Observations of Ozone Formation in Power Plant Plumes and Implications for Ozone Control Strategies

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Data taken in aircraft transects of emissions plumes from rural U.S. coal-fired power plants were used to confirm and quantify the nonlinear dependence of tropospheric ozone formation on plume NO_x (NO plus NO_2) concentration, which is determined by plant NO_x emission rate and atmospheric dispersion. The ambient availability of reactive volatile organic compounds, principally biogenic isoprene, was also found to modulate ozone production rate and yield in these rural plumes. Differences of a factor of 2 or greater in plume ozone formation rates and yields as a function of NO_x and volatile organic compound concentrations were consistently observed. These large differences suggest that consideration of power plant NO_x emission rates and geographic locations in current and future U.S. ozone control strategies could substantially enhance the efficacy of NO_x reductions from these sources.

Ozone is the most abundant tropospheric oxidant and an important component of photochemical pollution. Elevated concentrations of near-surface ozone that principally occur during the summer months have been shown to be harmful to human health and damaging to vegetation (*1*). Regulation of ozone precursor emissions under the U.S. Clean Air Act of 1970 and its subsequent amendments has been partially successful in reducing human exposure, but many areas of the country are still subject to episodes of high ambient ozone levels (*2*).

Tropospheric ozone is formed by photochemical reactions involving volatile organic compounds (VOCs) and the oxides of nitrogen NO and NO_2; a simplified scheme is shown in Fig. 1A (*3, 4*). Early ozone management strategies emphasized reductions of anthropogenic emissions of VOCs, such as those emitted in automobile exhaust. These strategies have successfully reduced peak ozone concentrations over time in cities where anthropogenic emissions dominate the ambient VOC mixture (e.g., Los Angeles) (*5*). In the eastern United States, however, large emissions of very reactive VOCs from biogenic sources [e.g., isoprene from oak trees (Fig. 1B)] have been shown to contribute substantially to ozone formation in both rural and urban areas (*6, 7*). Controls on anthropogenic NO_x in addition to anthropogenic VOCs are now thought to be needed to reduce ozone in regions characterized by strong biogenic VOC sources (*1*) (e.g., Atlanta).

Anthropogenic NO_x emissions in the United States are estimated to total 22.2 Tg (as NO_2) per year, with 53% from transportation, 25% from electric utility power plants, 12% from commercial fuel combustion, and the balance emitted from miscellaneous industrial processes (*2*). Early efforts to reduce anthropogenic NO_x were focused on the largest fraction, that from motor vehicles. Although these efforts have resulted in substantial reductions in automobile NO_x emission rates per mile driven, the reductions have been almost completely offset by an increase in vehicle miles traveled, so that total estimated U.S. vehicular NO_x emissions have remained relatively constant over the last two decades (*2*).

Most recently, regulatory attention has been turned to NO_x emissions from electric utility power plants, which have been shown to affect attainment of the U.S. ozone standard in two ways. First, ozone formation downwind of power plants can elevate background ozone levels on a regional scale, making compliance with existing and proposed ozone standards more difficult (*8*). Second, in isolated cases the contribution of an individual power plant plume can cause ozone concentrations to exceed the current 1-hour standard of 120 parts per billion by volume (ppbv) (*9*). In a series of actions intended to mitigate tropospheric ozone formation, the U.S. Environmental Protection Agency (EPA) has proposed a reduction of power plant NO_x emissions by about 0.45 Tg, or 500,000 tons per year, by 2003 (*10*). This amount represents 8% of current power plant NO_x emissions nationwide. The recent ruling, under section 126 of the Clean Air Act, calls for emission reductions at 392 large NO_x sources, chiefly coal-fired power plants located in rural areas in the eastern and mideastern United States (Fig. 1B).

To ease implementation, provide incentives for early reductions, and minimize costs to industry, the rule states that NO_x reductions are to occur within the framework of a cap-and-trade program, in which the total amount of reduction is fixed but trading of emissions within the affected states is allowed (*10*). A similar program has been used to reduce SO_2 emissions by 18% between 1989 and 1998 (*2*). These programs implicitly value all emissions equally, regardless of emission density or location. We demonstrate that control of tropospheric ozone formation by this type of program could be substantially enhanced if it were to additionally consider the NO_x emission rates and locations of the affected sources by providing additional incentives to transfer rural power plant NO_x emissions away from regions with the highest biogenic isoprene emissions.

We focus on observed production rates and yields of ozone from rural power plant NO_x sources, and illustrate how existing programs might benefit from potential improvements in the efficacy of NO_x emission reductions. Data from a field campaign using an instrumented aircraft in the summer of 1999 (*11*) are used to assess two critical factors that influence plume ozone production rate and yield: NO_x emission rate of the power plant, and biogenic isoprene emission in the area. Plume data from three research flights are shown to illustrate these general effects, which are confirmed and extended by the complete data set from multiple aircraft field missions over the course of several years. Example data (*12*) illustrating the observed dependence of ozone formation on NO_x emission rate are shown in Fig. 2.

The complex dependence of ozone formation on NO_x has often been described as nonlinear (*13*). This effect is most clearly illustrated in the flight data of 12 July 1999, in which plumes from two coal-fired power plants with NO_x emission rates differing by a factor of 8 were studied under similar mete-

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orological conditions over similar land use (green box in Fig. 1B). Nonlinearity here results in the familiar pattern of ozone concentration across plume transects (Fig. 2) (*14, 15*). Further, nonlinearity in NO_x has direct implications for ozone control strategies, as illustrated below by scaling observed production rates and yields of ozone to the NO_x emitted from the plants.

Considerable differences in the rate of ozone production are evident, with the less concentrated Johnsonville, Tennessee, plume (Fig. 3A) producing ozone more rapidly than the more concentrated Cumberland, Tennessee, plume (Fig. 3B). The Johnsonville plume was also characterized by more net ozone produced per NO_x originally emitted, in accord with 1995 observations (*16*) and theoretical predictions (*13*). By local sunset in the photochemically aged plumes, measurements showed that nitric acid (HNO_3) accounted for 65% or more of total reactive nitrogen NO_y. Because tropospheric HNO_3 represents an essentially irreversible sink for NO_x on plume ozone formation time scales, the majority of power plant NO_x was no longer available for future ozone production. At this point, the Johnsonville plume had produced ~7 molecules of ozone, and the Cumberland plume ~2 molecules, per NO_x molecule emitted from each source (Fig. 3). Measurements of VOCs on this flight showed similar average levels of isoprene, which at 1.1 ± 0.5 ppbv were sufficient to dominate ambient VOC reactivity in both plumes (*17*). Measured solar actinic flux, radical precursor concentrations, boundary layer height, plume dispersion, and wind speed were similar in these two plumes. Differences in ozone production rate and yield are therefore attributed to the factor of 8 difference in NO_x emission rate between these two plants. In light of a point source NO_x control strategy, we conclude that a hypothetical reduction in NO_x emissions from the Cumberland plant by a factor of 8 (the difference between the Cumberland and Johnsonville NO_x emissions) would have resulted in only a factor of 2.3 reduction in net ozone production (*18*).

Nonlinearity is therefore manifested in less than unit reductions (mol/mol) in ozone resulting from a unit reduction in NO_x emissions, as shown here from ambient measurements by contrasting two constant sources with different NO_x emission rates. Considering plumes with similar VOC reactivity, deliberate NO_x reductions from the largest point sources subject to the EPA ruling will have a lesser impact on ozone concentrations than would equivalent NO_x reductions from several smaller sources. Finally, deliberate NO_x reductions from the largest sources will be partially offset by an increase in the efficiency with which ozone is formed in those plumes (*19*).

Photochemical models of ozone formation [e.g., (*6, 7, 13, 20*)] additionally predict a strong dependence of ozone production rate and yield on the concentration and reactivity of ambient VOCs in these plumes. Because power plants do not emit substantial amounts of VOCs, these typically must be mixed into each plume from the surrounding atmosphere as the plumes are advected downwind. Aircraft observations in plumes sampled over regions of different land use, and hence different biogenic VOC emission sources, confirm this dependence. Further, these observations can be used to estimate the effect of power plant location and surrounding land use on plume ozone formation processes. We now present two examples showing the influence of biogenic isoprene concentration.

Ozone yield data from a flight on 7 July 1999, downwind of the Thomas Hill, Missouri, coal-fired plant, are shown in Fig. 3C. On this day the plume (blue box in Fig. 1B) was remote from isoprene sources, resulting in average measured isoprene concentrations of 0.3 ± 0.2 ppbv; these concentrations are lower than those observed on the 12 July flight by a factor of 3. Thus, ambient VOC reactivity was sufficiently low that background levels of carbon monoxide and methane contributed substantially to hydroxyl radical (OH) loss rates (*17*). Supporting onboard measurements of solar actinic flux, radical precursor concentrations, boundary layer height, and wind speed suggest that the Thomas Hill plume should exhibit ozone formation rates and yields intermediate between the Cumberland and Johnsonville plumes as a result of NO_x concentration differences alone. The relatively low VOC reactivity characteristic of the region resulted in delayed net ozone

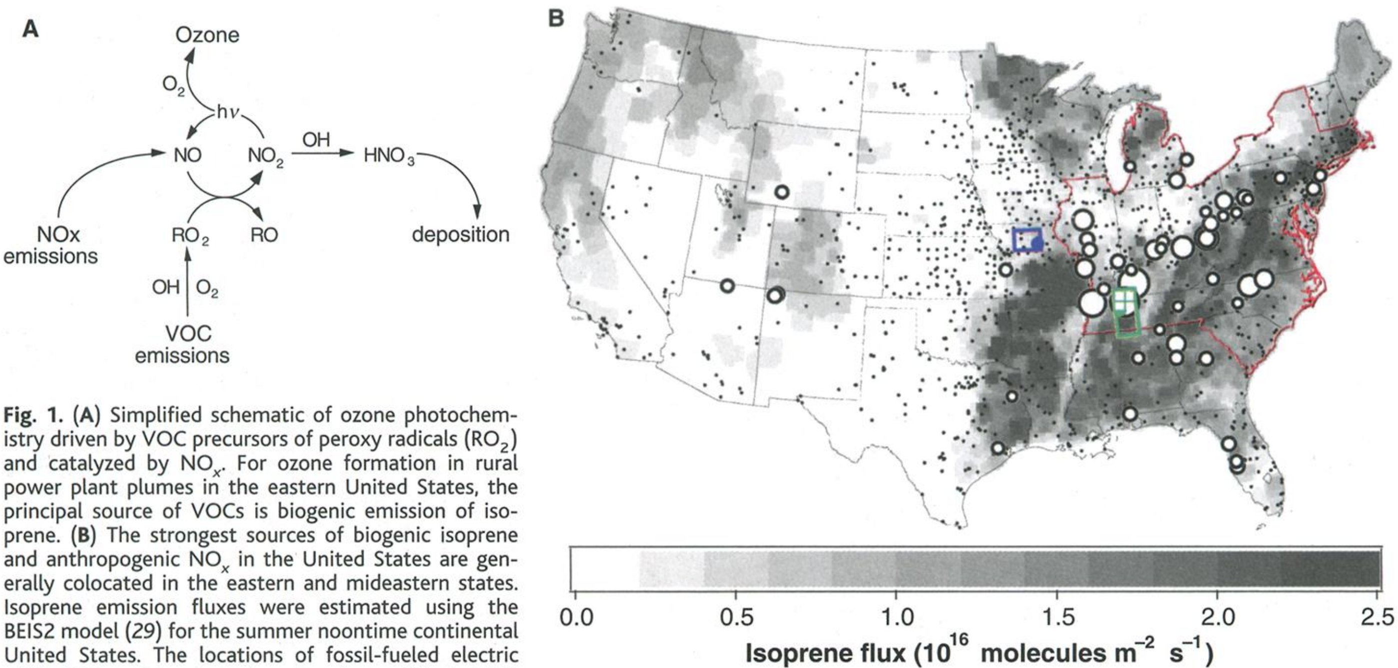

Fig. 1. **(A)** Simplified schematic of ozone photochemistry driven by VOC precursors of peroxy radicals (RO_2) and catalyzed by NO_x. For ozone formation in rural power plant plumes in the eastern United States, the principal source of VOCs is biogenic emission of isoprene. **(B)** The strongest sources of biogenic isoprene and anthropogenic NO_x in the United States are generally colocated in the eastern and mideastern states. Isoprene emission fluxes were estimated using the BEIS2 model (*29*) for the summer noontime continental United States. The locations of fossil-fueled electric utility NO_x point sources are also shown (black dots), with the 50 largest emitters shown as open circles sized according to NO_x source strength (data are available from EPA at www.epa.gov/AIRMARKET/egrid/index.html). Locations of the Thomas Hill (solid blue circle), Cumberland (green cross), and Johnsonville (solid green circle, at lower left of Cumberland) power plants are given, with the plume study areas of the 7 July and 12 July 1999 flights shown in blue and green boxes, respectively. States affected by the recent EPA call for NO_x reductions are outlined in red.

production and the lower observed yield of ~2 ozone molecules per NO_x emitted from the Thomas Hill plant (Fig. 3C).

A second illustration of the dependence of ozone production processes on the ambient concentration of isoprene is afforded by consideration of the partitioning of the oxidized nitrogen products in these plumes. The major products of atmospheric NO_x oxidation are HNO_3 and peroxyacetyl nitrate (PAN) and other organic nitrates (*21*). High ambient VOC concentrations lead to ozone formation initiated by the peroxy radical (RO_2) + NO reaction in the cycle catalyzed by NO_x (Fig. 1A). Low ambient VOC concentrations increase the relative importance of the OH + NO_2 reaction, thereby favoring HNO_3 formation. In an environment low in VOCs and organic peroxy radicals, in the first several hours after emission a proportionally greater fraction of power plant NO_x will be permanently removed by oxidation to HNO_3, with relatively little ozone formed. Contrasting the magnitude and timing of HNO_3 formation between plumes therefore provides a diagnostic of the relative contribution of organic peroxy radicals to NO_x oxidation, and of the relative efficiency with which plume NO_x has catalyzed ozone formation.

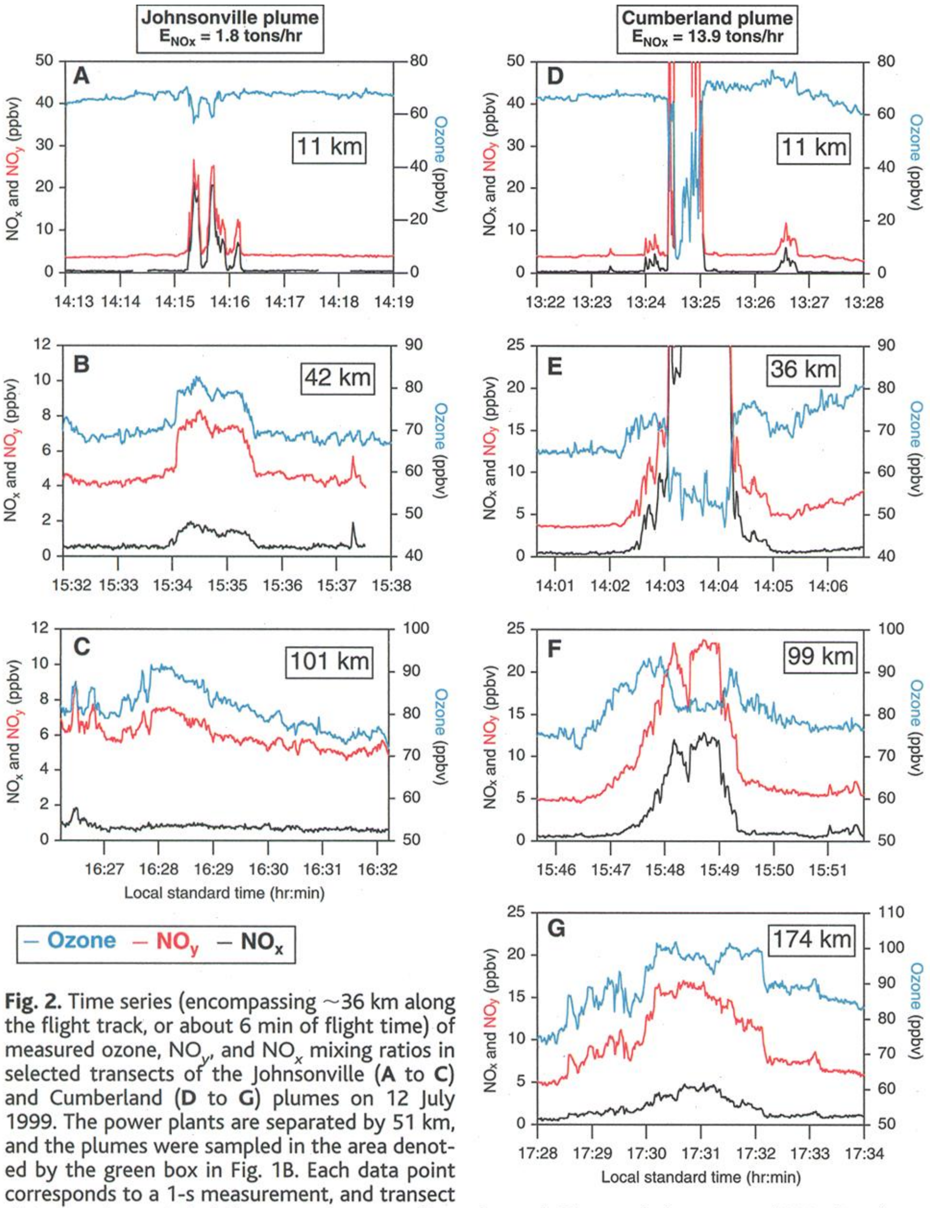

Fig. 2. Time series (encompassing ~36 km along the flight track, or about 6 min of flight time) of measured ozone, NO_y, and NO_x mixing ratios in selected transects of the Johnsonville (**A** to **C**) and Cumberland (**D** to **G**) plumes on 12 July 1999. The power plants are separated by 51 km, and the plumes were sampled in the area denoted by the green box in Fig. 1B. Each data point corresponds to a 1-s measurement, and transect distances downwind of the sources are noted in each panel. Plant emissions rates of NO_x (E_{NO_x}) are given at the top, in units of tons of NO_2 per hour. The nonlinear dependence of ozone production rate on NO_x concentration (*13*) is illustrated most clearly in (E). At this distance, ozone concentrations are enhanced at $NO_x < 10$ ppbv on the edges of the plume, whereas ozone is still suppressed at $NO_x > 15$ ppbv in the plume core. Differences in plume photo-oxidation rates are apparent; although NO_x has been completely oxidized by 101 km downwind in the Johnsonville plume (C), nearly 20% remains at 174 km downwind of Cumberland (G). Note that different panels have different *y* scales.

This contrast is illustrated by comparing measurements in a transect 6.5 hours downwind of the Thomas Hill plant on 7 July to those in a transect of the Johnsonville plume at the same elapsed time since emission on 4 July 1999 (Fig. 4). The NO_x emission rate of 3.0 tons/hour for Johnsonville on 4 July was 166% of the rate on 12 July, owing to changing plant loads under normal operating conditions. Comparing Johnsonville on 4 July to Thomas Hill on 7 July, the respective power plant NO_x emission rates are similar, but isoprene concentrations averaged 3 ± 2 ppbv on 4 July, a factor of 10 increase relative to the 7 July measurement. Similarity in other factors such as measured wind speed, solar flux, and plume dilution rate suggests that observed differences in ozone are primarily due to the large difference in isoprene concentration distinguishing these two plumes. Transect data show that although 80% of the NO_x had been oxidized in both plumes, the Thomas Hill plume included twice as much HNO_3 (Fig. 4A). The Johnsonville plume had produced significantly more organic nitrates, mainly PAN, reflecting a higher concentration of peroxy radicals in the plume. This is borne out in the amount of excess ozone formed in the Johnsonville plume transect, twice as much as at Thomas Hill (Fig. 4B). Thus, increased availability of reactive biogenic VOCs greatly enhances the ozone formation potential of power plant NO_x emissions. NO_x emitted into forested, isoprene-rich areas of the eastern United States will tend to produce more ozone per unit of NO_x emitted, and produce it more rapidly, than if those emissions had occurred in the agricultural Mideast (Fig. 1).

These findings are in accord with current theoretical models of the tropospheric photochemistry of ozone, and they suggest that changes in ambient ozone resulting from a program of power plant NO_x emission reductions will be affected by the size and location of sources that are controlled under such a program. The effectiveness of a cap-and-trade program applied to NO_x emissions could be substantially enhanced by further considering these factors. These considerations would recognize that a ton of NO_x emitted as a dilute plume into a high ambient loading of reactive VOCs would have a larger impact on local ozone levels than the same ton of NO_x emitted as a concentrated plume into a less reactive ambient environment. The latter scenario would produce less total ozone, and the ozone would be produced over a larger area. The data suggest that substantially different impacts, by factors of 2 or greater, on ozone production from power plant NO_x emissions might result from future trading schemes.

The measurements described here further suggest that relatively well-understood gas-phase chemistry involving NO$_x$, peroxy radicals, and OH is sufficient to describe the observed dependencies of ozone formation in these isolated plumes (*3*, *4*). Accurate evaluation of NO$_x$ emissions on the basis of their potential to form ozone should require only measurements or modeling of the few plume species shown in Fig. 4, namely NO$_x$, HNO$_3$, NO$_y$, and ozone itself. In many areas throughout the eastern United States, ambient VOC reactivity is determined by biogenic isoprene (*6*, *7*, *17*). Thus, measurements or models need not rely on extensive characterization of all possible VOCs to adequately simulate ozone formation in these plumes. Regional models using existing and verifiable EPA emissions inventories for power plant NO$_x$ and biogenic isoprene should be able to simulate plume ozone formation to the extent that they accurately disperse the NO$_x$ emissions of large point sources (*15*, *22*).

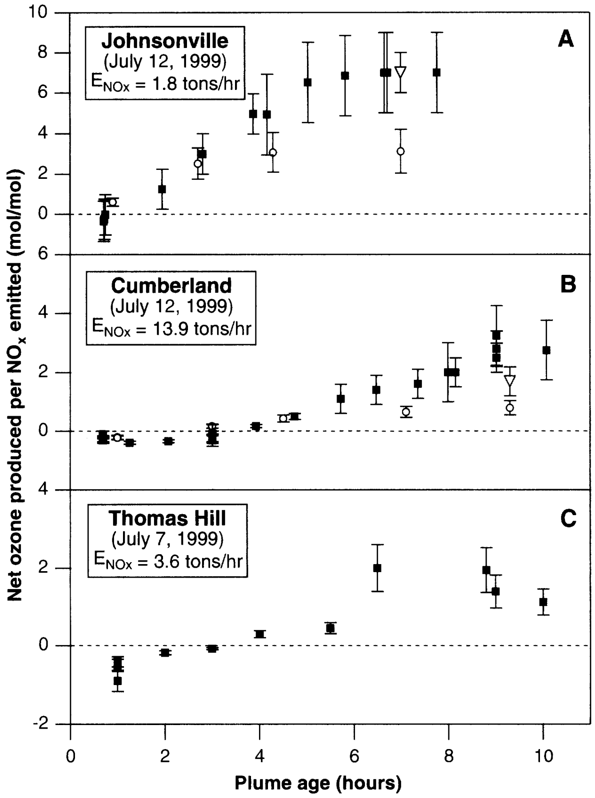

Fig. 3. Net ozone produced per NO$_x$ emitted (solid squares), shown as a function of time since emission from the Johnsonville (**A**) and Cumberland (**B**) plants on 12 July, and the Thomas Hill (**C**) plant on 7 July 1999. Upper and lower limits (open circles and triangles, respectively) for the Cumberland and Johnsonville plumes calculated from an earlier study (*16*) are included for comparison. Ozone production rates are inferred from the rate of increase in net ozone per NO$_x$ emitted and from differences in the time required to produce net ozone above zero in each plume. Note that different panels have different *y* scales.

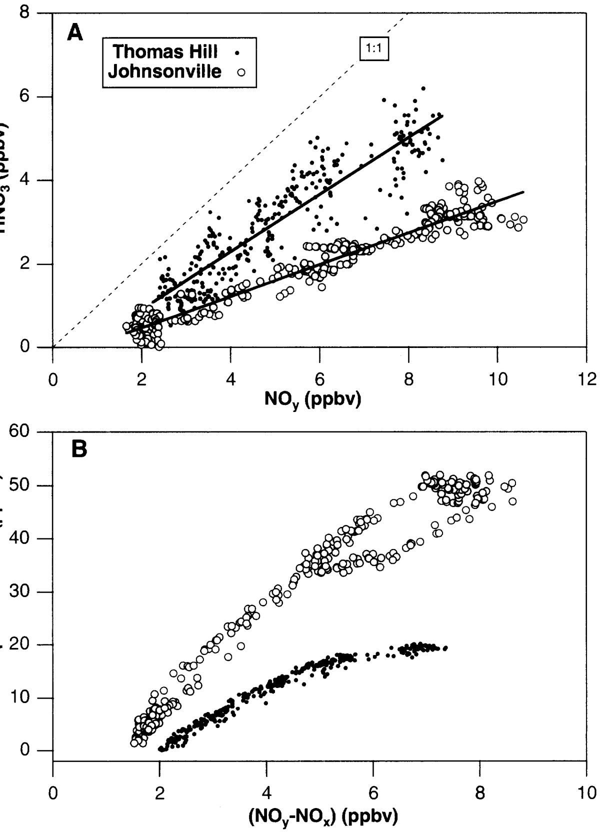

Fig. 4. Measurements (1 per second) during aircraft transects of the Johnsonville (circles; E_{NO_x} = 3.0 tons/hour, 4 July 1999) and Thomas Hill (dots; E_{NO_x} = 3.6 tons/hour, 7 July 1999) power plant plumes, on days characterized by similar meteorological conditions. Equal amounts (80 ± 2%) of the original NO$_x$ emissions had been oxidized in these transects. The lack of reactive VOCs in the Thomas Hill plume favored NO$_x$ oxidation via OH, illustrated by nearly a doubling of HNO$_3$ formation (**A**) at this transect. Solid lines represent linear-least-squares regression fits to the data, with the slopes providing a measure of the fractional contribution of HNO$_3$ to NO$_y$. Relatively high concentrations of biogenic isoprene in the Johnsonville plume favored peroxy radical formation, resulting in twice as much ozone from an equivalent amount of NO$_x$ emitted (**B**).

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11. Power plant plume chemical evolution over time was investigated using instrumentation aboard the NOAA WP-3D research aircraft in June and July 1999 (*23*). Oxidation processes as a function of time downwind of the emissions sources are estimated from plume transect data using two complementary approaches. In the first approach, the evolution of chemical tracer correlation slopes is interpreted to determine photochemical processing over time [e.g., (*16*, *24–26*)]. The second approach relies on a plume mass balance calculation to estimate net plume photochemical production and loss for each species (*16*, *27*, *28*). The approaches are complementary in that each is subject to different uncertainties, such that agreement between the two approaches provides increased confidence in the resulting conclusions. For the data presented here, agreement is typically within ±20%. Power plant NO$_x$ emission rates are derived from continuous emissions monitoring systems at each plant and agree within ±25% with extrapolations from onboard measurements.
12. These include measurements of total reactive gas-phase nitrogen oxides NO$_y$ (= NO$_x$ + HNO$_3$ + peroxyacyl nitrates + alkyl nitrates), which serve as a semiconserved tracer of power plant NO$_x$ emissions (*24*).
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18. Hypothetical reduction in net ozone by a factor of 2.3 is obtained by dividing the quantity (NO$_x$ emis-

sions × ozone yield) for Cumberland by that for Johnsonville. Any unreacted NO_x (about 20% in the 12 July case) remaining in the Cumberland plume at local sunset could continue ozone production upon sunrise the next day. That remaining 20%, having been diluted overnight, should produce ozone more efficiently, just as in the dilute Johnsonville plume. Assuming an efficiency of 7 the next day for this unreacted NO_x, including it in the calculation for the Cumberland plume increases its estimated total efficiency by 50% to 3 molecules of ozone per NO_x emitted, still lower than Johnsonville by a factor of 2. Any nighttime oxidation of unreacted NO_x via processes involving the nitrate radical would act to minimize this increase.

19. These findings additionally imply a strong dependence of ozone production rate and yield on the dilution induced by natural meteorological processes after emission. Given that these factors can change drastically in relatively short amounts of time, we should expect a range of ozone rates and yields from a single point source emitting NO_x at a constant rate. Meteorological changes may affect multiple parameters influencing ozone photochemistry (e.g., a cloud deck that attenuates actinic fluxes may also reduce the surface heating that drives vertical mixing of plume constituents in the boundary layer).
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30. We thank the air crew and systems engineering staff of the NOAA Aircraft Operations Center for their exceptional service, S. A. McKeen for providing the modeled isoprene emissions data shown in Fig. 1A, and two anonymous reviewers for suggestions that improved the manuscript. Portions of this work were funded through the NOAA Health of the Atmosphere and Climate and Global Change research programs.

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Dating Caral, a Preceramic Site in the Supe Valley on the Central Coast of Peru

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Radiocarbon dates from the site of Caral in the Supe Valley of Peru indicate that monumental corporate architecture, urban settlement, and irrigation agriculture began in the Americas by 4090 years before the present (2627 calibrated years B.C.) to 3640 years before the present (1977 calibrated years B.C.). Caral is located 23 kilometers inland from the Pacific coast and contains a central zone of monumental, residential, and nonresidential architecture covering an area of 65 hectares. Caral is one of 18 large preceramic sites in the Supe Valley.

The Late Archaic or Cotton Preceramic Period from ~5000 to 3750 years before the present (yr B.P.) was a time of substantial cultural change and the emergence of the first complex societies in the Americas. Here, we present radiocarbon dates from the site of Caral in the Supe Valley.

The Supe Valley, ~200 km north of Lima, was a focal point for early cultural development on the coast of Peru (Fig. 1). The Valley stretches ~90 km from the Pacific coastline to the sharply rising slopes of the Andes. There are, at present, 70 km^2 under cultivation and an average stream flow of 48,000,000 m^3 per year.

Supe has been perhaps best known in the archaeological literature as the location of the large coastal site of Aspero (Fig. 1). This site was first recorded in 1905 (*1*), and initial excavations at the site were carried out in 1941 (*2*). The site was revisited in the 1970s (*3*), when artificially constructed platform mounds were recognized for the first time. Further work at the site in the 1970s (*4, 5*) yielded dates of 3000 to 2400 calibrated years B.C. (Cal B.C.), which belong to the Preceramic Period in Peruvian prehistory. Aspero is distinctive in having an economy based primarily on maritime resources rather than on agriculture and domesticated plants. The site has been central to ongoing discussions of the maritime foundations of Andean civilization (*6, 7*).

A number of other large sites inland in the Supe Valley also have monumental architecture and lack surface ceramics (*8–10*). These include Caral (*11*) and 17 additional sites in the valley with extensive preceramic occupations. Caral (Fig. 2) is one of the largest and the most formally laid out of the major preceramic sites in the Supe Valley. It is located 23 km inland from the coast on a terrace 25 m above the floodplain of the Supe River on the south side of the valley (*12, 13*).

The central zone of Caral with monumental architecture covers an area of just over 65 ha, with the NW-SE axis measuring 1045 m and the NE-SW axis measuring 620 m (Figs. 3 and 4). This central zone includes six large platform mounds, numerous smaller platform mounds, two sunken circular plazas, an array of residential architecture, and various complexes of platforms and buildings. The largest of the platform mounds, the Piramide Mayor, measures 160 m by 150 m and is 18 m high. Testing within this platform mound indicates that, although there was much remodeling of the surface structures, the mound itself was constructed principally in two massive construction phases. The other five mounds range down in size to the smallest, which is ~60 m by 45 m and 10 m high. These other mounds appear to have been constructed in one or two major phases. All construction employed cut stone retaining walls with a combination of river cobbles and cut stone rubble fill. The fill material was transported to the mound in shicra bags [an open mesh bag made of reeds (*14*)], which were filled with stones and then placed inside the retaining walls, bag and all. The outer retaining walls were carefully faced and covered with multiple layers of colored plaster.

Secondary smaller mound alignments extend 1000 m to the northwest and 500 m to the southeast from the site center. These areas have not been tested, and at present, their chronological placement is uncertain. Details of these occupations are unclear because of much later ceramic-bearing architecture near these secondary mound extensions. In aerial photographs (Fig. 2), however, these long mound complexes appear to be integral parts of the original preceramic site. About 300 m to the southwest of the central mound complex at Caral is another sunken circular plaza and platform complex (which retains the name "Chupacigarro") (visible at the bottom center of Fig. 2). Its architecture covers an additional 23 ha. There is surface trash and residential architecture in between the central mound zone of Caral and the complex of Chupacigarro.

Excavations at Caral have revealed considerable diversity in residential architecture. Each of the six large mounds is associated with a large formally arranged residential complex. Each room complex covers an area of between 450 and 800 m^2, with carefully constructed and heavily plastered walls of cut stone. Domestic trash indicates that these rooms were residential in nature. A second kind of residential architecture is found in Sector A, covering an area of 5500 m^2 (Fig. 3). Rooms in this area are smaller and had walls built of wood poles, cane, and mud. This area shows extensive evidence of remodeling and sequential occupation, and construction of low platforms made of river cobbles in the later stages. Similar residential complexes are found in other parts of the site,

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