JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 92, NO. D12, PAGES 14,701-14,709, DECEMBER 20, 1987

NITROGEN AND SULFUR EMISSIONS FROM THE BURNING OF FOREST PRODUCTS NEAR LARGE URBAN AREAS

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Abstract. Airborne measurements of trace gases and particles in the smoke from a prescribed burn of forest products in the Los Angeles basin show significantly higher emissions of NO_x, SO_2, and particulate NO_3^- than do measurements in smokes from the burning of biomass in rural areas. It is postulated that the high emissions are due to the revolatilization of previously deposited pollutants. Implications for pollutant source inventories and the nuclear winter hypothesis are briefly discussed.

Introduction

Studies of the emissions of particles and gases from forest fires and prescribed burns of forest products were initially carried out to determine their effects on air quality [e.g., Radke et al., 1978, 1983; Sandberg et al., 1979]. Recently, interest in such studies has broadened to include the effects of burning on global chemistry [e.g., Crutzen et al., 1985] and on the environmental consequences of nuclear war, the so-called "nuclear winter hypothesis" [Crutzen and Birks, 1982; Turco et al., 1983].

This paper is concerned with a preliminary description of airborne measurements obtained in the emissions from a prescribed burn of forest products that took place near Los Angeles on December 12, 1986. Comparison of these measurements with those obtained from burns in more rural areas reveals significant differences in nitrogen and sulfur emissions.

Description of the Burns

The prescribed burn near Los Angeles, California, was designed to characterize gaseous and particulate emissions from a relatively large chaparral fire and to relate the emissions to remotely sensed fire radiative emissions, heating, and smoke optical properties. Located in Lodi Canyon (34°10'N, 117°47'W), on the San Dimas Experimental Forest, the prescribed burn encompassed ~40 hectares (0.4 km^2) of chaparral that was last burned by wildfire in 1960. The burn consumed 3-6 kg m^{-2} of foliage, deadwood, and small-diameter live stems of Ceanothus crassifolius and Adenostoma fasciculatum (chamise). Hereafter, we will refer to this as the Lodi burn.

The Lodi burn occurred on December 12, 1986 (Figure 1). It was ignited by flaming gasoline-gel globules dispensed from a helicopter. (We note that this fuel contains only insignificant levels of either sulfur or nitrogen.) Flames were commonly 6-12 m in length, with low rates of forward spread. Soil surface temperatures during burning peaked at about 850°C (F. Weirich, personal communication, 1986). The burn produced a series of discreet plumes, which tended to spread out horizontally at 2000-2400 m above mean sea level (MSL), with some hotter columns penetrating above these levels.

We will also present some airborne measurements obtained in the smoke from a prescribed burn ignited on December 3, 1986 near Ramona, California (33°3'30"N, 116°55'30"W). This burn involved a 30-hectare (0.3 km^2) subunit in which 2 kg m^{-2} of fuel was consumed, consisting of coastal sage scrub dominated by black sage (Salvia mellifera), sumac (Rhus laurina), and chamise (Figure 2). We will call this the Eagle burn.

Airborne Instrumentation and Sampling Procedures

The measurements to be described here were obtained aboard the University of Washington's Convair C-131A research aircraft (Table 1). Smoke samples were obtained by allowing ram-air pressure to fill a polyethylene bag, 1.5 m^3 in volume, aboard the aircraft. The various measurements listed in Table 1 were obtained by drawing samples from this bag. The bag was filled in 16 s when the aircraft was located near the center of the main convective plume from the fire. (The center of the plume was considered to be located where the particle number concentration and light-scattering coefficient reached peak values. Both of these parameters were measured continuously aboard the aircraft.)

Results

Chemical measurements obtained in the smokes from the Lodi and Eagle burns are listed in Table 2. For comparison, we list similar measurements in Table 3, obtained in the Pacific northwest. The concentrations of NO_x and SO_2 in the emissions from the Lodi burn, and to a lesser extent the NO_x from the Eagle burn, are much higher than those measured in the smokes from the burns in the much more rural areas of the Pacific northwest. Also, the NH_3 measured in the plume from the Lodi burn is unusually high.

The dichotomy between the measurements in southern California and the Pacific northwest is clearly shown in Figures 3-5; the ratios NO_x/CO, particulate NO_3^-/CO, and SO_2/CO in the smoke from the Lodi burn (and NO_x/CO in the smoke from the Eagle burn) were all much greater than they were in the smokes from the burns in the Pacific northwest. However, this dichotomy does not appear in the ratio particulate $SO_4^=$/CO (Figure 6).

Near the middle and upper part of the plume of smoke from the Lodi burn, the ozone concentrations were higher than in the ambient air, while near the base of the plume, the ozone concentrations were lower than ambient. Ozone deficits in the smokes from fires are to be expected and have been observed previously [e.g., Stith et al., 1981]; they are due to both heterogeneous and homogeneous reactions of ambient ozone with plume constituents. Ozone production in fire plumes has also been observed previously [Evans et al., 1974; Radke et al., 1978; Stith et al., 1981]; it may be due to photochemical

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Paper number 7DO799.
0148-0227/87/007D-0799$05.00

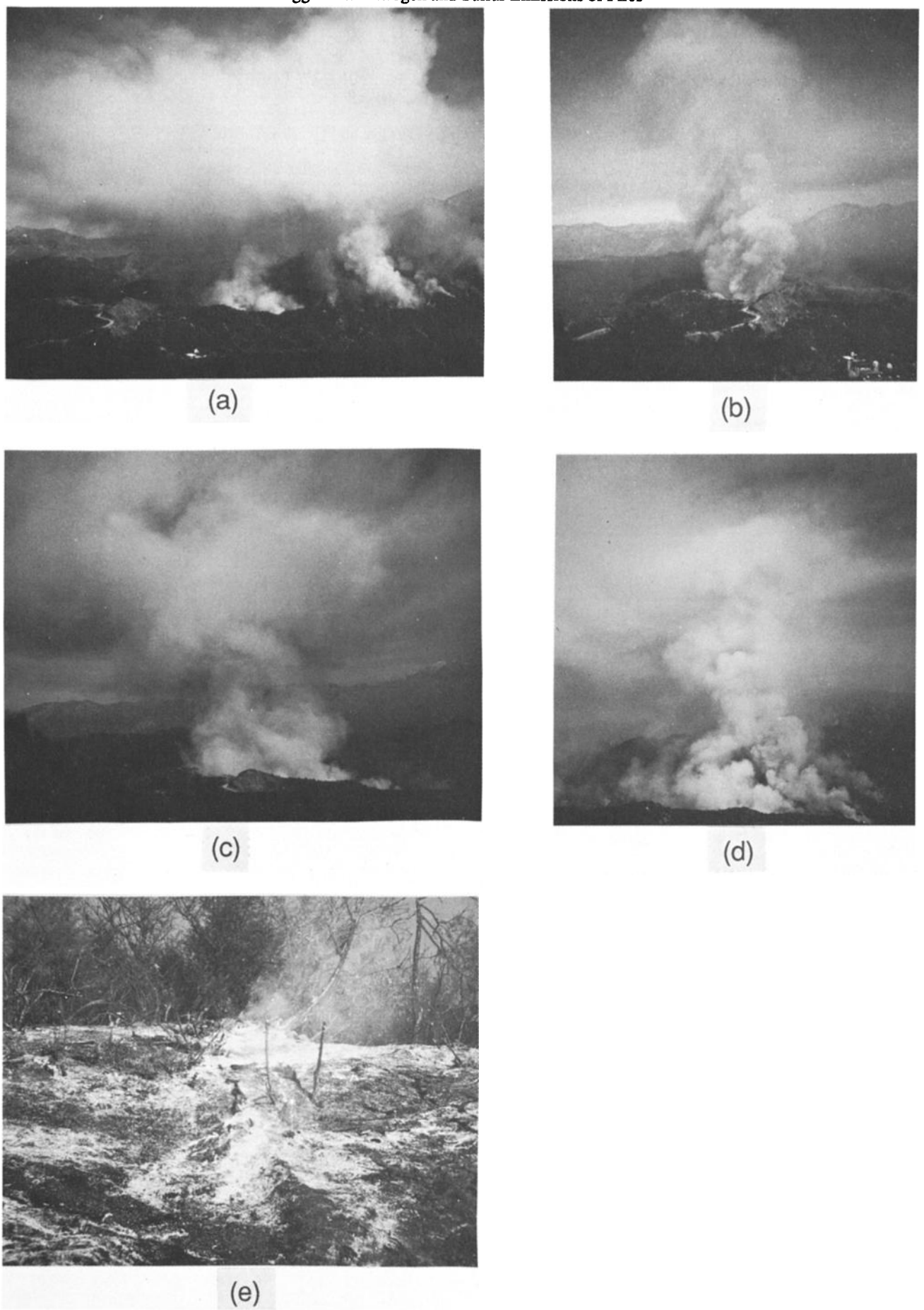

Fig. 1. Views of the Lodi burn at (a) 1106 PST (1 hour 9 min after ignition), (b) 1154 PST, (c) 1210 PST, (d) 1228 PST, and (e) 1322 PST on December 12, 1986. (Photographer: Peter V. Hobbs)

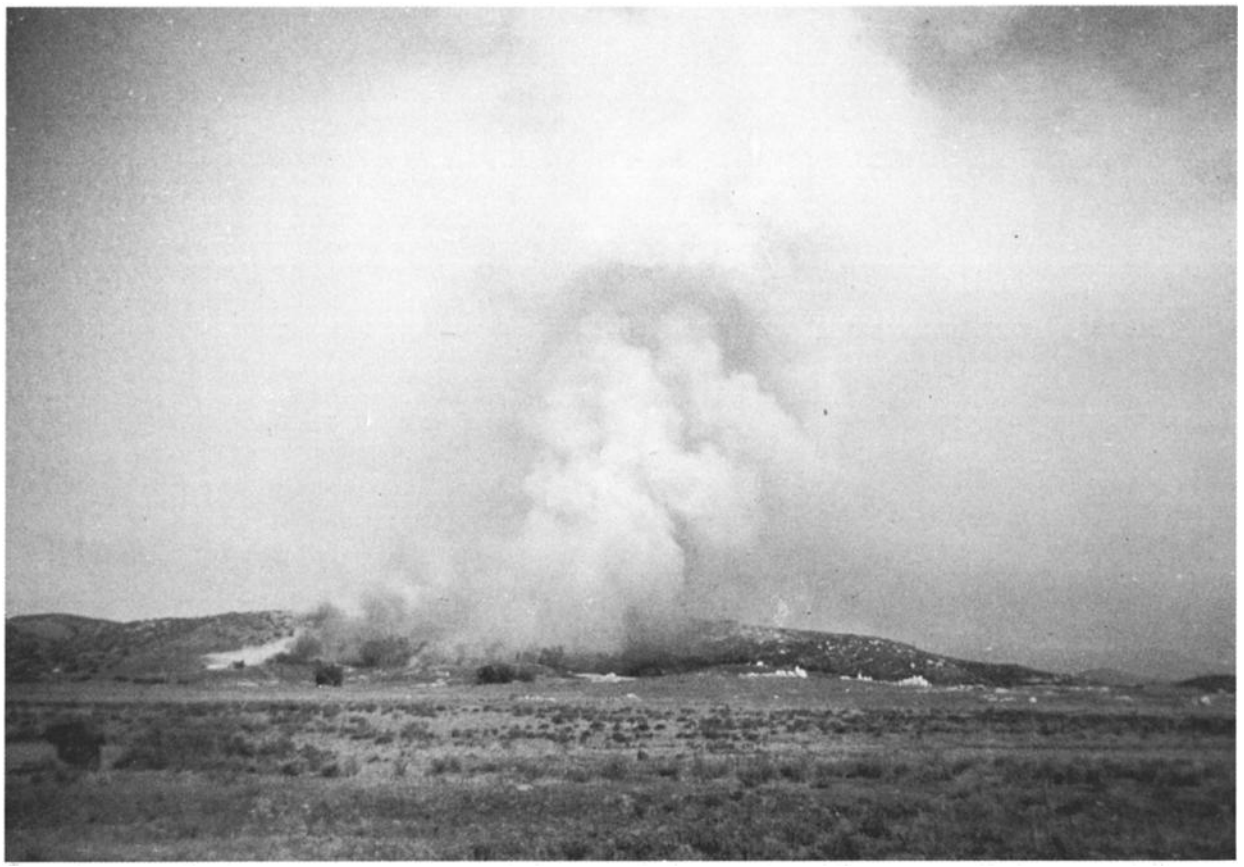

Fig. 2. View of the Eagle burn at 1200 PST on December 3, 1986.

TABLE 1. Measurements Obtained Aboard the C-131A Aircraft That are Utilized in the Present Analysis

Parameter	Instrument or Technique	Analytical Uncertainty
Soluble particulate anions	filtration with 47-mm stretched Teflon filter (Gelman)	$SO_4^=$ ± 4% or better; NO_3^- ± 11% or better; Cl^- ± 14% or better
O_3	Monitor Labs 8410 (C_2H_4 chemiluminescent)	±5 ppb
NO_2, NO	Monitor Labs 8840* (O_3 chemiluminescent)	±1 ppb
SO_2	Teco SP 43* (pulsed fluorescence)	±1 ppb
CO	Teco 48 (correlation IR spectrometer)	±100 ppb
CO_2	Miran 1A (IR spectrometer)	±4 ppm
NH_3	filtration with oxalic acid-impregnated filter preceded by Teflon filter	±15% or better

*Modified in house.

TABLE 2. Average Concentrations of Various Chemicals Across the Centers

Geographical Location	Fuel	Approximate Area Burnt, km^2	Location of Measurements With Respect to Fire	$[Cl^-]$, $\mu g\ m^{-3}$
San Dimas Experimental Forest, located about 10 km north of Pomana, near Los Angeles, California. ("Lodi" burn).	standing chaparral and brush (prescribed burn ignited with jellied gasoline by helicopter)	0.4	main convective plume‡	BDL
			main convective plume‡	1.23
			main convective plume‡	9.01
			main convective plume‡	BDL
			main convective plume‡	9.70
			main convective plume‡	7.75
			6.5 km downwind	BDL
			6.5 km downwind	BDL
			main convective plume‡	2.3
			main convective plume‡	3.10
			~8 km downwind	1.84
			~9.5 km downwind	BDL
			~10.5 km downwind	BDL
			~10.5 km downwind	BDL
			edge of the main convective plume	5.81
			edge of the main convective plume	7.19
			main convective plume‡	6.11
			main convective plume‡	BDL
			main convective plume‡	NM
			main convective plume‡	NM
			~10.5 km downwind	NM
			~10.5 km downwind	NM
			ambient air above the surface inversion layer	BDL
				BDL
			same as above	BDL
			main convective plume‡	1.08
			main convective plume‡	1.29
About 2.5 km east of Ramona and ~40 km NE of San Diego, California ("Eagle" burn).	coastal sage scrub	0.3	main convective plume‡	BDL
			main convective plume‡	BDL
			main convective plume‡	BDL
			main convective plume‡	BDL
			main convective plume‡	BDL
			ambient air above the surface inversion layer	NM

BDL and NM indicate "below detection limit" and "no measurement", respectively. Approximation sign indicates that measurements could be as much as a factor of 2 in error.

* Parameters to the left of the brackets in this column were measured simultaneously by exposing two filters (both values listed) while parameters to the right of the brackets are average values measured over the time interval for which the two filters were exposed.

† Of the NO_x, 70% or more is NO_2.

‡ Measurements averaged over a distance of ~1.5 km across center of plume.

reactions near the top of a plume as it mixes with the ambient air [Evans et al., 1977].

Discussion

Several possible explanations can be suggested for the differences in the emissions from the burns in southern California and the Pacific northwest. Differences in the composition of the biomass undergoing combustion might be responsible. In the Lodi burn, which produced high ratios of NO_x/CO, particulate NO_3^-/CO and SO_2/CO as well as NH_3, the biomass was primarily Ceanothus crassifolius and Adenostoma fasciculatum (chamise). Ceanothus supports active nitrogen fixation in root nodules, and the standing vegetation should contain substantially more fixed nitrogen than the biomasses consumed in either the Eagle burn (Salvia mellifera, Rhus laurina (sumac and chamise)) or the Pacific northwest burns (primarily P. menziesii (Douglas fir)). However, this would leave unexplained both the high NO_x/CO ratio in the Eagle burn and the high SO_2/CO ratio in the Lodi burn compared to those in the Pacific northwest burns.

An alternative, and we believe more likely, explanation for the differences in the emissions between the burns in southern California and Pacific northwest lies in differences in the deposition of pollutants to the vegetation. Lodi Canyon is located just 10 km north of the San Bernadino Freeway (Interstate 10) and ~50 km to the east of the center of Los Angeles. Nitrate deposition in this area is the highest in the United States, and it exceeds that in typical watersheds in the Pacific northwest by a factor of 4 or more [Riggan et al., 1985]. Furthermore, while direct depositional measurements are scarce, on the basis of the relatively high levels of NO_x and SO_2 found in the Los Angeles basin [Hoggan et al., 1980], and the well established direct proportionality between dry

of Smoke Plumes From the Burning of Forest Products in Southern California

$[NO_3^-]$, $\mu g\ m^{-3}$	$[SO_4^=]^*$ $\mu g\ m^{-3}$	$[O_3]$, ppbv	[CO], ppbv	$[CO_2]$, ppmv	$[NO_x]^\dagger$, ppbv	$[SO_2]$, ppbv	$[NH_3]$, ppbv
9.17 7.03	4.47 5.54	85	1212	375	NM	3	NM
26.9 34.1	26.1 30.6	108	4644	442	>50	16	NM
59.4 54.0	26.1 29.1	106	5135	448	>100	18	NM
10.7 19.0	4.18 4.83	185	1026	370	53	2	NM
21.4 24.2	11.8 13.2	NM	2475	NM	NM	NM	NM
41.0	13.3	200	1746	388	108	5	NM
18.7	7.55	178	1437	379	189	4	NM
16.0 15.7	4.71 4.98	182	2330	387	NM	2	NM
15.3 16.6	13.5 16.1	62	3298	413	353	6	NM
21.2	20.7	17	5125	429	359	5	NM
18.7	12.9	22	3402	408	343	6	127
NM	NM	82	2730	417	166	6	NM
NM	NM	99	2220	397	107	BDL	NM
NM	NM	143	1840	377	161	2	NM
NM	NM	141	1853	378	156	1	NM
BDL BDL	0.49 0.48	88	345	356	19	BDL	NM
0.83	0.20	128	126	361	NM	BDL	~6
5.23 5.85	3.79 4.26	103	422	365	NM	2	NM
7.44	4.75	111	1057	375	143	4	NM
4.03 5.71	3.21 2.99	108	478	396	NM	3	NM
4.54 6.14	1.89 2.34	123	529	411	NM	4	NM
NM	NM	71	83	359	~6	2	NM

deposition (the dominant deposition mechanism for NO_x and SO_2) and air concentration, it is quite likely that deposition of SO_2 and NO_x are also anomalously high. We postulate that during burning the nitrogen and sulfur species that have been previously deposited on the vegetation are volatilized, resulting in these species being resuspended in the atmosphere. Pollutant, and in particular nitrate, deposition in the region of the Eagle burn is probably not as great as in the Los Angeles area, but, with the exception of one case (see next paragraph), it probably significantly exceeds that at the rural locations in the Pacific northwest for which emissions from burns are listed in Table 3.

The interesting exception in the Pacific northwest are the measurements obtained at the location listed under number 4 in Table 3. These measurements were obtained in the emissions from a burn located ~25 km downwind of a 1000-MW coal-fired electric power plant. This plant is a major source of odd nitrogen emissions [Hegg and Hobbs, 1980], and it undoubtedly produces high deposition of these materials downwind. Note that the NO_x measured in the emissions from this burn (250 parts per billion by volume (ppb)) are comparable to the highest measured in the emissions from the Lodi burn. Although CO measurements were not made in this Pacific northwest burn, any reasonable assumption would yield an NO_x/CO emission ratio comparable to those measured in the Lodi burn and much higher than those measured in the emissions for the other Pacific northwest burns. This observation provides further support for the importance of prior atmospheric deposition in determining the emissions of nitrates from burning biomass.

Turning to SO_2 emissions, Figure 5 shows that the SO_2/CO ratio is also much higher in the Lodi burn than it is in the Pacific northwest. We also attribute this to the resuspension of previously deposited pollutants, since there are far more

TABLE 3. Average Concentrations of Various Chemicals Across the

Geographical Location	Fuel	Approximate Area Burnt, km^2	Location of Measurements With Respect to Fire	[Cl^-], $\mu g\ m^{-3}$	[NO_3^-], $\mu g\ m^{-3}$
Near Montesano, ~55 km west of	conifer slash (prescribed burn)	?	main convective plume‡	6.17	19.9
			main convective plume‡	6.27	20.9
			main convective plume‡	2.90	10.2
			main convective plume‡	3.43	10.46
			main convective plume‡	1.59	11.2
			main convective plume‡	1.60	11.0
~50 km southeast of Olympia, Washington.	~5 x 10^6 kg of moist conifer slash (prescribed burn ignited by diesel oil and gasoline and hand lit)	1.13	~1.6 km downwind§	NM	NM
~50 km south of Eugene, Oregon.	conifer slash (prescribed burn)	?	near plume center§	NM	NM
~60 km south of Tacoma, Washington, and ~25 km ENE of the Centralia coal-fired power plant.	~4.6 x 10^6 kg of of conifer slash (prescribed burn ignited by diesel oil and gasoline)	0.35	~13 km downwind§	NM	NM

BDL and NM indicate "below detection limit" and "not measured" respectively. Approximation sign indicates measurement could be as much as a factor of 2 in error.

* Parameters to the left of the brackets in this column were measured simultaneously be exposing two filters (both values are listed) while parameters to the right of the brackets are average values measured over the time interval for which the filters were exposed.

† Of the NO_x, 70% or more is NO_2.

‡ Measurements averaged over a distance of 1.5 km across center of plume. The three sets of values listed were obtained at different times and locations in the plume.

§ Measurements averaged over a distance of 300 m across center of plume.

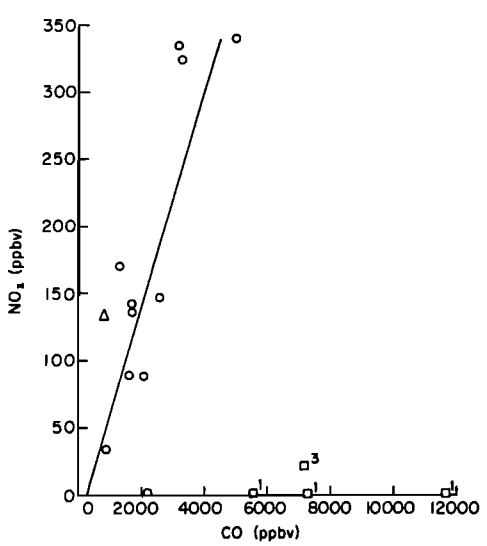

Fig. 3. NO_x versus CO concentrations in smokes from forest fires: the Lodi burn near Los Angeles (circles), the Eagle burn near San Diego (open triangle), and burns in the Pacific northwest (squares; numbers refer to data sets as listed in Table 3). Ambient values have been subtracted for the Lodi and Eagle measurements. The line is a best fit to the Lodi measurements. Analytical uncertainties are listed in Table 1.

sources of SO_2 in the Los Angeles basin than in the Pacific northwest. And, as mentioned previously, air concentrations in the Los Angeles basin are in fact much higher than in the Pacific northwest. Also, it is likely that much of the SO_2

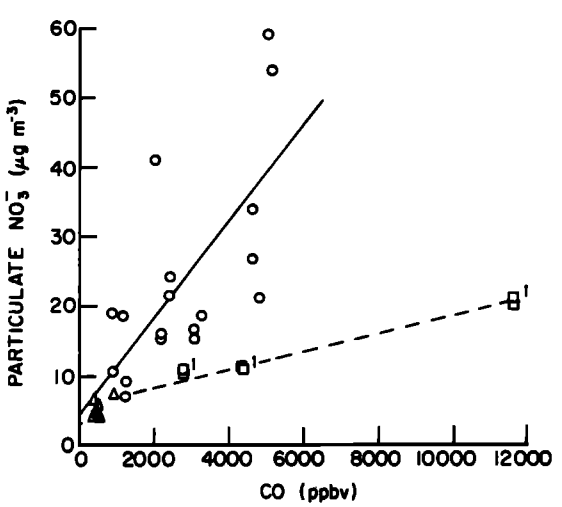

Fig. 4. As for Figure 3, but for particulate NO_3^- versus CO. The dashed line is a best fit to the data from the Pacific northwest.

Centers of Smoke Plumes From the Burning of Forest Products in the Pacific Northwest

$[SO_4^{=}]^*$, $\mu g\ m^{-3}$	$[O_3]$, ppbv	[CO], ppbv	$[CO_2]$, ppbv	$[NO_x]^\dagger$ ppbv	$[SO_2]$, ppbv	Reference
62.3 64.5	~8	11,800	491	~1	~1	this paper
24.4 26.0	13	2,885	43	~1	~2	this paper
15.3 16.8	~3	4,489	423	~1	BDL	this paper
NM	140	NM	~500	20	≤ 2	Stith et al. [1981]; (CO_2 information from J. Stith (personal communication, 1986)
8.7	~40	7,250	367	27	BDL	Radke et al. [1983]
NM	~100	NM	NM	250	BDL	Radke et al. [1978]

emissions in the Pacific northwest are oxidized to sulfate either before or after deposition, this being the normal fate of SO_2. However, in the Los Angeles basin, a considerable amount of S(IV) is stabilized in organic and inorganic adducts, even in the aqueous phase, as a result of the abnormally high concentrations of suitable reactants, such as aldehydes [Eatough and Hanson, 1983; Richards et al., 1983]; sulfur from such species will likely volatilize as SO_2.

The similarity in the particulate $SO_4^{=}/CO$ ratios in the emissions from the burns in southern California and the Pacific northwest (Figure 6) is to be expected, since sea salt is a major source of sulfate deposition in both regions [Liljestrand and Morgan, 1981; Vong and Waggoner, 1983]. Indeed, estimates of sulfate deposition derived from the studies of Roberts [1975] and Liljestrand and Morgan [1981] for the Los Angeles basin are quite similar to sulfate deposition values derived from Vong and Waggoner [1983] for the Pacific northwest.

Implications

Whether the variable and unexpectedly high emissions of nitrogen and sulfur species presented here are due primarily to differences in the composition of the biomass or, more likely, to different exposures to pollutants, they have implications for both air pollution and the nuclear winter hypothesis.

As far as air pollution is concerned, the results presented here indicate that wild fires or prescribed burns of biomass, particularly those near major urban areas, may be major sources of odd nitrogen and S(IV). The source strengths of such fires for odd nitrogen may be similar to or larger than that of a coal-fired power plant.

Model simulations indicate that the particles that would be emitted into the atmosphere by the widespread fires that would accompany a nuclear war could cause significant decreases in global surface temperatures [Crutzen and Birks, 1982; Turco et al., 1983; National Research Council (NRC), 1985; Pittock et al., 1986]. Gaseous emissions from such fires will also perturb the chemistry and physics of the atmosphere. The measurements presented in this paper suggest that the NO_x (and possibly, the NO_3^- and $SO_4^{=}$) emission factors that have been used in numerical model simulations of the nuclear winter scenario may have been too small. For example, in the NRC and SCOPE [Pittock et al., 1986] reports on the nuclear winter hypothesis, the molar ratio of NO_x to CO_2 in smokes from forest fires was taken as 3×10^{-3} and 2×10^{-3}, respectively. These values were based on measurements in the laboratory [Patterson and McMahon, 1984] and in smokes from forest fires in the Amazon basin [Crutzen et al., 1985], neither of which would include effects due to the remobilization of deposited pollutants from urban sources. Crutzen and Birks [1982] presumably used a similar ratio of NO_x to CO_2 in their original paper on nuclear winter, although this is not explicitly stated in the text. Turco et al. [1983] did not include NO_x emissions from forest fires in their model simulations of a nuclear winter (although they did include NO_x in the stratosphere from the nuclear "fireball").

Shown in Table 4 are average values for the molar ratios of NO_x to CO_2 and NO_x to CO derived from the measurements presented here and from values that have been measured or

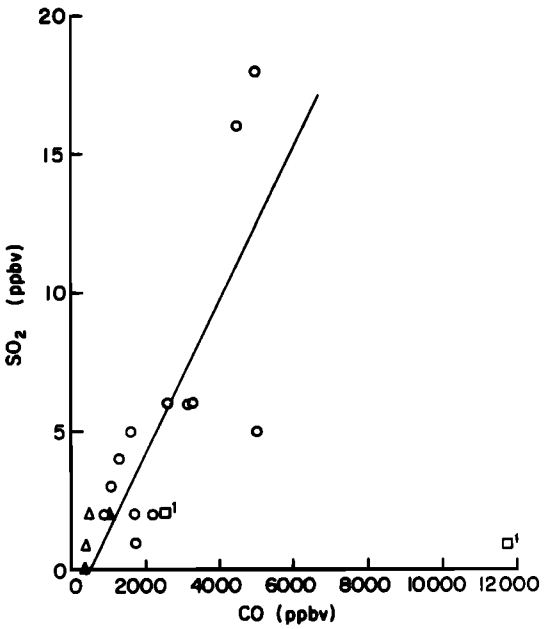

Fig. 5. As for Figure 3, but for SO_2 versus CO.

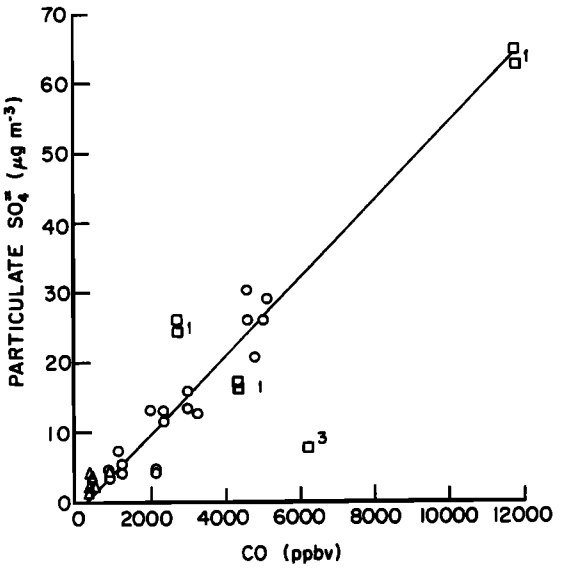

Fig. 6. As for Figure 3, but for particulate $SO_4^=$ versus CO. The line is a best fit to both the southern California and Pacific northwest data.

TABLE 4. Averages and Standard Deviations for the Ratio of NO_x to CO and NO_x to CO_2 in Smokes Fires

Location of Fire	NO_x/CO	NO_x/CO_2	Comments
Near Los Angeles (Lodi Burn)	0.08 ± 0.03	0.006 ± 0.003	measurements presented in this paper (If the NO_x/CO_2 ratio is calculated from our measured NO_x/CO ratio using Pittock et al.'s [1986] value for CO/CO_2 (=0.13), a value of 0.011 is obtained for NO_x/CO_2*. Our measurements give a ratio of CO/CO_2 of 0.072 ± 0.018.)
Near San Diego (Eagle Burn)	0.14	0.013	measurements presented in this paper
Near Montesano, ~55 km west of Olympia, Washington	$<10^{-4}$	$<10^{-5}$	measurements presented in this paper
~50 km southeast of Olympia, Washington	...	0.0001	Radke et al. [1983].
~50 km south of Eugene, Oregon	0.003	0.0001	Radke et al. [1983]
Several different locations in the United States	0.02 -0.1	0.003 - 0.017	NO_x/CO_2 ratio is calculated from Sandberg et al.'s [1979]measurements of NO_x/CO, using Pittock et al.'s [1986] CO/CO_2 ratio.
Nuclear winter numerical model simulations	0 -0.023	0-0.003	The NO_x/CO_2 values cover the range used by Crutzen and Birks [1983], Turco et al. [1983], National Research Council [1985], Thompson and Schneider [1986], Vupputuri [1986], and Pittock et al. [1986]. The NO_x/CO values were calculated from the NO_x/CO_2 values using Pittock et al.'s [1986] value for CO/CO_2.

Ambient concentrations of the gases have been subtracted.

* This method is often used to determine NO_x/CO_2. We measured both NO_x/CO and NO_x/CO_2; the former is more accurate because of the large contrast between CO concentrations in smoke and ambient air compared to CO_2.

used by other workers. The ratios NO_x/CO_2 and, in particular, NO_x/CO that we measured in the Lodi and the Eagle burns are more than an order of magnitude larger than the values for these ratios that we have measured in more rural locations, and 2-4 times larger than the largest values that have been used in numerical model simulations of the nuclear winter scenario. In the compilation of emission ratios by Sandberg et al. [1979], values comparable to those we measured in the Lodi and Eagle burns can be found, although most of their values are lower than ours.

What weight should be given in numerical modeling studies of the nuclear winter hypothesis to the high values of NO_x/CO that we have measured in smokes from fires near urban areas in southern California? Certainly, this region cannot be considered as representative of forested areas in general. However, for reasons outlined in the following paragraph, the NO_x/CO values we measured in southern California may be more representative for numerical modeling studies of nuclear winter than those for rural areas.

First, pollutant deposition is commonplace near large urban areas. Indeed, nitrogen deposition in western European woodlands can be as much as 3 times greater than that in the Los Angeles basin [van Breeman et al., 1982]. Second, in a nuclear war, urban areas could be preferential targets. Finally, it is important to note that we do not know what fraction of the remobilized pollution is derived from the burning of the forest products themselves (i.e., plant materials) and what fraction is derived from the heating and burning of the forest floor. If a significant fraction originated from surface materials and soils, then remobilized pollution will probably increase the emission factors from the burning of other materials subject to deposition and accumulation of urban pollutants. This overall increase in NO_x emissions from combustion of ground material could render such NO_x emission comparable, or possibly even greater than, those produced in nuclear fireballs.

In view of these considerations, we suggest that the NO_x emission factors that we have measured in southern California may be more representative than those that have been used to date in numerical modeling studies of the nuclear winter scenario.

Acknowledgments. Thanks are due to all of those, too numerous to mention by name, who participated in the Lodi Canyon fire experiment. Support and management of fire operations for the Lodi burn by the Angeles National Forest, County of Los Angeles Fire Department and the California Department of Forestry are gratefully acknowledged. This research was supported by the Defense Nuclear Agency, through funds made available under NRL contract N00014-86-C-2246.

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(Received June 10, 1987;
revised September 1, 1987;
accepted October 12, 1987.)