, or more slowly during the smoldering process. The composition and amount of trace gas production depends primarily upon the rates of energy release (intensity) and fire spread. Both of these are strongly controlled by fuel moisture content, fuel type, fuel array, ignition pattern, terrain, and weather.

To date, most of the large field studies of trace gas production in biomass burns have occurred in the tropics, or, to a lesser degree, in the western and southeastern United States [*Crutzen et al.*, 1979, 1985; *Delmas*, 1982; *Greenberg et al.*, 1984; *Ward and Hardy*, 1984]. The limited sets of measurements of gas emissions from these fires have been extrapolated to estimate gas emissions from biomass burning on a planetary scale [*Seiler and Conrad*, 1987]. Tropical fires have received special emphasis because they are believed to constitute the most significant fraction of atmospheric emissions from biomass burning, particularly during the dry season [*Seiler and Crutzen*, 1980]. Tropical fires, however, are not necessarily characteristic of fires in other regions of the world.

Interestingly, less field research on trace gas production has centered on fires at mid-latitudes in the world's temperate forests, which are generally believed to be more intense fires. Our efforts have been directed toward the study of mid-latitude fires and, in particular, to trace gas emissions from these fires. Chaparral fires may be particularly interesting because of their typically intense combustion.

The measurements of trace gas emissions from biomass burning reported in this paper were obtained during a prescribed burn on December 12, 1986, conducted by the U.S. Department of Agriculture Forest Service in the Lodi Canyon. The Lodi Canyon is in the San Dimas Experimental Forest, part of the Angeles National Forest in Los Angeles County, California. The climate and ecology of the region are discussed in the following section. The measurements obtained during this burn are important to our understanding of the role of biomass burning on the budgets of trace amtospheric gases, since very little information is available about gaseous emissions from biomass burns from a Mediterranean-type chaparral ecosystem. In addition, the characteristics (total mass, moisture level, nutrient level, etc.) of the living and dead biomass were assessed and inventoried prior to the fire, providing a unique prefire data base.

Smoke plume samples were also obtained on December 3,

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1986, during an attempted burn of the Lodi Canyon that was aborted within minutes after its initiation. Results from these collections are reported as samples 1, 2, and 3 in this text.

Among the various gases produced during biomass burns, several (CO_2, CH_4, and N_2O) are greenhouse gases and impact the climate of our planet [*Ramanathan et al.*, 1985]. Other biomass-burn-produced gases control the chemistry and photochemistry of the troposphere and stratosphere. Carbon monoxide and CH_4 control the chemical destruction of the hydroxyl (OH) radical [*Sze*, 1977; *Levine et al.*, 1985], the most chemically active species in the troposphere, which in turn controls the atmospheric lifetime of almost every tropospheric species [*Levy*, 1972; *Logan et al.*, 1981]. In addition, CH_4, CO, and several other hydrocarbons lead to the photochemical production of ozone (O_3) in the troposphere [*Stith et al.*, 1981; *Delany et al.*, 1985]. In the troposphere, NO is chemically transformed to nitric acid (HNO_3), the fastest growing component of acid precipitation [*Galloway and Likens*, 1981]. Nitrous oxide, chemically inert in the troposphere, diffuses into the stratosphere, where it is chemically transformed into NO and is responsible for about 70% of the annual global destruction of stratospheric ozone via the nitrogen oxide catalytic cycle [*Turco*, 1985].

Interestingly, atmospheric levels of several trace gases (CO_2, CH_4, N_2O, CO, and ethane) produced during the burning of biomass that impact atmospheric chemistry and climate appear to be increasing with time [*Keeling et al.*, 1984; *Rinsland et al.*, 1985; *Rinsland and Levine*, 1985, 1986; *Weiss*, 1981].

LODI CANYON FIRE

The Lodi Canyon, site of the 400-acre prescribed fire, is located within the San Dimas Experimental Forest (34°10'N, 117°47'W) in the San Gabriel Mountains of southern California. It was last swept by wildfire in 1960. It is a semiarid Mediterranean-type chaparral ecosystem. Chaparral is subject to extensive and severe wildfires and is adapted to recurrent fire. Chaparral fires are typically very energetic (intense) fires with fuel consumptions in the range of 2–7 kg/m^2. They are usually propagated through live vegetation, largely consuming foliage and small stems less than 1 cm in diameter (P. J. Riggan, unpublished data, 1987). The Lodi prescribed fire was ignited by a thin stream of gasoline gel spread by helicopter. The fire burned only on the south facing aspects, in predominantly *Ceanothus crassifolius* and *Adenostoma fasciculatum* chaparral, with low rates of fire spread but with typical flame lengths of 6–12 m. Fire intensities have not been resolved at this time. At the time of the fire relative humidity and temperature were 40% and 21°C, respectively. Fuel moisture was about 100% for live fuel and 9% for dry sticks. Slightly more than 80% of the fuel was live. Surface winds were light and variable during the December 12 burn, and discrete smoke plumes dispersed into lenses at 2000 to 2400 m above mean sea level.

SAMPLING EQUIPMENT

Smoke samples were collected from plumes using an air-sampling system aboard a Los Angeles County Fire Department Bell 204B helicopter. The sampling system consisted of three aluminum sampling probes (1.7 cm ID) that protruded about 30 cm forward of the helicopter nose through a modified forward nose access hatch. Flexible Tygon hose (2.0 cm ID) was attached to the base of each probe and run about 4 m to fabricated expansion cones attached to Hi-Vol air pumping/filtration systems. All hardware was attached to a floor pallet and consisted of three air pumping/filtration systems, a power control box, and assorted mounting hardware. One pump/filtration system was used exclusively for pumping particle-filtered burn produced gases into 20-L Tedlar air-sampling bags, and another for collection of smoke particles on Teflon filters. Filters were changed after each complete sampling. A single-throw sliding valve was used to direct the gas from the exhaust sampling line into the Tedlar bags during actual sampling. The exhaust lines from the two pumps converged into one large flexible hose that exited through a modified overhead cargo vent. Sampling flows (velocities) through the probes were chosen to match the planned forward aircraft speed of 40 knots (77 km/h). Rotor downwash was well behind the probes at forward speeds in excess of 30 knots (55 km/h). The Tedlar bags had been tested in our laboratory for the gases reported herein and determined suitable for gas collection/storage for periods of several hours. However, hydrocarbon tests in these bags employed the use of only very simple surrogate mixtures.

SAMPLING SYNOPSIS

Typically, an extensive smoke plume would be visually identified emanating along a canyon ridge or fire line. The identified plume would be primarily associated with either vigorous flaming combustion or mixed combustion with a visually substantial smoldering component. Generally, however, both types of combustion were going on simultaneously at different points along any sampling traverse. A sampling altitude was chosen and a traverse initiated. The pump/filtration systems were turned on seconds before the helicopter penetrated the smoke plume. After entry into the plume (visually determined) the sampling valve was thrown to divert flow into the Tedlar bag. Upon departure from the plume, valving was changed to divert flow back into the exhaust lines and the systems were turned off. About 15 s were required to totally fill a 20-L bag. Traverses through the plume typically involved about 5–7 s, so at least two and sometimes three collections were made through the same region of smoke plume in order to acquire sufficient sample volume for subsequent analyses. Each numbered sampling therefore consisted of an integrated gas sample involving two or three collection passes. Several samples were collected upwind of the canyon at various times during the controlled burn and were used to measure background trace gas composition. Without exception, after two bags were filled, the helicopter returned to our field laboratory, where the samples were immediately transfered (by pump) from the Tedlar bags to electropolished stainless steel sample canisters. In no instance did gas samples remain in the Tedlar bags for more than 30 min before transfer. Chemical analysis was begun immediately after transfer in the on-site laboratory.

CHEMICAL ANALYSIS

Chemical analyses for the trace gases CO_2, CO, hydrogen (H_2), CH_4, total nonmethane hydrocarbons (TNMHC), and N_2O were performed at our field laboratory in the San Dimas Experimental Forest. Except for the CH_4/TNMHC measurements, which required substantial gas volume, all analyses were performed using aliquots from the stainless steel canisters. The hydrocarbon measurements were made by attaching bags directly to the instrument inlet, from which air sample was withdrawn by the automated system. The TNMHC instrument/technique has been described by *Cofer and Purgold* [1981] and *Cofer* [1982], and utilized gas chromatography with flame ionization detection of the CH_4 and hydrocarbons. NMHC data are reported as methane response. CO and H_2 were analyzed using the hot mercury oxide technique, in which CO and H_2 were separated chromatographically, then reduced and detected as mercury vapor [*Cofer et al.*, 1986*a*]. The technique is well established. The precision of our technique was assessed to be about 2%, as performed with calibration gas surrogates at our field laboratory. Analysis for N_2O was performed by electron capture gas chromatography. The technique used for these analyses has been described by *Cofer et al.* [1986*b*]. Analysis for CO_2 was performed using gas chromatography with thermal conductivity detection. A 2-m by 0.32-cm-diameter Porapak N column operated at 60°C was used for separation. Again, a precision of about 2% was obtained with calibration surrogates under field conditions. All gas calibration standards, except the CO_2 standard, which was certified at $\pm 2\%$, were master gravimetric standards certified by Scott Specialty Gases (Plumsteadville, Pennsylvania) at the $\pm 1\%$ level.

A Daedalus DEI-1260 Multispectral Scanner was flown onboard a NASA Ames U2 aircraft 19.8 km above the Lodi Canyon during the burn. The scanner was used in the 0.4- to 12.0-μm spectral range, and transects were flown continuously during the burn. Data were recorded on high-density tape for image processing and analysis. A pixel size of 24 by 24 m was used in this analysis. The scanner data are currently being reduced to yield mean temperatures for each pixel. However, after examination of the uncalibrated thermographic images for the Lodi Canyon fire, we have determined that these digitally enhanced images in their existing qualitative form supply much information about the relationship of our collections to the nature of the fire.

TABLE 1. Sampling Parameters for Lodi Canyon Prescribed Burn Experiment

Sample	Category*	Date (1986)	Time, PST	Altitude, m	Passes
1	B	December 3	1000	215	1
2	B	December 3	1005	215	1
3	M	December 3	1040	215	2
4	F	December 12	1105	60	2
5	B	December 12	1110	60	1
6	M	December 12	1145	35	2
7	F	December 12	1150	90	3
8	B	December 12	1155	90	1
9	F	December 12	1230	90	3
10	M	December 12	1235	670	2
11	B	December 12	1240	670	1
12	M	December 12	1256	310	2
13	M	December 12	1303	90	2
14	F	December 12	1326	90	3
15	F	December 12	1335	60	3
16	B	December 12	1340	90	1
17	M	December 12	1358	370	2
18	M	December 12	1402	90	2
19	B	December 12	1415	90	1

*B indicates background; F, vigorous flaming; M, mixed stages of combustion.

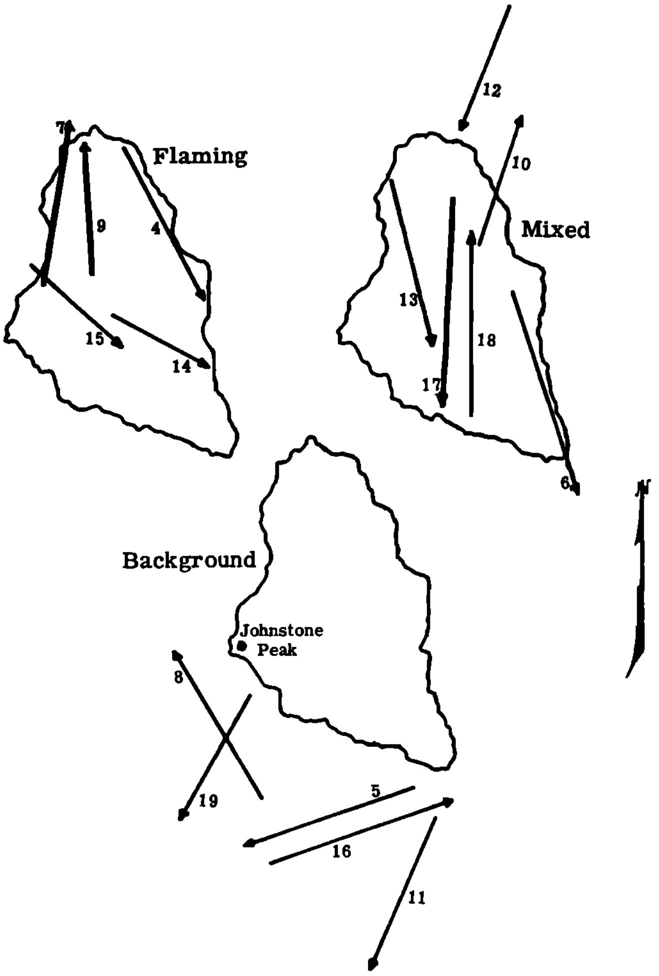

Fig. 1. Approximate flight paths for the helicopter collections over the Lodi Canyon.

RESULTS

Sampling parameters for the helicopter collections during the Lodi Canyon prescribed biomass burn are presented in Table 1. These data, when viewed with the flight paths indicated on Figure 1, provide an accurate description of the helicopter collections. As shown in Table 1, most of the collections were in the vicinity of 60–90 m above ground level, with about an equal number of collections dedicated to vigorous flaming and mixed stages of combustion. Flames frequently rose 6–12 m above the burning chaparral during stages of vigorous combustion. Flames were usually visible in portions of the chaparral fire categorized as mixed; however, the flames appeared much less intense and extensive. Examination of enhanced thermography images recorded nearly coincidental in time with each of the helicopter collections confirmed a reduced but still significant component of intense combustion associated with collections categorized as mixed. This is illustrated in Figure 2 for collections 17 and 18, in which the appropriate helicopter traverses have been superimposed on the thermographic image. Pixels that appear white indicate areas of active combustion. The illustration shown in Figure 2 is representative of all of our mixed combustion stage collections. Incoming air samples (background) were collected and analyzed at regular intervals during the burn and used

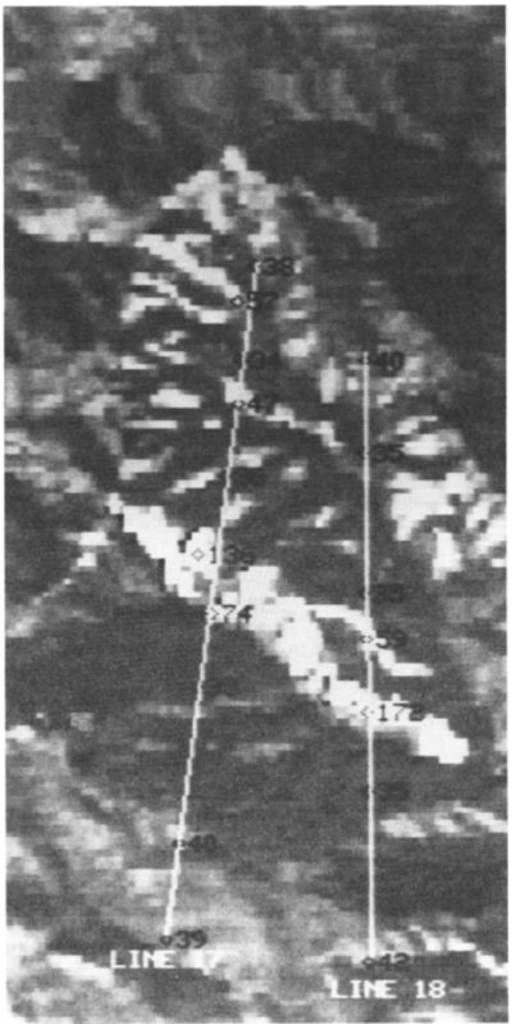

Fig. 2. Enhanced thermography image at 1400 PST.

with plume determinations to determine the concentrations (concentrations above background) for the measured trace gases.

The results of the chemical analyses are grouped by category and presented in Table 2. Mixing ratios determined for the trace gases in the background air are somewhat higher than would be expected for "clean" mid-latitude air. The lowest background trace gas levels determined on December 12 were for sample 11, the highest altitude collection. The fact that the mixing ratios of CO_2 determined in the smoke plumes were significantly above background in the plume measurements is important, since the determination of emission ratios for CO, H_2, CH_4, TNMHC, and N_2O relative to CO_2 ($\Delta X/\Delta CO_2$, vol/vol) was an objective of our experiments. These ratios not only help to assess the emissions overall, but also provide a method for comparison of the $\Delta X/\Delta CO_2$ emission ratios from both flaming and mixed stage combustion.

CO_2 normalized emission ratios ($\Delta X/\Delta CO_2$, where X is equal to each species, vol/vol) obtained from this data set for the measured gases, comparing the mean and standard deviation for vigorous flaming and mixed stage emission categories, are presented in Table 3. Mean concentrations determined from the background trace gas measurements were subtracted from each individual smoke plume measurement to determine emission ratios. Means and standard deviations for flaming and mixed collections of the trace gases were then calculated. The results shown in Table 3 do not indicate any significant differences in mean emission ratios for samples collected over vigorous flaming or mixed stages of combustion.

DISCUSSION

CO_2 normalized emission ratios ($\Delta X/\Delta CO_2$, vol/vol) for trace gas samples collected above the Lodi Canyon prescribed biomass burn are generally lower than the results reported by others involved in large field experiments investigating biomass fires [*Crutzen et al.*, 1979, 1985; *Greenberg et al.*, 1984; *Ward and Hardy*, 1984]. These prior results have assessed CO_2 normalized emission ratios for CO, H_2, CH_4, TNMHC, and N_2O at about 10, 3, 1, 1, and 0.02%, respectively. With the exception of our CO_2 normalized N_2O emission ratios, which are in good agreement with these previously cited results, our emission ratios determined for CO, H_2, CH_4, and TNMHC are all lower, generally by about a factor of 2–3. This result deserves discussion.

The complex relationship between trace gas emissions and fire intensity, fire spread, fuel type, array and size, and fuel moisture content has been well established in the earlier fire literature. For example, the results of *Gerstle and Kemnitz* [1967] indicate that in the very early stages of biomass combustion, CO_2 is emitted in significantly larger proportions than CO and hydrocarbons. Results from *Darley et al.* [1966] describe the dependence of $\Delta CO/\Delta CO_2$ and hydrocarbon ratios upon the ratio of green/dry fuel. CO_2 normalized CO emission ratios ranged from 4.0% for dry native California (San Joaquin Valley) brush to about 14% for green native brush. *Boubel et al.* [1969] report $\Delta CO/\Delta CO_2$ (vol/vol) ratios ranging from 5–20% for grass stubble and straw, again pri-

TABLE 2. Mixing Ratios Determined From Helicopter Sampling of Lodi Canyon Prescribed Burn Experiment

Sample	Category*	CO_2	CO	H_2	CH_4	NMHC	N_2O
1	B	375	0.40	0.80	1.83	0.235	0.314
2	B	370	0.40	0.75	1.81	0.185	0.317
5	B	370	0.60	0.80	1.83	0.235	0.316
8	B	375	0.70	0.85	1.82	0.215	0.318
11	B	365	0.40	0.60	1.82	0.145	0.317
16	B	370	0.80	0.85	1.84	0.170	0.320
19	B	375	0.75	0.95	1.83	0.220	0.317
4	F	570	12.6	5.40	2.44	0.887	0.337
7	F	690	14.3	7.20	2.84	1.336	0.359
9	F	545	10.8	5.60	2.48	0.946	0.345
14	F	600	12.0	4.90	2.63	1.028	0.353
15	F	570	9.4	4.30	2.75	1.141	0.346
3	M	510	4.10	2.40	2.35	0.930	0.331
6	M	420	3.00	1.90	2.11	0.415	0.330
10	M	395	2.80	1.80	1.93	0.295	0.325
12	M	405	4.10	1.75	1.98	0.345	0.328
13	M	460	2.95	2.00	2.07	0.440	0.324
17	M	410	3.85	1.75	2.05	0.361	0.330
18	M	425	3.00	1.80	2.06	0.420	0.321

Units are parts per million by volume.

*B indicates background; F, vigorous flaming; M, mixed stages of combustion.

marily depending upon moisture content. *Ward and Hardy* [1984] report $\Delta X/\Delta CO_2$ ratios for CO and CH_4 ranging from 5 to 11% and from 0.3 to 0.7%, respectively, for samplings from actively flaming prescribed slash fires in the Pacific Northwest. The complex dependency of emission ratios on so many factors, as indicated in these citations, can lead to a high degree of variability in individual measurements and data sets. That our determinations indicate notably lower CO_2 normalized emission ratios is consistent with several characteristics of the Lodi Canyon chaparral fire. Combustion during this fire was dominated by foliage and limited to small-diameter stem material. This produced a fire with a large surface-to-volume fuel ratio. Although the fire had low rates of spread, the chaparral that was burning burned intensely, producing 6- to 12-m-high flame lengths. These conditions should favor efficient combustion and lead to low CO_2 normalized emission ratios for CO, CH_4, H_2, and NMHC.

That our N_2O emission ratios are in relatively good agreement with previous studies may indicate that N_2O production in biomass burning is less dependent on fire intensity than are the other gasses. *Dash* [1982] reported results from wood-burning fireplaces that suggested that the emission ratio ($\Delta NO_x/\Delta CO_2$) did not change much during flaming and smoldering combustion and may have depended largely upon nitrogen content in the fuel. However, *Ceanothus crassifolius* stands in the San Dimas Experimental Forest have a high nitrogen content relative to many other biomass fuels (see Table 4.). The higher nitrogen content of our fuel may have masked any potential disagreement in our N_2O emission ratios with other previously reported determinations.

Our CO_2 normalized emission ratios for samples primarily collected over intensely burning (vigorous flames) chaparral are indistinguishable from those made over mixed stages of combustion. While our data for CO and CH_4 emission ratios are within the range reported by *Ward and Hardy* [1984] for samples from actively burning fires, results from our mixed collections do not indicate any trend toward increases in $\Delta X/\Delta CO_2$ ratios for the reported trace gases. Our emission ratios would be anticipated to increase as proportionally more gaseous emissions from the less efficient transitional and smoldering combustion stages were incorporated into the smoke plumes.

Calculations of emission ratios using the results of *Ward and Hardy* [1984] over smoldering fires range from 13- to 29% and 1.0 to 2.5% for CO and CH_4, respectively. These emission ratios, however, resulted from burns of logging slash, which might be expected to contain lower relative amounts of high surface-to-volume fuel and thus potentially to yield a less efficient combustion and higher CO_2 normalized emission ratios. D. E. Ward (personal communication, 1987) has indicated that his emission ratios for CO determined over flaming combustion during a September 1986 test burn at the Lodi Canyon are in relative agreement with ours.

TABLE 3. The $\Delta X/\Delta CO_2$ for All Samples, Vigorous Flaming Combustion, and Mixed Stages of Combustion

Gas	All	Flaming	Mixed
CO	0.056 ± 0.024	0.051 ± 0.008	0.060 ± 0.031
H_2	0.022 ± 0.008	0.021 ± 0.004	0.023 ± 0.010
CH_4	0.0041 ± 0.0009	0.0036 ± 0.0006	0.0043 ± 0.0010
TNMHC	0.0040 ± 0.0007	0.0039 ± 0.0006	0.0041 ± 0.0008
N_2O	0.00018 ± 0.00010	0.00014 ± 0.00002	0.00021 ± 0.00012

TABLE 4. Above-Ground Nitrogen Content of Several Biomass Fuels

Vegetation	Biomass	Foliage	Reference
Ceanothus crassifolius	355	130	P. J. Riggan (unpublished data, 1987)
Douglas fir	294	65	*Cole and Rapp* [1981]
Red alder	240	100	*Cole and Rapp* [1981]

Values are given in kilograms of nitrogen per hectare.

Since the digitally enhanced thermograpic images indicated that our mixed collections should be considered as mixtures of flaming, transitional, and smoldering combustion, we offer an interpretation of these results. Since much larger volumes of gas per unit of area are released during intense stages of combustion, emissions from the flaming combustion might well have dominated the collections from the mixed smoke plumes, even though visual evidence suggested the less intense combustion (smoldering) to be more extensive. Additionally, the large surface-to-volume ratios of the chaparral fuel would tend to decrease the less efficient smoldering-type emissions.

CONCLUSIONS

Samples of trace gases collected during a December 12, 1986, prescribed burn of the Lodi Canyon yielded CO_2 normalized emission ratios for CO, H_2, CH_4, and TNMHC about a factor of 2 lower than the bulk of emission ratios reported in the literature for these gases. The lower emission ratios determined in this study quite likely resulted from efficient combustion typical of intense fires with high surface-to-volume fuel ratios. Emission ratios determined for N_2O, however, were at levels comparable to those determined in other studies. That N_2O emission ratios were in good agreement with N_2O emission ratios in other studies may indicate less dependency of N_2O emissions on fire intensity or may reflect the higher nitrogen contents in the chaparral ecosystem. No significant differences in emission ratios were determined between samples collected over vigorously flaming and over mixed stages of combustion. The absence of any discernable differences probably reflects both the nature of the combustion of a chaparral fire (small amout of fuel consumed during the smoldering stages) and the fact that our mixed collections contained emissions from flaming combustion. Flaming combustion would be expected to inject disproportionately voluminous emissions per unit area undergoing combustion and thereby dominate composition of mixed stage smoke plumes. Future work will attempt to better isolate emissions from the different stages of combustion and to assess their contribution to the total emission burden.

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