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Authors

ELLIOTT, SCOTT
SHEN, MEI
BLAKE, DONALD R
et al.

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Atmospheric Effects of the Emerging Mainland Chinese Transportation System at and Beyond the Regional Scale

SCOTT ELLIOTT [1], MEI SHEN [2], DONALD R. BLAKE [3], RONG LU [2], ARMISTEAD G. RUSSELL [4], C. Y. J. KAO [1], GERALD E. STREIT [5], XUE PENG ZHAO [2], EDWARD IAIN MCCREARY [5], F. SHERWOOD ROWLAND [3], MICHAEL J. BROWN [5] and RICHARD P. TURCO [2]

[1]*Earth and Environmental Sciences Division, Geoanalysis Group, Los Alamos National Laboratory, Los Alamos, NM 87545, U.S.A.*
[2]*Atmospheric Sciences Department, University of California, Los Angeles, CA 90024, U.S.A.*
[3]*Chemistry Department, University of California, Irvine, CA 92715, U.S.A.*
[4]*Civil Engineering Department, Georgia Institute of Technology, Atlanta, GE 30033, U.S.A.*
[5]*Technology and Safety Assessment Division, Energy and Environmental Analysis Group, Los Alamos National Laboratory, Los Alamos, NM 87545, U.S.A.*

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Abstract. Local surface travel needs in the People's Republic of China (mainland China) have traditionally been met largely by nonpolluting bicycles. A major automobile manufacturing/importing effort has begun in the country over the last decade, and planning documents indicate that the Chinese may strive to acquire more than 100 million vehicles early in the next century. By analogy with large automotive fleets already existing in the western world, both regional and global scale pollution effects are to be expected from the increase. The present work adopts the latest projections of Chinese automobile manufacture and performs some quantitative assessments of the extent of pollution generation.

Focus for the investigation is placed upon the oxidant ozone. Emissions of the precursor species nitrogen oxides and volatile organics are constructed based on data for the current automotive sector in the eastern portion of the United States. Ozone production is first estimated from measured values for continental/oceanic scale yields relative to precursor oxidation. The estimates are then corroborated through idealized two dimensional modeling of the photochemistry taking place in springtime air flow off the Asian land mass and toward the Pacific Ocean. The projected fleet sizes could increase coastal and remote oceanic ozone concentrations by tens of parts per billion (ppb) in the lower troposphere. Influences on the tropospheric aerosol system and on the major greenhouse gas carbon dioxide are treated peripherally. Nitrogen oxides created during the vehicular internal combustion process will contribute to nitrate pollution levels measured in the open Pacific. The potential for soot and fugitive dust increases should be considered as the automotive infrastructure develops. Since the emerging Chinese automotive transportation system will represent a substantial addition to the global fleet and all the carbon in gasoline is eventually oxidized completely, a significant rise in global carbon dioxide inputs will ensue as well.

Some policy issues are treated preliminary. The assumption is made that alterations to regional oxidant/aerosol systems and to terrestrial climate are conceivable. The likelihood that the Chinese can achieve the latest vehicle fleet goals is discussed, from the points of view of new production, positive pollution feedbacks from a growing automobile industry, and known petroleum reserves. Vehicular fuel and maintenance options lying before the Chinese are outlines and compared. To provide some perspective on the magnitude of the environmental changes associated with an Asian automotive buildup, recent estimates of the effects of future air traffic over the Pacific Rim are described.

Key words: Chinese vehicles, ozone, aerosols, carbon dioxide, alternative fuels.

1. Introduction

The current mainland Chinese transportation system is to a large extent nonmotorized. The People's Republic of China today has fewer automobiles per capita and less paved roadway per unit area than any major country on earth (World Resources Institute, 1992; World Bank and Chinese Ministry of Communications, 1994). There is bicycle for every four human beings in the P.R.C., but until recently fewer than a million private citizens possessed automobiles (Zhihao *et al.*, 1990; Sathaye *et al.*, 1994; Brown, 1995). Energy demands have traditionally been met mainly through coal combustion (Wu *et al.*, 1985; Zhao and Sun, 1986; Smil, 1988; Bhatti and Streets, 1991; Sinton *et al.*, 1992; Smith *et al.*, 1994; Qung and Songkong, 1995; Yan-Qing, 1995). However, a major shift toward petroleum based fuels is underway, and is driven largely by the demand from/for vehicles (Xu and Zhu, 1991 and 1994; Sinton *et al.*, 1992; Qin and Chan, 1993; Smith *et al.*, 1994; Zhang *et al.*, 1995; Tang *et al.*, 1995). Mainland China has the largest population of any country on earth, on the order of one billion (U.N. 1989 and 1990; World Resources Institute, 1992). The market for automobiles is larger than in the United States and Europe combined (Sathaye *et al.*, 1994; Barnathon *et al.*, 1996). Recent indications are that the Chinese will attempt to develop a vehicular transportation system along lines pioneered in the U.S., Western Europe and Japan. Trade journals and the popular press echo P.R.C. policy statements to this effect (Tyler, 1994; Brown, 1995; Barnathon *et al.*, 1996; Engardio and Roberts, 1996). Estimates are that the national production capacity will be on the order of 3 million automobiles and trucks per year by the year 2000 (Brown, 1995; Engardio and Roberts, 1996). Documents drafted by the Chinese Government discuss the inadequacies and deteriorating state of the current highway infrastructure (World Bank and Chinese Ministry of Communications, 1994). Remedies are consistent with more than one hundred million vehicles coming on line early in the next century.

The figures are as yet quite speculative, but much smaller numbers of automobiles have long been known to lower air quality even in the largest urban areas (Haagen Smit *et al.*, 1951 and 1959; Haagen Smit, 1952; Leighton, 1961; Ferman *et al.*, 1981; Kretzschmar, 1993, 1994; MARI, 1994). Furthermore, international fleets of comparable size constitute a serious regional pollution concern (Hough and Derwent, 1990; Crutzen and Zimmerman, 1991; McKeen *et al.*, 1991a, b; Parrish *et al.*, 1993; Langman and Graf, 1995). Detriments attributable to automotive primary emissions and the associated secondary materials include the health effects of carbon monoxide, volatile organics and aerosols (Kittelson and Dolan, 1980; NAS, 1983a; Andrade *et al.*, 1994; Villalobos-Piatrini *et al.*, 1995), and the agricultural/natural impacts of ozone and particulates (Trijonis, 1989; Chameides *et al.*, 1994). Chinese scientists have for several decades studied the ramifications of pollution sources already existing in their country (UDC, 1982; Zhao and Zhao, 1985; Mingxing, 1985; Dai *et al.*, 1986; Zhao and Xiong, 1988; Tao *et al.*, 1992; Xu *et al.*, 1993). Because of the potential for severe alterations to the chemistry of the regional atmosphere, we take the most recent Chinese vehicle production projections as a starting point for consideration of new Asian emissions. The size of the impending fleet portends global level atmospheric effects as well. The total number of internal combustion vehicles on earth would rise significantly (Lu, 1991; World Resources Institute, 1992; Cutter Information, 1992). Automobiles and trucks are already major contributors to the terrestrial carbon dioxide loading (Marland and Rotty, 1984; Marland *et al.*, 1985; Houghton *et al.*, 1990).

Our primary concern in the present work will be the ozone generation which takes place through the oxidation of volatile organics in the presence of nitrogen oxides. Existing model results (Liu *et al.*, 1987; Crutzen, 1988; Lin *et al.*, 1988; Jacob *et al.*, 1993a, b; Chin *et al.*, 1994) and computational tools which several of us have been involved in developing (Elliott *et al.*, 1993a, b; Elliott *et al.*, 1995a, b; Shen *et al.*, 1996) permit a zeroth order but quantitative analysis of the upcoming Asian ozone situation. We will also perform a scoping overview of the potential for alterations to Pacific Rim nitrate fields (Kondo *et al.*, 1987; Prospero and Savoie, 1989; Akimoto *et al.*, 1994; Kondo *et al.*, 1996; Koike *et al.*, 1996), which feed into the regional aerosol system (Duce *et al.*, 1991). The large scale aerosol effects are for the moment difficult to simulate rigorously (Erickson *et al.*, 1991; Langner and Rodhe, 1991; Penner *et al.*, 1994; Benkowitz *et al.*, 1994). However, because understanding of the physicochemistry of particles is thought to contain the most significant uncertainties associated with predictions of global climate change (Twomey, 1977; Twomey *et al.*, 1984; Albrecht, 1989; Charlson *et al.*, 1991 and 1992; Kiehl and Briegleb, 1993), we feel that at least some preliminary comments are warranted. General circulation model calculations with carbon dioxide levels sufficient to represent the potential Chinese vehicular contribution have already been performed as medium/worst case scenarios (e.g. Houghton *et al.*, 1990; Houghton *et al.*, 1992). Although the CO_2 enhancements resulting from an enlarged Asian fleet may have important net climate effects, we here discuss them only on a budgetary level (Marland and Rotty, 1984; Marland, 1989; Lu, 1991; Kato *et al.*, 1991).

Our text opens with a brief review of background tropospheric chemistry for East Asia and the Western Pacific (e.g. Chung, 1986; Sunwoo and Carmichael, 1992; Sunwoo *et al.*, 1992; Liu *et al.*, 1992; Blake *et al.*, 1996a, b; Sing *et al.*, 1996a, b). The information base is far from complete, and so data are drawn when necessary from a meteorological analog region, the Atlantic coast of North America (Galloway *et al.*, 1992; Anderson *et al.*, 1993; Parrish *et al.*, 1993; Benkowitz *et al.*, 1994; Berkowitz *et al.*, 1995). We then move on to another short review, this one of urban pollution in Asia and most particularly in the P.R.C. Again analogies with other parts of the world are useful (NRC, 1991; United Nations, 1992; Kretzschmar, 1993, 1994), but a large amount of high quality research is in fact accessible (e.g. Qin and Chan, 1993; Xu and Zhu, 1994; Tang *et al.*, 1995; Moon *et al.*, 1995). A description of the continental scale meteorological backdrop follows, drawing the transport connection between sources of urban and vehicular pollution and pristine areas of the Pacific Rim (Rex and Goldberg, 1958; Shaw *et al.*, 1980; Duce *et al.*, 1980; Prospero and Savoie, 1989). We make note of a particularly simple wind situation which exists in the springtime, when the prevailing westeries are in force at most altitude or pressure levels and from the center of the continent far out over the open ocean (Merrill *et al.*, 1985; Kotamarthi and Carmichael, 1990). Next, a set of estimates of ozone production is undertaken relative to an enhanced Chinese automobile fleet. Vehicular and other emissions of the nitrogen oxides have been compiled several times for East Asia (Kato *et al.*, 1991; Fujita *et al.*, 1991; Kato and Akimoto, 1992; Akimoto and Narita, 1994). For the nonmethane hydrocarbons and carbon monoxide, the corresponding data are sparse (Graedel, 1994; Guenther *et al.*, 1995; Yuhua *et al.*, 1995). We adopt the simple expedient of extrapolating from the Eastern United States (NRC, 1991; Hameed and Dignon, 1991). This necessarily entails an initial assumption that new vehicles will be traditional in engine design (Chock *et al.*, 1994). Several atmospheric chemistry modeling groups have conducted generic simulations of ozone production over a range of concentration combinations for the precursors nonmethane hydrocarbons (NMHC) and the nitrogen oxides (Liu *et al.*, 1987; Jacob *et al.*, 1989). Effective yields have been tabulated, and can be applied to Chinese emissions to give estimates of ozone production levels. We also describe corroborative calculations which employ some of the coding we have developed in recent years (Elliott *et al.*, 1993a, b; 1995a, b; Shen *et al.*, 1996). Highly idealized representations of the simple springtime synoptic conditions are assumed in the framework of a two dimensional, full photochemical tracer transport model. Alterations to Asian nitrate, soot and mineral dust loadings are treated (Prospero and Savoie, 1989; Parungo *et al.*, 1994). Carbon dioxide releases are dealt with on an absolute basis and superimposed conceptually on contributions from the present global fleet (Marland and Rotty, 1984; Sinton *et al.*, 1992; Sathaye *et al.*, 1994).

The various computations and budgets suggest that significant alterations to tropospheric chemistry could result from the potential Chinese automotive infrastructure of the next century. In our idealized springtime photochemical simulations, ozone increases of tens of ppb ensue in some likely scenarios. The vehicles we assume will add significantly to nitrogen oxide and nitrate concentrations in East Asia and the Western Pacific. Both ozone and the nitrates are already considered among the major Pacific Rim pollution problems (Su, 1978; Kondo *et al.*, 1987; Prospero and Savoie, 1989; Kotamarthi and Carmichael, 1990; Rodhe *et al.*, 1992). Poorly maintained vehicles and roads will bolster the export of all compositional types of particulates to pristine areas (Kittelson and Dolan, 1980; Ji *et al.*, 1993;

Parungo *et al.*, 1994; Zhou *et al.*, 1994; Zhang *et al.*, 1995; Zhu *et al.*, 1995). The augmented carbon dioxide flux may accelerate global climate change (Marland and Rotty, 1984; Houghton *et al.*, 1990; Lu, 1991). It is clear that any new and massive automotive transportation system in the P.R.C. will impact the environment at a variety of scales. We conclude our text with some discussion of policy implications for the impending vehicle buildup. It is valid to question the ability of the Chinese economy to sustain a major automobile fleet. We briefly outline prospects for increased vehicle production, increased pollution emissions resulting from new production, and directions for growth in the Chinese energy sector (Smil, 1988; Sinton *et al.*, 1992; Sathaye *et al.*, 1994; Smith *et al.*, 1994). The gasoline powered automobiles upon which we base our calculations are likely for several decades to offer a cost effective means for transporting the citizens in developing areas (Lu, 1991; Zhang *et al.*, 1995; Brown, 1995). However, petroleum reserves are not unlimited (Sinton *et al.*, 1992; DeCicco and Ross, 1994). Alternative fuels have been considered for propelling ground transportation (Bravo *et al.*, 1991; Russell *et al.*, 1990; Chock *et al.*, 1994; Lave *et al.*, 1995), along with the incumbent environmental side effects (Williams, 1989). We take the liberty here of speculating on their efficacy under East Asian conditions. Issues such as vehicle design and maintenance are touched upon briefly (DeCicco and Ross, 1994; Zhang *et al.*, 1995). As a novel perspective on the postmillennial surface vehicular system in the P.R.C., we review recent estimates of the changes to remote tropospheric chemistry which will be connected with subsonic aircraft (Baugham *et al.*, 1993; Brasseur *et al.*, 1996).

Also in the final discussion section, several uncertainties inherent in our arguments are listed. We reiterate that the Asian automotive fleet of the next century is for the moment largely a speculation (Sathaye *et al.*, 1994; Brown, 1995), but then move quickly to more immediate issues. It is acknowledged, for example, that verifiable data are not available regarding the volatile organic to nitrogen oxide ratio for emissions from present day Chinese vehicles (Kotamarthi and Carmichael, 1990; Tang *et al.*, 1995). Furthermore, the reader will note that the meteorology which drives our photochemistry computations has been greatly idealized (Kotamarthi and Carmichael, 1990). We conclude nevertheless that our major points remain intact; emissions expected from a massive buildup in automobiles in the People's Republic of China will lead to regional scale increases in tropospheric ozone, nitrates and aerosols, as well as global carbon dioxide.

2. Background Asian Air Chemistry

We begin here by surveying the literature on composition for the portion of the remote troposphere which acts as a backdrop for our deliberations. The background atmosphere over central Asia has not received the level of measurement campaign attention which has been devoted to North America or Europe over the last few decades. Several lines of evidence can be called upon to construct likely species distributions. Global scale modelers and reviewers of global tropospheric chemistry have drawn on data for more thoroughly studied continents (Logan *et al.*, 1981; Logan, 1983, 1985; Crutzen and Zimmerman, 1991; Langner and Rodhe, 1991; Penner *et al.*, 1991; Kanakidou and Crutzen, 1993; Muller and Brasseur, 1995). Measurements in the continental outflow can at certain times of the year be revealing (Duce *et al.*, 1980; Kondo *et al.*, 1987; Prospero and Savoie, 1989). For example, the Pacific Exploratory Missions (PEM A and B) were conducted in the fall and spring seasons to respectively minimize and maximize input to the Western Pacific from continental air masses (e.g., Hoell *et al.*, 1996; Kondo *et al.*, 1996; Koike *et al.*, 1996; Blake *et al.*, 1996a, b). Satellite data can sometimes be deconvoluted to provide insight into the chemistry of the troposphere (Sunwoo *et al.*, 1992), and routine urban surface measurements often reflect background conditions (Xu and Zhu, 1994; Tang *et al.*, 1995). Data for the PEM B springtime field experiment are likely to be useful, but are in the review stages as we are composing this work.

Carbon monoxide concentrations recorded at surface stations across China display variation from low remote levels of just over 100 ppb, through the several hundred ppb typical of regional scale photochemical pollution and into the urban ppm range (e.g. Qin and Chan, 1993; Tang *et al.*, 1995; Zhang *et al.*, 1995). Several of us have been involved in surface determinations made outside of cities in the P.R.C. (DRB and FSR). Continental data indicate widespread carbon monoxide emissions. Measurements from the Western Pacific display the higher background concentrations at middle latitudes which have often been noted in major reviews (Figure 1). They develop in part because of low winter removal rates (Singh and Zimmerman, 1992), but at times also reflect layers of pollution. We adopt coarse profiles from Logan *et al.* (1981) here. The situation for the nitrogen oxides is similar. Some urban data sets provide a glimpse of the background troposphere opportunistically (Xu and Zhu, 1991, 1994; Xu *et al.*, 1992; Tang *et al.*, 1995). The studies of Kondo and associates (Kondo *et al.*, 1987; Kondo *et al.*, 1996; Koike *et al.*, 1996) on air masses just off the Chinese coast suggest NO_x levels of between 10 and 100 ppt, consistent with results of lower dimensionality models which attempt to represent the global troposphere (Logan *et al.*, 1981) and with more recent three dimensional simulations (Penner *et al.*, 1991). NO_y levels are on the order of 100 to 1000 ppt. To our knowledge, volatile organics have but rarely been measured over the Asian continent. Limited emissions data for natural hydrocarbons such as the terpenes have been constructed based on determinations in vegetated areas (Yuhua *et al.*, 1995). Urban results tend to focus on tailpipe level measurements (Tang *et al.*, 1995; Zhang *et al.*, 1995). We will take remote measurements for longer lived species such as the low molecular weight alkanes from the works Blake and Rowland (1986), Rudolph (1988) and Sing *et al.* (1988). A relatively complete review which we will use as a resource is that of Singh and Zimmerman (1992).

The urban measurement stations may provide some opportunistic ozone concentration data in addition to insight into CO and the nitrogen oxides (Xu and Zhu, 1993 and 1994; Tang *et al.*, 1995). Ground based measurements in Japan during

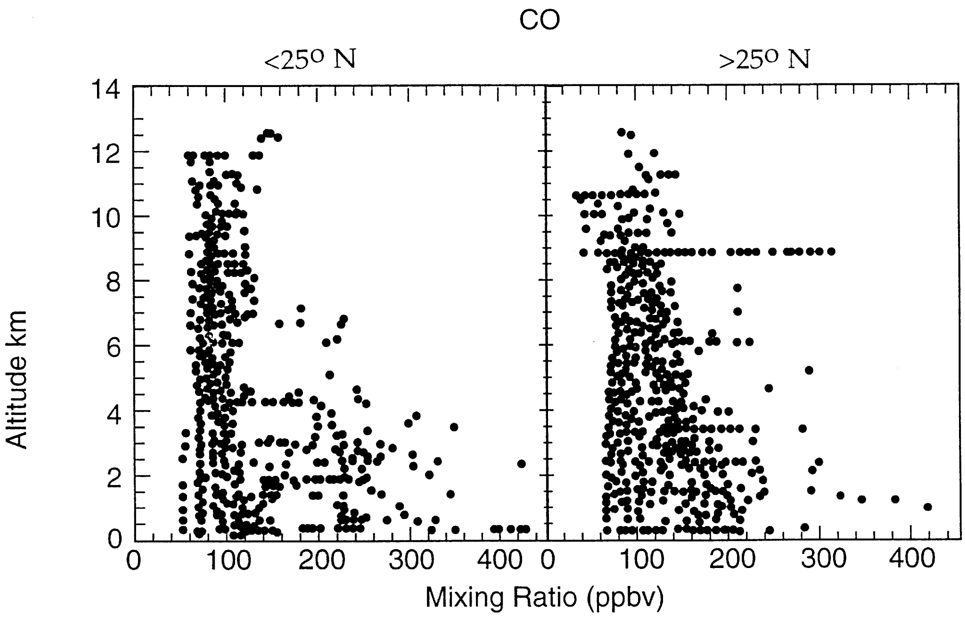

Figure 1. Carbon monoxide measurements from the autumn and springtime Pacific Exploratory Missions (Blake *et al.*, 1996b). The data are in close accord with background vertical profiles adopted for our modeling purposes from Logan *et al.* (1981).

periods of Asian outflow vary from roughly 20 ppb in the fall to 60 ppb in the spring (Sunwoo *et al.*, 1994). Satellite measurements of ozone have been performed over the entire East Asian region through the expedient of subtracting stratospheric concentrations from the total column to yield tropospheric residuals (Sunwoo *et al.*, 1992). Column concentrations of ozone are on the order of 30 Dobson units over the Pacific Rim, with springtime maxima. We will not attempt here to comment on the relative strengths of surface and stratospheric ozone sources (Singh *et al.*, 1978; Fishman and Crutzen, 1978; Gidel and Shapiro, 1980; Mahlman *et al.*, 1980; Logan, 1985). The satellite column levels are consistent with background concentrations of several tens of ppb and a small anthropogenic input (Kotamarthi and Carmichael, 1990). Latitudinal averages from low dimensionality models and three dimensional structures generated by newer tracer transport programs are both in accord (Logan *et al.*, 1981; Crutzen and Zimmerman, 1991).

Distributions in the aerosol system are somewhat less well characterized. The Chinese have been sensitive to aerosol health effects for several decades and so have monitored particulates in urban areas and conducted medical studies (Weiguang, 1981; WHO, 1985; Zhao and Zhao, 1985). Classical source receptor analyses have often been applied to the monitoring results (Dai *et al.*, 1986; Mingxing, 1985; Zhang *et al.*, 1987). Acid precipitation has been identified as a pollution problem in remote areas (Zhao and Sun, 1986; Zhao and Ziong, 1988; Wang and Shi, 1991) and several trace elements have been tracked on trajectories over the Pacific (Savoie and Prospero, 1989; Prospero and Savoie, 1989; Duce *et al.*, 1991; Parungo *et al.*, 1994). Only a handful of publications discuss remote aerosols on the Chinese mainland separately from the sulfuric acid issues (Su *et al.*, 1991; Parungo, 1993; Zhu *et al.*, 1995). An exception to this rule of thumb is the phenomenon of the Kosa dust storm, which is known to transport mineral dust from central Asia far out onto the Pacific (Windom *et al.*, 1967; Duce *et al.*, 1991). Hemispheric to global scale models of the aerosol system are less well developed at present than their photochemical counterparts, and so are not as instructive. Several currently deal with the competition between continental and oceanic sulfur sources to produce remote marine cloud condensation nuclei (Erickson *et al.*, 1991; Benkowitz *et al.*, 1994).

Our investigations into aerosol effects will be less rigorous than those of the ozone system. For our purposes it will be sufficient to assume a typical continental background mix of mineral dusts (Ji *et al.*, 1993; Zhu *et al.*, 1995), aqueous sulfate/nitrate aerosols (Rodhe *et al.*, 1992; Zhu *et al.*, 1994), and black carbon (Parungo *et al.*, 1994; Tang *et al.*, 1995). Several conceptual models of large to global scale aerosol systems have been proposed (Toon and Pollack, 1976; Shettle and Fenn, 1979; Nemesure *et al.*, 1995; Boucher and Anderson, 1995). The assumptions we adopt will be consistent with them. Our conclusion regarding the regional effects of the emerging Chinese automotive network on aerosol systems will be restricted to computations of the conversion of nitrogen oxides to nitrate, which can enter particulates, and to qualitative statements on dust and soot.

Assumptions regarding the composition of the background Asian atmosphere can be validated to some extent by comparison with the more complete data sets available for the U.S. east coast and the North Atlantic ocean. The two areas lie at latitudes similar to populous parts of the Peoples Republic of China, and are also related to one another by transport in the geostrophic westerlies. Studies of continental to ocean exchange of the nitrogen oxides (Galloway *et al.*, 1992), non-methane hydrocarbons (Penkett *et al.*, 1993), ozone (Parrish *et al.*, 1993; Anderson *et al.*, 1993; Chin *et al.*, 1994; Berkowitz *et al.*, 1995), and particles (Harriss *et al.*, 1984; Galloway *et al.*, 1992) are all readily available in the Western literature.

3. Asian Urban Air Chemistry

Reports on the air pollution chemistry of Chinese cities are quite numerous. Several dozen can be obtained within the Western scientific literature (e.g. Xu and Zhu, 1994; Tang *et al.*, 1995) and many more have been published in Asia (e.g. UDC, 1982; Environmental Protection Agency of China, 1991; Zhu and Xu, 1993). Although the volume of information does not approach that of the Western air pollution literature, it will still be necessary for us to focus on a handful of definitive papers dealing with major urban areas (Su *et al.*, 1991; Qin and Chan, 1993; Xu and Zhu, 1994; Tang *et al.*, 1995). A detailed literature also exists on air pollution health effects in China (Tao *et al.*, 1992; Xu *et al.*, 1993, 1994, 1995). In some cases non-Chinese urban analogs will be useful, especially in the Koreas (Kong and Sang, 1991; Moon *et al.*, 1995; Lee, 1995).

Our own Chinese carbon monoxide measurements extend into the urban arena (DRB and FSR), and indicate that episodic concentrations typical of western urban zones can be approached (e.g. Mayrsohn *et al.*, 1975, MARI, 1994; blake and Rowland, 1995). Research conducted by P.R.C. scientists is in concert (Qin and Chan, 1993). Carbon monoxide emissions from individual vehicles currently in operation seem to be higher than in the United States (Zhang *et al.*, 1995; Tang *et al.*, 1995). Qin and Chan (1993) offer surface measurements of CO over the course of several days in Guangzhou, and from them local emissions can be deduced. The fluxes approach those of Mexico City (MARI, 1994; Fernandez-Bremauntz and Ashmore, 1995). Nitrogen oxide diurnal cycles have also been recorded (Xu and Zhu, 1994) and absolute concentrations are again in the range observed in Western cities (MARI, 1994). We have not located urban measurements of the nonmethane hydrocarbons for China. However, hydrocarbon data are even in scarce supply domestically (Seinfeld, 1989; Seila *et al.*, 1989).

Qin and Chan (1993) state based on mass balance computations that even at present traffic levels, automobiles are the major sources of CO, nitrogen oxides and volatile organics in their area. Several sets of authors note that per vehicle emissions of all three of these materials are higher than in the United States or Europe (Zhang *et al.*, 1993; Tang *et al.*, 1995; Zhang *et al.*, 1995). Volatile organic to NO_x emissions ratios appear to be on the order of 10 in one study (Qin and Chan, 1993), but are undoubtably highly variable (NRC, 1991). Meteorological factors such as inversion heights and wind speeds are stressed as reasons for variability in pollution episodes (Xu and Zhu, 1993 and 1994). Traffic cycles do not seem to be uniform across the P.R.C., suggesting that perhaps cultural differences in work habits must be reckoned with. In Guangzhou, for example, a square wave in CO or NO_x emissions is observed reminiscent of Mexico City (Qin and Chan, 1993; MARI, 1994; McNair *et al.*, 1996). Shanghai, on the other hand, exhibits morning and evening NO_x peaks which may point to a rush hour phenomenon (Xu and Zhu, 1994). Cities in nearby Asian mainland countries record traffic patterns more like those of Shanghai (Moon *et al.*, 1995; Lee, 1995).

The secondary pollutant ozone is strongly in evidence in the Chinese urban areas reporting measurements. As in Western cities (MARI, 1994), concentrations can reach many tens of ppb but are seasonally dependent (Tang *et al.*, 1995; Xu and Zhu, 1994). In Beijing the highest ozone levels are recorded in the summertime. A recurring theme in the Chinese air pollution literature is concern over the growing number of motorized vehicles in major cities (Qin and Chan, 1993; Xu and Zhu, 1994; Tang *et al.*, 1995). Qin and Chan (1993) cite an annual rate of increase of over 25% in the number of autos and trucks in Guangzhou. A consensus is apparent that increased NO_x and hydrocarbon inputs will worsen the ozone pollution dilemma.

Aerosol composition has been studied in Chinese cities several times from the point of view of source receptor modeling (Mingxing, 1985; Dai *et al.*, 1986; Zhang

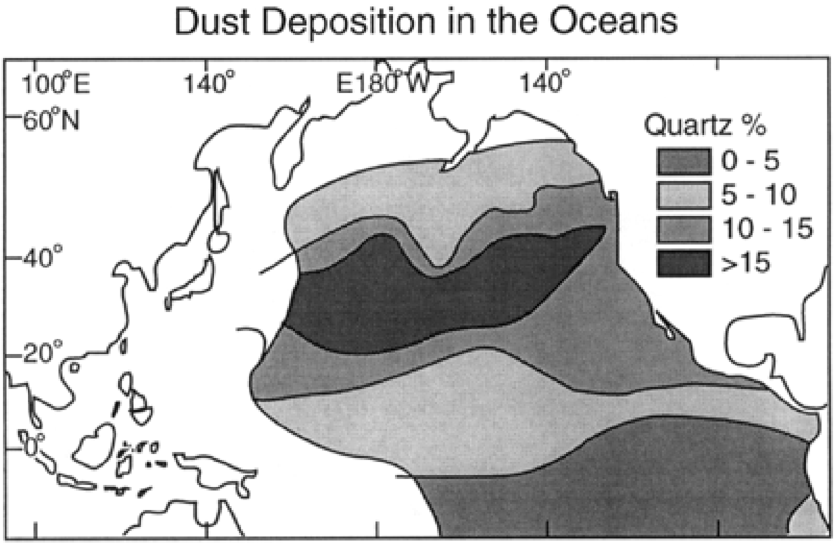

Figure 2. Mapping of the quartz distribution in Pacific Ocean sediments; a reflection of dust transport from central Asia with the prevailing westerlies (adapted from Windom, 1975).

et al., 1987; Zhu *et al.*, 1995). Particulate health effects have been a major concern recently to the Chinese and other world governments (UDC, 1982; WHO, 1985; Ross and Silk, 1987; Wu, 1989). Several authors have investigated the chemical composition of aerosols within major Chinese urban areas (Weiguang, 1981; Su *et al.*, 1991). Coal combustion has often been found to be a major source of the sulfate in particulates (Zhao and Sun, 1986; Zhu *et al.*, 1995). This is in accord with data indicating that coal has been until recently the major national energy source (Zhao and Sun, 1986; Smil, 1988). Natural dust and road dust are other major sources to the aerosol mass loading (Ji *et al.*, 1993; Zhu *et al.*, 1995). At least one Chinese group has undertaken numerical simulations of the urban aerosol (Xu *et al.*, 1992).

4. Asia-Pacific Air Mass Exchange

Major questions in the evaluation of any new Asian pollution source will concern influence on the pristine Pacific environment. Our consideration of transport of emissions from an impending Chinese automobile fleet will begin with a description of Asian winds and continent to ocean exchange. The wind systems could be characterized through several lines of reasoning. The well known Kosa dust storms, however, serve as an effective tutorial starting point (Iwasaka *et al.*, 1983; Hirose and Sugimara, 1984). They have long been studied as a major aeolian source of certain elements to the central Pacific (Ferguson *et al.*, 1970). Transport of dust minerals has been documented as far away as the Hawaiian Islands (Rex *et al.*, 1969; Shaw, 1980). Figure 2 typifies some of the early data (Windom, 1975).

In their studies of Kosa and other marine aerosols, Duce and colleagues (Duce *et al.*, 1980; Uematsu *et al.*, 1983; Duce *et al.*, 1991) emphasized the distinction between spring and summer flow patterns. In the spring, westerly winds blow steadily from China far into the Pacific. During the summer, northward expansion of the trade winds weakens the geostrophic flow and brings a halt to the regularity of the westerlies. The PEM missions studying the atmospheric chemistry of the Western Pacific have recently taken advantage of this situation. The initial PEM A campaign was conducted during the fall precisely because it is a period of minimal continental influence on the marine troposphere (Hoell *et al.*, 1996; Bachmeier *et al.*, 1996). Conversely, the as yet unpublished PEM B took place in the spring to maximize exchange.

We employ a simple approach here relying on meteorological trajectory analyses to define Asian winds. The Duce conclusions regarding seasonality of particulate transport were supported through a comprehensive suite of trajectory calculations (Uematsu *et al.*, 1983; Merrill *et al.*, 1985; Duce *et al.*, 1991). During the spring season, the transport paths deduced confirm that flow is steadily from the west, sometimes extending as far as the Marshall Islands. Kotamarthi and coworkers (Kotamarthi and Carmichael, 1990; Kotamarthi *et al.*, 1991) have constructed an idealized representation of springtime Asian air movements based on Merrill *et al.* (1985). In the Kotamarthi scenario, air travels from central Asia over populated areas of China, then Japan, and finally out over the Pacific. We will utilize a very similar idealization of flow in our two dimensional photochemical computations.

During the summer and fall, the transport situation can be much more complex (e.g. Bachmeier *et al.*, 1996). Chinese meteorological data have shown that there are at least some periods of steady westerly flow (Kondo *et al.*, 1987; Lamb *et al.*, 1990; Hong, 1993). We take the conservative position here that transport in summer/autumn is sufficiently intricate to require three dimensional simulations and realistic wind inputs. In winter prevailing winds may be largely westerly (Kondo *et al.*, 1987), but little photochemical activity is taking place.

5. Ozone Precursor Emissions

Tabulations of present day Asian nitrogen oxide emissions estimates are readily available in the literature (Kato *et al.*, 1991; Fujita *et al.*, 1991; Kato and Akimoto, 1992; Akimoto and Narita, 1994). In some cases the data are offered in conjunction with sulfur oxide fluxes. Information on the input of the volatile organics from the Asian continent is scarce. An exception which proves the rule is a report on terpene sourcing by Yuhua *et al.* (1995). While global NMHC emissions data bases necessarily enfold Asian values, the resolution is low and uncertainties are large (Muller, 1992; Graedel, 1994; Guenter *et al.*, 1995). In view of the lack of organic fluxes, we have elected to adopt some reasonable simplifications in constructing ozone precursor source estimates.

Our starting point is the NCAR Community Climate Model (CCM) NO_x input set developed by Dignon and Hameed (Dignon, 1991 and 1992; Hameed and Dignon, 1991). We choose the GCM gridded values in order to facilitate comparisons with other parts of the world, and especially the Eastern United States. A rough average nitrogen oxide flux of 1×10^{11} molecules cm^{-2} s^{-1} can be derived from the CCM gridding for the populous regions of China delineated in Figure 3. This will be termed $ENO_{x,c}$ here, signifying current nitrogen oxide emissions. The value is in broad agreement with the Asia specific works. Since we will be attempting to compute ozone production stemming from an as yet unbuilt fleet of automobiles and there are large uncertainties in the present vehicular contribution to the volatile organic budget, our default assumption will be that the latter is insignificant. In other words, we will postulate that natural organic inputs dominate. Although the Chinese transportation sector is growing rapidly, it is still small by Western standards. For natural background sourcing of hydrocarbons a composite of data from Yuhua *et al.* (1995), Singh and Zimmerman (1992) and NRC (1991) will be adopted where necessary. A lower limit will be used for the natural volatile organic flux, in order to permit highlighting of the upcoming vehicular ozone production. All natural hydrocarbons are inserted into the model as isoprene. The species is often employed as a surrogate under such circumstances (Liu *et al.*, 1987; Jacob and Wofsy, 1988, 1990; Pickering *et al.*, 1992a, b).

The U.S. analogy will be drawn upon for an estimate of ozone precursor generation by new Chinese vehicles. Note in Figure 3 that Eastern U.S. and Chinese areas of highest population density correspond in latitude and in surface area. Total automobiles and trucks in the eastern part of the United States can be derived from values for the number per family or person and standard demographic materials (United Nations, 1992; World Resources Institute, 1992; Cutter Information, 1992). The populous area shown in the Figure contains 50 to 75 million vehicles.The CCM grid nitrogen oxide emissions are roughly 2×10^{11} molecules cm^{-2} s^{-1} (Hameed and Dignon, 1991). About half of U.S. NO_x can be traced to automotive sources (NRC, 1991). Crudely, then, we will attach a value of 1×10^{11} molecules cm^{-2} s^{-1} to autos/trucks in the left portion of Figure 3. The flux from vehicles will in general be denoted $ENO_{x,v}$ here. It will be a major variable in our calculations. Clearly there are several large sources of uncertainty in the manipulations conducted above. They are both geographically and demographically informal. We will construct our ozone production arguments on a scaling basis, however, so that this particular source of error will be of little consequence. A square wave emissions function is applied in all cases.

The ratio of volatile organic carbon to nitrogen oxide emissions (moles C to moles N) from the automotive sector varies widely across the world from as little as 2 to as high as 20 (EPA, 1989; Placet *et al.*, 1990; NRC, 1991; MARI, 1994). Kotamarthi and Carmichael (1990) quote the value 5 for total NMHC to NO_x in present day China. In the U.S. the value is close to this level for both vehicular and integrated anthropogenic inputs (NRC, 1991). We adopt the Kotamarthi and

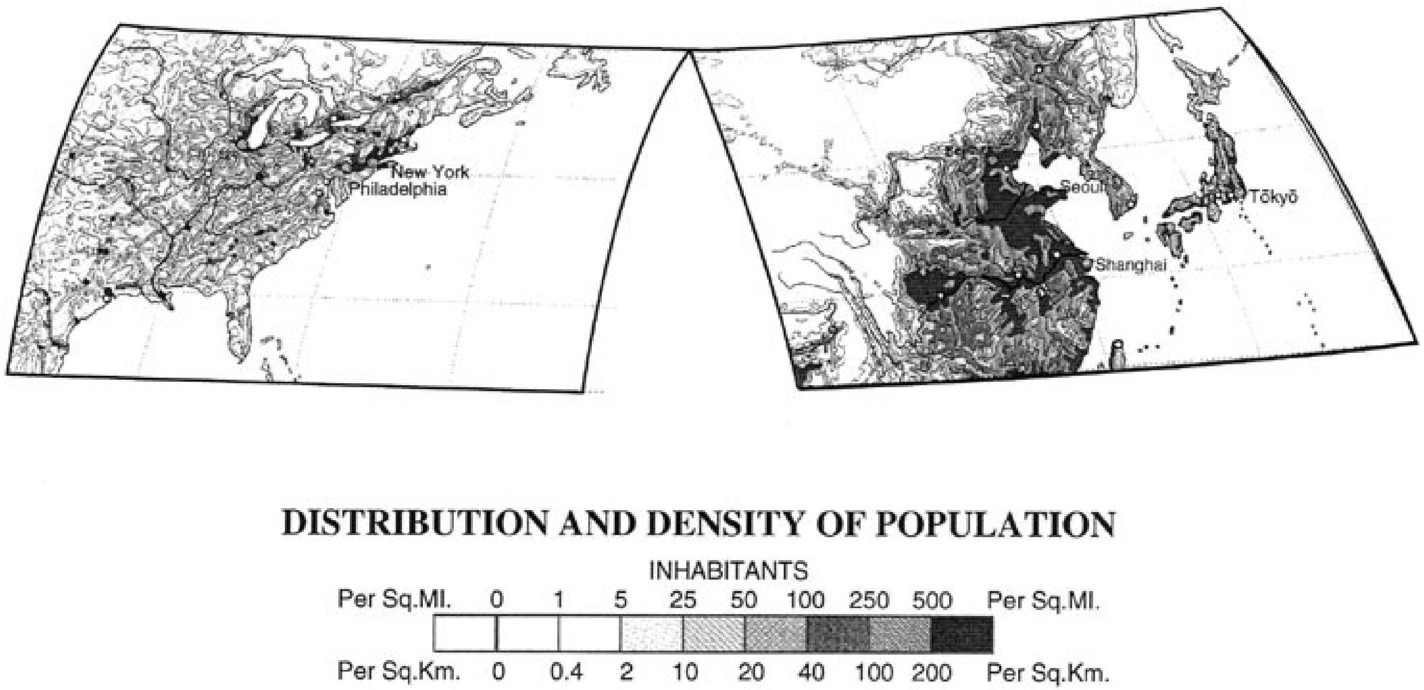

Figure 3. Population density contours for two regions of the middle latitude Northern Hemisphere – the Eastern United States and mainland China/Japan. Latitude increments indicated by the dashed lines are 10 degrees, longitude increments separated by dashed curves are 15 degrees (adapted from the London Times Atlas of the World, 1990 edition).

Carmichael ratio here but treat it as a variable parameter worthy of sensitivity testing. We partition our automobile organic emissions across the spectrum of model primary species according to morning measurements in several large cities worldwide (Mayrson *et al.*, 1975; Sexton and Westberg, 1984; Seiler *et al.*, 1989; MARI, 1994; Blake and Rowland, 1995).

6. Organic and Nitrogen Oxidation Yields

The ozone production of interest can be quantified on a preliminary and verificational level from regional scale yields which have been documented for precursor oxidation in the Western world. Inputs of hydrocarbons and nitrogen oxides are known for the United States (NAPAP, 1990; NRC, 1991) and outflow of ozone has been monitored (Anderson *et al.*, 1993; Parrish *et al.*, 1993; Berkowitz *et al.*, 1995). Many three dimensional models have been applied to organizing the ozone budget for large portions of the North American continent (Sillman *et al.*, 1990a, b; McKeen *et al.*, 1991a, b; Jacob *et al.*, 1993a, b). Useful discussions of overall oxidation yields have appeared from several valid viewpoints (Liu *et al.*, 1987; Lin *et al.*, 1988; Jacob *et al.*, 1989; Chin *et al.*, 1994).

Ozone production is most efficient per unit concentration of the nitrogen oxides at intermediate NO_x levels (Liu *et al.*, 1987). As titration points are reached for NO to dominate HO_2 or ozone in reaction with peroxy radicals, ozone production efficiencies of various hydrocarbon oxidation sequences maximize. The titration levels are generally in the 1 to 100 pptv range (e.g. Crutzen, 1988), but they vary with species, causing a smearing of what might otherwise be sharp transitions (Lin *et al.*, 1988). Increasing amounts of organic material enhance overall oxidation rates on both a mass basis (Jacob *et al.*, 1989) and because of raised hydrogen oxide and radical input rates (Crutzen, 1988), and this also causes stretching of the ozone production profiles. Eventually, NO_x begins to counteract these trends by converting the hydroxyl to nitric acid (Logan *et al.*, 1981). Large quantities of hydrocarbons will also suppress OH and reactive, unsaturated species such as isoprene will themselves consume ozone (Jacob and Wofsy, 1988 and 1990).

We will first paraphrase an organic oxidation yield standpoint. Among the simplest hydrocarbons in the atmosphere is methane. Its breakdown is initiated by hydroxyl radical attack, and in standard reaction mechanisms a total of five ozone molecules could be produced as the single carbon atom is converted to carbon dioxide (e.g. Logan *et al.*, 1981; Crutzen, 1988). As the number of carbon atoms increases moving toward larger organics, however, the per molecule ozone yield drops. Liu *et al.* (1987) argue effectively that a minimum for any one organic is two stoichiometric units of ozone. It is a simple matter to show, however, that yield should also scale with the chain length. Elliott *et al.* (1995b) derive a qualitative upper limit of three ozone molecules per carbon for several of the low molecular weight alkanes including ethane. Jacob *et al.* (1989) indicate that similar results can be obtained for unsaturated species. They then employ a kinetic box model to conclude that actual yields in the atmosphere are on the order of 1.5, and are relatively independent of the identity of the molecule. Reductions from the upper limit of three are attributable to storage of long lived intermediates such as ketones and alkyl nitrates. Jacob *et al.* (1989) also exclude the ultimate oxidation of CO to carbon dioxide from their analyses.

An alternative is to deduce ozone yields as a function of oxidation rates for the other major precursor set, the nitrogen oxides. Liu and coworkers both pioneered and promulgated this approach (Liu *et al.*, 1987; Lin *et al.*, 1988). McKeen *et al.* (1991a, b) and Jacob *et al.* (1993a, b) have applied the nitrogen oxide yields to continental ozone budgets, and Liu *et al.* (1987) have even extended them to the global scale. It is assumed that NMHC and NO_x levels are proportional to one another in a set of air parcels. As NO_x concentrations rise, ozone production P fails to track them in a linear fashion. Production can be normalized to NO_x as a matter of conceptual convenience, and we will denote then new variable as P'. Local nitrogen oxide concentrations can be expressed roughly as the product of emissions E and the lifetime T. The triple product $P'TE$ can then be seen either as production of ozone per unit NO_x ($P'T$) multiplied by emissions, or as P' multiplied by a concentration. The perspectives are distinct but both are valid. The former permits the estimation of ozone generation over large spatial scales given the nitrogen oxide input pattern.

We can now move to some sample computations, with the Eastern United States serving as a test case. During the early history of an air mass passing over the region the quantity $ENO_xR(VOC/NO_x)1.5$ should provide an estimate of ozone production from the organic yield perspective. The symbol R here indicates the volatile organic to nitrogen oxide emission ratio. The round value 5 is adopted for $R(VOC/NO_x)$ (NAPAP, 1990; NRC, 1991) and the overall ENO_x of 2×10^{11} molecules cm^{-2} s^{-1} is taken from the CCM gridded data (Hameed and Dignon, 1991). We assume westerly winds moving over a 1000 kilometer population corridor at 5 meters s^{-1} (Figure 3; Kotamarthi and Carmichael, 1990; Parrish *et al.*, 1993; Berkowitz *et al.*, 1995). Residence time over the developed zone is about two days, long enough for a significant fraction of reactive organics to oxidize. Total uptake of organic carbon atoms by the air mass is 2×10^{17} cm^{-2} and total ozone creation is 3×10^{17} molecules cm^{-2}. Relative to a 30 Dobson tropospheric column burden or an average of 40 ppb throughout the column (Logan *et al.*, 1981) this represents an addition of a few tens of percent. Parrish *et al.* (1993) report that pollution levels in surface air parcels traveling the great circle route from Washington D.C. to London are consistent with this analysis. Results in Anderson *et al.* (1993) show that the free Atlantic troposphere contains from 40 to 60 ppb of ozone depending on local pollution histories. Logan *et al.* (1981) give Northern Hemispheric free tropospheric averages of 40 ppb in winter and 60 ppb in summer. Quantification of ozone production over the Atlantic is of course a nontrivial matter, and actual sources may be quite difficult to identify. High ozone concentrations can result from stratospheric folding or long distance transport of oxidant rich air (Levy *et al.*, 1985; Oltmans and Levy, 1992).

Similar conclusions can be drawn from the standpoint of NO_x. Chin *etal.* (1994) summarize values which have been obtained for the ratio of ozone molecules produced to nitrogen oxide molecules consumed. In the parlance of Liu *et al.* (1987), this is the quantity $P'T$. Values for the Eastern half of North America range from about 5 to 10 in the summer. Annual variations are small down to 0.1 to 1.0 ppb NO_x. As oxidation rates and so also production P drop in the winter, the lifetime T rises and compensates. Both P and T are controlled largely by the hydroxyl. We will apply the summertime values from Chin *et al.* (1994) without adjustment. Some extremes in the summer $P'T$ are worth noting. Jacob *et al.* (1993a, b) obtain 6 for a national scale budget computation performed in a three dimensional model deriving from the GISS GCM. In interpreting the Jacob *et al.* model results, Chin *et al.* (1994) ignore dry deposition and infer a lower limit of 1.7. Olszyne *et al.* (1993) computed a value of 12 in the hydrocarbon rich Southeast. The reasons for and potential extent of such variability are outlined in Liu *et al.* (1987). Yields for the regional scale tend to cluster near 5. Our ozone production estimate is now $ENO_xP'T$ or 10^{12} molecules cm^{-2} s^{-1}. For the two day residence time over sources, we conclude that 2×10^{17} molecules cm^{-2} of ozone are generated. Another outlook is this: the total amount of NO_x incorporated into an air mass over the corridor is 4×10^{16} molecules cm^{-2}, and oxidation of each leads (through interactions with organics) to 5 ozone.

Asian meteorology bears real resemblances to that of the Eastern U.S., at least during seasons of westerly dominance (Duce *et al.*, 1980; Merrill *et al.*, 1985; Kotamarthi and Carmichael, 1990; Oltmans and Levy, 1992; Galloway *et al.*, 1992; Parrish *et al.*, 1993). The length of the Chinese population corridor is similar as well, although densities are much higher (Kotamarthi and Carmichael, 1990; Figure 3). In our North American calculations half of the ozone increase can be assigned zeroth order to automobiles since they produce half of the nitrogen oxides (NRC, 1991). Further, that half of the increase can be attributed to only about 50 to 75 million total vehicles. Given the Chinese fleet projections we are dealing with here, the potential for Asian ozone production is several times as large.

7. Ozone Production in an Idealized Two-Dimensional Model

The yields cited for precursor species in the Eastern United States may of course not translate directly to like latitudes on the Western Pacific Rim. No analysis of the Asian organic/NO_x/ozone relationsips has been put forward in the atmospheric chemical literature. Our crude estimates can to some extent be substantiated, however, through photochemical modeling. Several of us are presently at work constructing global scale tropospheric chemistry codes in general circulation model frameworks (*X.P.Z.*, *R.L.*, *C.Y.J.K.*, *R.P.T.*; see Elliott *et al.*, 1993b, 1995a, b; Shen *et al.*, 1996). Our strategy here will be to overview effects of the emerging Chinese automotive sector in a two dimensional framework. We will rely heavily on the highly idealized springtime dynamics described by Kotamarthi and Carmichael (1990).

The kinetics solver in our coding is adapted from a fast tropospheric photochemistry package which we have lately designed in order to meet the needs of the global modeling community. The package is documented in detail in the series Elliott *et al.* (1993a, b; 1995a, b) and Shen *et al.* (1996). Intercomparisons have been conducted against better established integrators. For example, Figure 4 displays daily ozone production at Niwot Ridge in the Colorado Rockies as computed in Liu *et al.*, 1987 and in Elliott *et al.* (1995a, b) for the same conditions. Several hydrocarbon combinations were assayed, including single carbon species alone, single carbons with natural NMHC represented by isoprene, and all NMHC including those in polluted air masses emanating from the Denver metropolitan area. Our kinetics package has found application recently in several one dimensional studies. For example, Shen *et al.* (1996) compare the influence of isoprene and the terpenoids on tropospheric chemistry at a suite of sites across North America. We list some salient properties of the chemistry module below.

Many traditional tropospheric kinetics models integrate the photochemical continuity equations through the linear multistep backward differentiation formulae (Byrne and Hindmarsh, 1987). Typical examples include the Gear code (Chang *et al.*, 1973), and the reverse Euler (Wofsy, 1978; Logan *et al.*, 1981; Kasting and Singh, 1986; Jacob *et al.*, 1989). While Jacobson and coworkers have recently succeeded in streamlining the Gear solver to the point where it may be feasible to insert into global three dimensional frameworks (Jacobson, 1994; Jacobson and Turco, 1994), a practical alternative to individual species integrations is offered by the family method (Brasseur and Solomon, 1984; Brasseur *et al.*, 1990). In the troposphere Crutzen and coworkers have been prominent practitioners of the application of family groupings (Crutzen and Gidel, 1983; Kanakidou *et al.*, 1991; Crutzen and Zimmerman, 1991). Our own family programs encompass several features which render them convenient for application in a variety of tracer transport schemes and in global scale three dimensional models. These include refinements in mechanism selection, numerics and optimization (Elliott *et al.*, 1993a, b, 1995a, b).

In addition to standard background oxygen, nitrogen, hydrogen and single carbon species kinetics (Crutzen, 1973; Fishman and Crutzen, 1978; Logan *et al.*, 1981), six primary nonmethane hydrocarbons have been selected and inserted to serve in the current work as surrogates for the overall tropospheric organic system (Elliott *et al.*, 1995a, b; Shen *et al.*, 1996). The primaries in the code are ethane, ethylene, propane, propylene, acetylene and isoprene. Full oxidation sequences are included for each. For example, the Paulson and Seinfeld (1992) mechanism has been adopted for isoprene. The choice of surrogates is typical for recent tropospheric modeling investigations (Liu *et al.*, 1987; Chatfield and Delany, 1990; Pickering *et al.*, 1992a). The total number of species in the model version used is around 100, and the total number of photochemical and photolysis reactions is over 200.

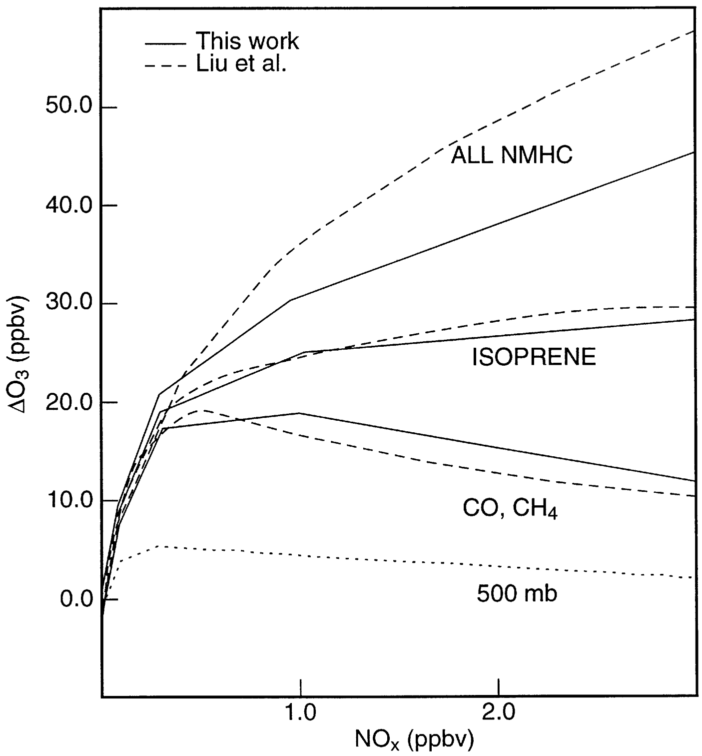

Figure 4. Comparison of ozone production in a code we have developed with results from established integrators. Dashed simulations are for the Niwot Ridg site (Liu *et al.*, 1987; Trainer *et al.*, 1987). The lone dotted curve labeled 500 millibars is an upper level calculation which matched Levy *et al.* (1985) values for the free troposphere over the open Pacific.

The two dimensional transport framework into which the photochemistry code is placed derives directly from Kotamarthi and Carmichael (1990) but is even more simplified. The grid consists of 17 one kilometer deep layers and 52 one hundred kilometer wide columns. Placement on the earth's surface is depicted in Figure 5. Merrill *et al.* (1985) springtime trajectories and the position of the Kotamarthi and Carmichael (1990) grid are shown for comparison. Columns 10 through 20 in our arrangement correspond roughly to highly populous areas of China, and also to areas of heavy vegetation. Columns 20 through 30 lie over the ocean en route to Japan, 30 to 40 over Japan itself extending from the western coastline to major cities of the east and to the Pacific. Horizontal transport is accomplished through the simple expedient of applying a forward Euler (Rood, 1987). Wind velocities are textbook averages for the spring and match the values in Kotamarthi and Carmichael (1990) closely; we begin the computations with an average of 5 m s^{-1} in the lower 5 km, and 15 m s^{-1} in the second 5 km (Merrill *et al.*, 1985). The horizontal wind field is ultimately treated as an important variable parameter. Vertical transport is simulated as pure eddy diffusion, with diffusion coefficients

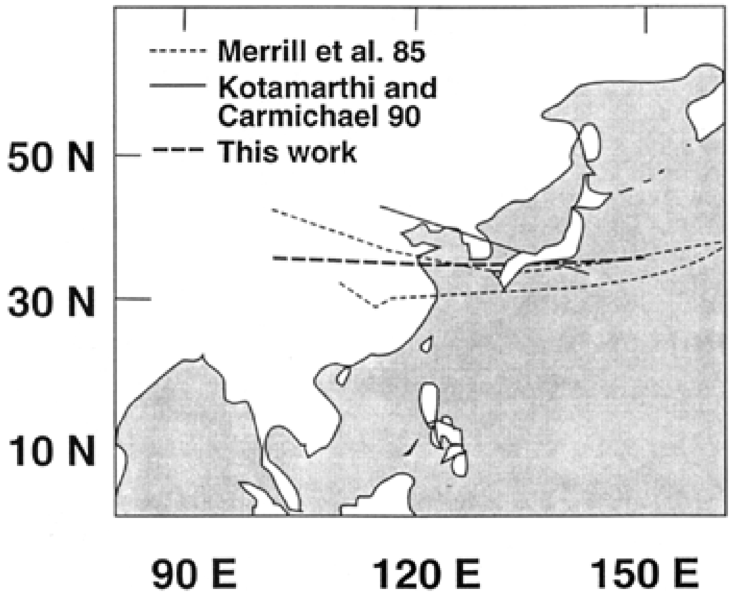

Figure 5. Springtime air mass trajectories and two dimensional model positioning over the Western Pacific Rim. Lighter dashed lines represent trajectory computations by Merrill *et al.* (1985) for parcels eventually reaching the Marshall Islands. The slanting straight line is a top down view of the model grid from Kotamarthi and Carmichael (1990), and the darker horizontal dashed line at constant latitude denotes the grid from the present work.

set as in early one dimensional tropospheric chemistry models (e.g. Thompson and Cicerone, 1982; Liu *et al.*, 1984). Deposition velocities are taken from Kotamarthi and Carmichael (1990) for continental and ocean surfaces. The radiative transfer driving photolysis is handled as in Elliott *et al.* (1995b). All computations are for clear sky, consistent with at least some local meteorological data (Wang *et al.*, 1993; Zhou *et al.*, 1994). Incoming solar radiation is attenuated first through Beer's Law absorption by molecular oxygen and ozone. The resulting intensities are then adjusted for multiple scattering.

Simulations begin on the vernal equinox and continue for 25 days with boundary conditions unchanging. The eventual results reported can be thought of as perpetual, equinoctial steady states. Initial concentration profiles for water vapor and ozone are independent of longitude and are taken directly from Logan *et al.* (1981) and Logan (1985). Background measurements from Kondo *et al.* (1987) are used to initialize the nitrogen oxides. Emissions of NO_x and the accompanying hydrocarbons are made exclusively into the lowest layer of columns 10 through 20. Some of the modeled ozone concentration data are given in Figure 6. We will focus our attention here on the profiles from column 40. The baseline simulations are driven by the $ENO_{x,c}$ from above and natural hydrocarbon emissions. An increase of 10 ppbv or so is computed over the lowest 5 km of the model regime.

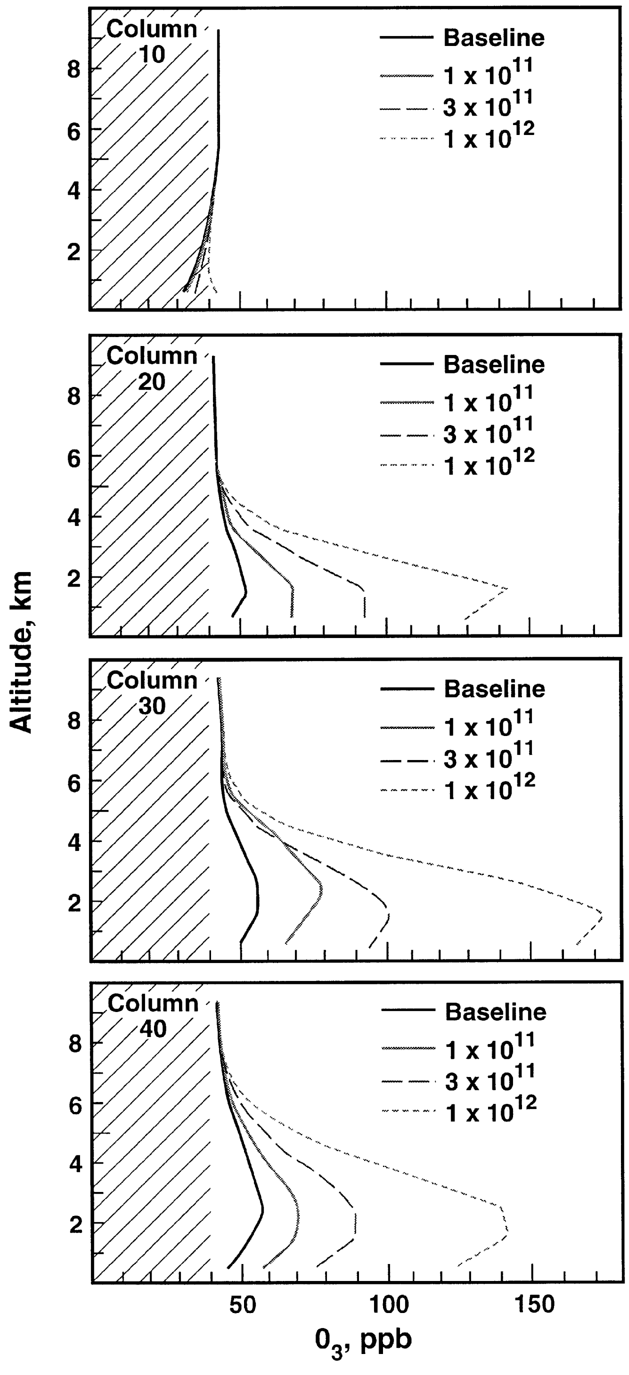

Figure 6. Vertical ozone concentration profiles at key locations from west to east across the two dimensional model. The fluxes in the legend are the quantity $ENO_{x,v}$ defined in the text. Units are molecules cm^{-2} s^{-1} of the nitrogen oxides.

There are two useful literature reference points for comparison with our baseline. In the most closely related model of chemistry in flow from the continent, Kotamarthi and Carmichael (1990) obtain buildup of ozone to 100 ppb in the lower 3 kilometers by the Osaka-Kobe area. This suggests that we have successfully established a lower limit, as intended. In the study of tropospheric ozone residuals via satellite, Sunwoo *et al.* (1992) detect 5 Dobson units more ozone in the 30 to 40 degree latitude band over the oceanic rim than in the surrounding regions. The populous Asian corridor generates about 15 percent more ozone than neighboring latitudes. The authors tentatively attribute the larger column density to anthropogenic activity. Our own column increase is on the order of ten percent. Since we do not attempt to include human hydrocarbon inputs, our results again seem reasonable.

We now describe the addition to our simulations of representations for the potential new vehicle emissions. The values of $ENO_{x,v}$ which we have chosen are listed in the legends for Figure 6. The increase in the lower tropospheric baseline is 10 to 15 ppb for the first increment in $ENO_{x,v}$ of 1×10^{11} molecules cm^{-2} s^{-1}. This is a little over 10 percent of the ozone column density. The result is consistent with our yield estimates/observations for the United States east coast. As a further check, we have asked the program to compute the per carbon and nitrogen atom efficiencies over the first 40 boxes in the grid. The values obtained are listed in Table I. Agreement with the literature on oxidation yields is fairly satisfactory (Liu *et al.*, 1987; Jacob *et al.*, 1989; Jacob *et al.*, 1993a, b; Chin *et al.*, 1994). The figures are higher for the baseline run because of the high reactivity level of isoprene, our surrogate for all natural NMHC. The yields near 3 for the nitrogen oxides are a bit lower than those of Jacob and co-workers for the Eastern U.S. (around 4; 1993a, b; Chin *et al.*, 1994). There are several potential explanations for the discrepancy. Our idealized and very direct horizontal transport may be moving organic oxidation intermediates such as carbonyls and nitrates to the Pacific before they can make a contribution to overall production. It may also be that our ozone deposition rates are overestimated. In either case, however, indications are that we have computed lower limits to overall ozone production from the new transportation sector.

A set of sensitivity calculations has been performed to accompany the major runs. Figure 7 summarizes the conclusions. The $ENO_{x,v} = 3 \times 10^{11}$ molecules cm^{-2} s^{-1} flux serves as the basis for the comparisons. It is shown as the solid curve in the figure. Two crucial variables underlying the calculations were treated as uncertain parameters. First, the R(VOC/NO_x) for the potential Chinese automobile fleet was raised from 5 to 15. The organic to nitrogen oxide ratio is a perennial uncertainty, and particularly so in Asia. Because of the standard tropospheric chemical nonlinearities, ozone production over and above the $ENO_{x,c}$ level did not triple. Tests of lower R(VOC/NO_x) were also conducted to confirm that the effects on ozone would lie in the opposite direction. Next we increased the horizontal wind velocity in the lower few kilometers from the initial value 5 m s^{-1} to 10 m s^{-1}. Ozone production was significantly reduced under these circumstances.

Table I. Ozone production efficiencies from the idealized two dimensional model of photochemistry in air flowing from the Asian continent. Yields are derived relative to both the number of organic carbon atoms injected and the number of nitrogen oxide molecules. The new vehicular nitrogen oxide fluxes are given in units of molecules cm^{-2} s^{-1}. The null emission is the baseline, which includes only existing NO_x fluxes and natural hydrocarbons

$ENO_{x,v}$	Efficiency relative to carbon	Efficiency relative to nitrogen
0×10^{11}	2.5	8.6
1×10^{11}	1.2	3.4
3×10^{11}	1.2	3.3
1×10^{12}	1.2	3.3

Table II. Ozone production efficiencies from the idealized two dimension model as exhibited during sensitivity tests. Nitrogen oxide emissions $ENO_{x,v}$ are set constant at 3×10^{11} molecules cm^{-2} s^{-1}. The first line in the table is a repetition of the run already displayed in Figure 6 (Table I). Volatile organic fluxes and near surface winds are then multiplied alternately by factors of three and two. R has the meaning R(VOC/NO_x) as defined in the text, U is an average horizontal wind speed in units meters s^{-1}

$ENO_{x,v}$	R	U	Efficiency: carbon	Efficiency: nitrogen
3×10^{11}	5	5	1.2	3.3
3×10^{11}	15	5	1.4	10.3
3×10^{11}	5	10	2.0	6.1

Efficiency computations corresponding to those in Table I are collected in Table II. Experiments were run on several partitioning strategies for the natural and vehicular organics. The major conclusions are not altered by the organic runs and so the results are not shown here. The key sensitivity tests can be interpreted as follows: (1) raising of the volatile organic input enhances the NO_x efficiency mainly because there are more hydrocarbon moieties available to oxidize (2) at higher surface wind speeds, overall nitrogen oxide concentrations are reduced because of shortened advective residence times, and organic efficiencies rise slightly due to the standard nonlinearities.

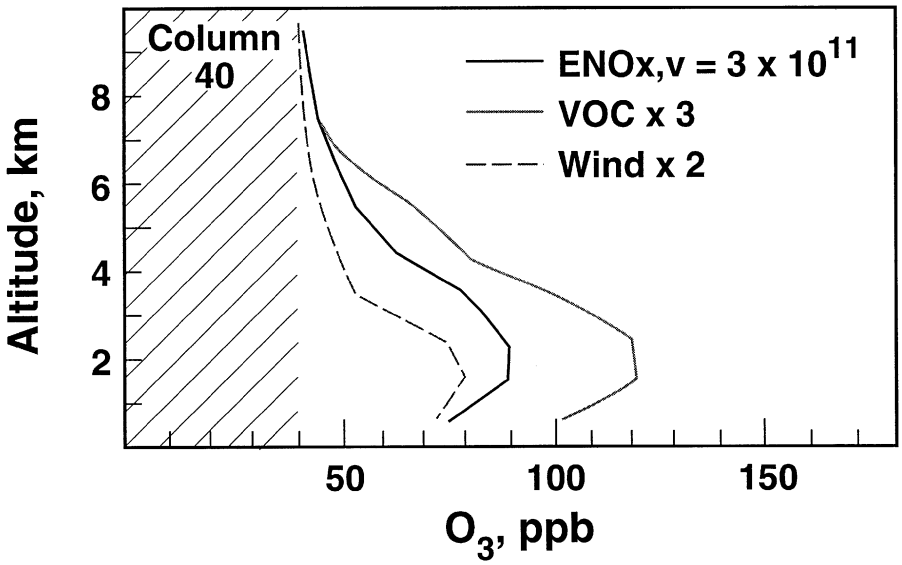

Figure 7. Vertical ozone concentration profiles at column 40 in the $ENO_{x,v} = 3 \times 10^{11}$ molecule cm^{-2} s^{-1} emissions scenario, in the original model configuration (darker solid curve), and with the volatile organic flux and horizontal wind velocities multiplied in turn by factors of three and two.

8. Nitrate and Particulate Production

Automobiles in the western world are considered major emitters of the nitrogen oxides, but not of the sulfur oxides (United Nations, 1992; Russell *et al.*, 1993; MARI, 1994). The latter are thought to derive mainly from stationary inputs (Chang *et al.*, 1987; NAPAP, 1990; Sillman *et al.*, 1990a). Coal combustion is the dominant source of atmospheric sulfur in China today (Smil, 1988; Sinton *et al.*, 1992). We begin our brief assessment of aerosol effects with the assumption that potential Chinese vehicles of the next century will add significantly to gaseous nitrogen but not to sulfur budgets. There could of course be feedbacks to sulfur if coal were burned to sustain expanded automobile-related industries. The nitrogen oxide species can contribute to urban aerosol mass loadings after they have reacted with hydroxyl radical to give nitric acid, which is relatively involatile.

Pollutant nitrate is detectable in the Northern Hemisphere well into both the Pacific and Atlantic Oceans (Duce *et al.*, 1991; Galloway *et al.*, 1992). Prospero and Savoie (1989), for example, note co-seasonality of nitrate collected on Whatman filters with Kosa dust across the SEAREX Pacific Island network. It can be deduced that over half the nitric acid and nitrate ion sampled by the filters is continental in origin, and the authors note that most continental sources are thought to be anthropogenic. The nitrogen oxide emissions adopted in the photochemistry section imply direct impacts upon nitric acid and nitrate pollution levels in the open Pacific. They can be assessed preliminary through a set of fundamental budget calculations.

The initial $ENO_{x,v}$ value of 1×10^{11} molecules cm^{-2} s^{-1} can be used to demonstrate. In the spring season the contact time of a typical air mass with a 1000 km emissions zone on a continental east coast is on the order of two days (Merrill *et al.*, 1985; Kotamarthi and Carmichael, 1990). Accumulation of NO_x is then approximately 2×10^{16} molecules cm^{-2}. Logan *et al.* (1981) calculate an average lifetime of one day for conversion of the NO/NO_2 couple to nitric acid. Results in recent global models of nitrogen species distributions are consistent with the figure (Penner *et al.*, 1991). We will assume here that NO_x is converted rapidly to HNO_3. Mixing depths for pollutants traveling to the Pacific Rim are on the order of 3 kilometers in Kotamarthi and Carmichael (1990), and 5 km given global average tropospheric diffusion coefficients (Thompson and Cicerone, 1982; Liu *et al.*, 1987). About half the tropospheric column integrated concentration of 2×10^{25} molecules cm^{-2} is contained in the lowest five kilometers of the atmosphere. Even in our lowest $ENO_{x,v}$ situation, the potential exists for the addition of several ppb of nitrate to the continental tropospheric chemistry system which influences the open Pacific. This would represent a significant increase relative to the latest measurements at the Rim (Kondo *et al.*, 1987; Kondo *et al.*, 1996; Koike *et al.*, 1996) and also against the nitric acid/nitrate aerosol data from remote island stations (Prospero and Savoie, 1989; Duce *et al.*, 1991).

Automobiles are well known for production of soot and organic aerosols, and the role of these substances in radiative forcing of climate is very uncertain (Seinfeld, 1986; Aldape *et al.*, 1993; Miranda *et al.*, 1994; United Nations, 1992; Novakov and Penner, 1993; Penner *et al.*, 1994). Parungo *et al.* (1994) have measured pollutant levels of black carbon at a series of sites extending from populated regions of mainland China out into the ocean. Unless a large future vehicle fleet is quite well maintained increases in soot transport are to be expected (Zhang *et al.*, 1995; Jiqun *et al.*, 1995). The situation for fugitive dust is similar. China currently has few paved roads or highways upon which to operate its upcoming fleet (World Resources Institute, 1992; World Bank and Chinese Ministry of Communications, 1994; Brown, 1995). Construction of new vehicles is likely to long precede new roadways; congestion is a norm in burgeoning Asian cities such as Bangkok and Singapore (Sinton *et al.*, 1992; Sathaye *et al.*, 1994). The effects of the central Asian dust storm phenomenon upon populated regions of the country (Zhou *et al.*, 1994; Zhu *et al.*, 1995) and upon the open Pacific (Rex and Goldberg, 1958; Duce *et al.*, 1980, 1991) are well documented. A few emissions inventories and aerosol studies have already hinted that fugitive dust increases may accompany increasing vehicle traffic (Ji *et al.*, 1993; Zhu *et al.*, 1995).

9. Carbon Dioxide and Global Climate

In this section we place emissions of a potential twenty first century Chinese automotive fleet in the context of global climate change and the major greenhouse gas, carbon dioxide. Even when maintenance procedures are substandard, most of

Table III. Annual carbon dioxide emissions to the global atmosphere from the combustion of major fossil fuel types, for years close to 1980 (Marland and Rotty, 1984). Gaseous fuels include natural gas, liquids are mainly crude oil and solids encompass coals with a wide range in quality. Units are millions of metric tons of carbon per year. Values are rounded to the nearest hundred

	Gases	Liquids	Solids	Oil flares	Total
10^6 ton C	700	2300	1900	100	5000
Percent	14	46	38	2	100

the gasoline used as fuel in automobiles is converted rapidly to CO_2 (Zhang *et al.*, 1995). The carbon dioxide molecule itself comprises the majority of emissions. The exhaust fraction of carbon monoxide is dictated to a large extent by thermodynamics within the engine block (Seinfeld, 1986), but CO is in any case oxidized by the hydroxyl radical to CO_2 within about a month in the troposphere (Logan *et al.*, 1981). The fraction of unburned organics is small (on the order of a percent or less; Marland and Rotty, 1984). From the standpoint of terrestrial climate, a major impact of the internal combustion engine is to generate carbon dioxide.

Early carbon budget studies demonstrated that about half of all the CO_2 emissions stemming from fossil fuel use could be traced to crude oil production and consumption (Keeling, 1973; Marland and Rotty, 1984). For example, a global emissions breakdown for the average of several years before and after 1980 is presented in Table III. In the same data bases it is concluded that in developed countries, half of all crude oil is generally consumed by the transportation sector. Over the several decades for which reliable energy statistics were available at the time (1950–1980), the major change in this overall distribution was a slight relative drop in the importance of CO_2 generation from coal/solid fuels. More recent analyses tell essentially the same story (Lashof and Tirpak, 1990; Cutter Information, 1992). One quarter of fossil fuel carbon dioxide emissions to the atmosphere can be attributed to the energetics of transport. Of this quarter, close to 90 percent are to be connected with surface vehicles (Cutter Information, 1992).

Absolute automobile registration figures in different countries/regions have been compiled by several groups (Motor Vehicle Manufacturer’s Association, 1990; World Resources Institute, 1992). The number of vehicles is in fact the most convenient scaling factor for our purposes here. The automobile total for the United States is now about 140 million, which is consistent with the estimates proffered earlier for eastern areas. The number of automobiles registered for operation globally was close to 400 million in the late nineteen eighties. Vehicular transportation is growing only slowly in developed countries, and their numbers should be only 10 to 40 percent larger by the year 2050 (DOE, 1990). The projections for Chinese auto production which we are adopting here suggest a real enhancement of the global fleet in the near future. Each hundred million vehicles coming on line will increase the world total for the next century by about one fifth. At current mileages (Cutter Information, 1992), the associated gasoline consumption will add perhaps 5 percent to the carbon dioxide inputs from fossil fuel combustion.

We will not attempt to speculate on any specific global climate effects which it may be possible to assign to the upcoming Chinese fleet. A common yardstick for assessing climate change is CO_2 doubling. Such levels should be reached by the end of the twenty first century, and will lead to a warming of two degrees centigrade or so averaged over the earth's surface (NAS, 1983b; Houghton *et al.*, 1990). Fossil fuels are by far the major anthropogenic carbon source (Cline, 1992). Clearly the contribution from an upward ramping of the automotive industry in the People's Republic would be significant. It is also of interest here that greenhouse gases other than carbon dioxide produce a radiative forcing which is as large as that of CO_2 alone (Ramanathan *et al.*, 1987; Ramanathan, 1988; Lacis *et al.*, 1990; Kiehl and Briegleb, 1993; Hauglustaine *et al.*, 1994). The nitrogen oxides, volatile organics and ozone derived from vehicle use all qualify as trace level forcers.

10. Discussion

Several lines of reasoning have been presented suggesting that a major buildup in China of vehicles constructed by western standards would entail increases in tropospheric pollution levels. We now address some policy relevant concerns which follow rather directly from the atmospheric chemistry investigation. Most critically, it is noted that the surface transportation system for which the People's Republic appears to be striving may not be attainable. Questions are sketched regarding the approach to and provision for a large, steady state vehicular fleet in China. It is also the case that novel design options will be open to the automotive engineers of the P.R.C. For example, we touch upon the kinds of alternative fuel which it may be feasible to rely upon. Some peripheral environmental pollution issues are interwoven with this discourse. The effects of increased air traffic upon the Pacific free troposphere are mentioned in order to provide a future perspective from another of the transportation industries. To conclude, the uncertainties underlying our development are evaluated.

Our preliminary estimates of the size of the impending Chinese automotive fleet derived from two main sources. Independently operated international trade journals have stated that the P.R.C. will be able to produce several million cars and trucks per year by the year 2000 (Brown, 1995; Engardio and Roberts, 1996). The logic is based on rates at which bicycle, tractor and truck production have been ramped in the past. In fact the figures range from two to four million per year. A linear extrapolation over several decades leads readily to total of 50 to 100 million vehicles. The second source of information is the Chinese government itself. The Ministry of Communications, which contains counterparts to western departments of transportation, forecasts even larger fleet numbers (World Bank and Chinese Ministry of Communications, 1994). In one important sense, the projections are of course eminently defensible. At the Western ratio of half an automobile per person (Cutter Information, 1992), the populace of the P.R.C. could instantly create demand for half a billion cars (World Resources Institute, 1992; Sinton *et al.*, 1992; Sathaye *et al.*, 1994; Tyler, 1994; Barnathon *et al.*, 1996).

The automobile fleet in the United States currently totals 140 million and averages less than 10 years of age (Cutter Information, 1992; United Nations, 1992). Under a steady state approximation the sum of automobile production and imports is thus on the order of 10 million per year or greater. Clearly a Chinese vehicle total of more than one hundred million would require large productions increases beyond the year 2000. Even if cars and trucks could be kept on the road for a long average lifetime of a decade, the projections for early next century do not support 100 million units. Further expansion in the Chinese industrial infrastructure would be required to yield the augmented production, and new pollution problems are implied. Coal combustion is now the major source of energy to the P.R.C. and contributes heavily to sulfur oxide loadings and acid rain (UDC, 1982; Zhao and Sun, 1986). Increased coal consumption might be needed to sustain enlarged steel and rubber industries.

Continuing for the moment under the assumption that additional autos would be gasoline powered, it is relevant to examine the reserves of crude oil available to the Chinese (Smil, 1988; Sinton *et al.*, 1992). Government reports have been inconsistent in this area. Production is now concentrated on oil shale deposits in the Northeast of the country, but extraction with water has become increasingly difficult. Exploration of offshore fields is proceeding through foreign contractors. ARCO is in operation off Hainan, for example. The western desert may harbor significant reserves, but government comments on this have been difficult to interpret. The most useful independent estimate of late has come from experts at British Petroleum. They are willing to say only that Chinese recoverable reserves are equal to or larger than those of the United States.

Rising personal automobile usage has traditionally led to congestion in developing Asian cities, but a major response has often been to limit driving habits, as opposed to construction of new roads (Sathaye *et al.*, 1994; Smith *et al.*, 1994). In the cases of the island nations of Hong Kong and Singapore this is in part the result of a lack of space. The conclusion extends, however, to mainland cities such as Bangkok and Beijing, and to some extent to Manila (Lodge, 1992). New types of taxation and penalties sometimes cause vehicle registration levels to fall (Sathaye *et al.*, 1994). Nevertheless, history teaches that the transition to private autos will be difficult to prevent (Sathaye *et al.*, 1994; Goldemberg, 1995). The prospects for large scale pollution increases will then act as a motivation to investigate alternative fuel supplies.

Alcohol fuels could be produced from the substantial Chinese coal deposits. Methanol has been considered as a means for reducing domestic auto emissions (Carter, 1986; Russell *et al.*, 1990). Alcohol has actually been employed/tested in several megacities of the Americas (Szwarc and Branco, 1985; DeAndrade and

Miguel, 1985; Williams, 1989). Aldehyde pollution has been a noteworthy side effect. Another leading contender for automotive fuel of the future is natural gas (NRC, 1 991; Cutter Information, 1992). Global reserves are greater than those for oil. Indonesia possesses vast holdings, but the Chinese do not. The volatile organics produced/emitted during natural gas combustion are relatively unreactive. Furthermore, over the integrated production and distribution cycle it may in the long run prove cheaper to use than gasoline. Hydrogen has drawn some study as well (Cutter Information, 1992; DeCicco and Ross, 1994), but burns hot and so produces substantial amounts of the nitrogen oxides. In many parts of the world the combination of NO_x with natural NMHC inputs would still lead to ozone production (NRC, 1991). Storage, production and distribution of H_2 fuel all offer difficult engineering challenges. Electric cars are actually on the market now in California, but serious lead pollution problems have not been adequately addressed (DeCicco and Ross, 1994; Lave *et al.*, 1995).

It is worthwhile to consider whether the vehicle fleet in the P.R.C. is likely to be well maintained (NRC, 1991; MARI, 1994). Remote sensing studies demonstrate that engine upkeep may be the single most important factor in determining the level of automotive emissions (Bishop *et al.*, 1989; Lawson *et al.*, 1990; Beaton *et al.*, 1992; Zhang *et al.*, 1995), outweighing control technologies and age in that regard. Scandinavian countries tend to have strong records for vehicle service. The centralized economies of Eastern Europe, on the other hand, were known for polluting vehicles during the Communist era (Klemm and Schaller, 1994; Grosse, 1995; Langman and Graf, 1995). It is difficult to predict the level of maintenance a Chinese fleet will receive. Trade journals and other sources point out that the quality of Chinese products in high technology industries has been high in the last few years (Sinton *et al.*, 1992; Brown, 1995; Barnathon *et al.*, 1996; Engardio and Roberts, 1996).

Studies of aircraft effects on tropospheric chemistry have developed in parallel with urban pollution research. Early work focused on nitrogen oxide injections into the lower stratosphere from supersonic transports (Johnston and Podolske, 1978; Johnston *et al.*, 1989). It was thought that catalytic cycling involving the simple nitrogen oxides could efficiently remove stratospheric odd oxygen (Johnston and Podolske, 1978; Brasseur and Solomon, 1984). Interest later shifted toward the upper troposphere, where enhanced nitrogen oxide seems likely to increase ozone production (Hidalgo and Crutzen, 1977; Liu *et al.*, 1980; Derwent *et al.*, 1982). Ozone acts as a greenhouse gas in the lower stratosphere and upper troposphere. Radiative forcing of surface temperature is much more sensitive to aircraft than to surface inputs of oxidized nitrogen (Johnson *et al.*, 1992; Beck *et al.*, 1992).

A good deal of research activity into subsonic aircraft emissions has been spurred by the recent NASA subsonic assessment program. Attempts have been made to quantify aircraft related pollutant inputs into the upper troposphere (Baugham *et al.*, 1993). Growth in aviation transport is anticipated in all the major flight corridors including those running over the remote Pacific. Injections of NO_x along the corridors leads not to local but to hemispheric scale alterations in chemistry. Three dimensional photochemical modeling efforts indicate ozone concentrations could rise on the order of 5 to 10 percent by the middle of the next century, relative to a hypothetical aircraft-free situation (Brasseur *et al.*, 1996). This is to be contrasted with surface increases over Asia and the western Pacific of tens of ppb.

We conclude our discussion with a listing of some major uncertainties in our work. Although People's Republic policy statements are consistent with an intent to create a surface transportation system of one hundred million or more automobiles (World Bank and Chinese Ministry of Communications, 1994), and although trade journals offer evidence that the necessary production capacity is within reach (Brown, 1995), the fleet remains entirely a speculation. To support it large production increases must be assumed early in the next century, and an ability to cope with several types of new pollution. Our estimates of ozone production by new vehicles, whether based on precursor oxidation yields or on full photochemical modeling, imply several potential sources of error. Volatile organic to nitrogen oxide ratios remain uncertain for present day Asian emissions, and are even more so in the future. Asian meteorology has been idealized in several ways in our manipulations, whether through the procedure of drawing on an analogy with eastern North America, or by concentrating on synoptically simple times of the year. Maintenance and vehicle design assumptions are open to question. There are indications that the Chinese will build quality cars and trucks, just as they have computers and other high technology products of late. It is also true, however, that large centralized economies have not fared well in the design and engineering of automobiles.

11. Summary

The People's Republic of China constitutes the world's largest underexploited marketplace for automobiles (World Resources Institute, 1992; Sathaye *et al.*, 1994). The government has signaled an intent to dramatically enlarge the surface transportation sector (World Bank and Chinese Ministry of Communications, 1994) and the international press estimates that production of several million new vehicles per year may be possible in China early in the next century (Tyler, 1994; Brown, 1995; Barnathon *et al.*, 1996; Engardio and Roberts, 1996). It is conceivable that one hundred million or more new cars and trucks may operate on the roads of the P.R.C. within a few decades. Experience in the Eastern United States, Europe and Japan suggests that a fleet of this size will create new regional pollution problems and add to the pace of global change (Marland and Rotty, 1984; Hough and Derwent, 1990; Akimoto *et al.*, 1993; Jacob *et al.*, 1993a, b). We have here quantified some of the large scale eff ects on the chemistry of the troposphere, and consider some related policy matters as well.

Our focus has been upon the rise in regional ozone which is likely to follow a massive Chinese automobile buildup. Ozone increases are first constructed following U.S. studies of yields from the major precursors the organics and the nitrogen oxides (Chin *et al.*, 1994). The increases are then corroborated through two dimensional modeling of the photochemistry taking place in air flow off the Asian continent. In either case we conclude that several tens of ppb of ozone over and above the present baseline are likely to result. Alteration in tropospheric aerosol fields and carbon dioxide loadings are anticipated as well. We estimate the column increase in nitric acid, an involatile partitioned to a large extent into particle phases over the open Pacific (Duce *et al.*, 1991). Soot is already considered a pollutant in the pristine Pacific marine environment (Parungo *et al.*, 1994) and poorly maintained vehicles will contribute to the problem. The addition of fugitive dust from unpaved roads is considered (Ji *et al.*, 1993; Zhu *et al.*, 1995). By scaling against the extant global fleet (Cutter Information, 1992), it is calculated that twenty first century Chinese vehicles could add several percent to the CO_2 input from fossil fuel combustion.

Accompanying all these manipulations we perform cursory reviews of the scholarly literatures regarding Asian background tropospheric chemistry (e.g. Sunwoo *et al.*, 1992), urban air pollution in China (e.g. Qin and Chan, 1993), and the meteorology of the western Pacific Rim (e.g. Merrill *et al.*, 1985). Our two-dimensional photochemistry simulations borrow from these data bases to construct initial concentration distributions, trace gas emissions, and wind fields. In particular, springtime meteorological conditions were identified which permit highly idealized treatment of chemistry and tracer transport together (Kotamarthi and Carmichael, 1990). Pollutant NO_x and volatile organic inputs were constructed by extrapolation of U.S. data since Asian vehicular and hydrocarbon studies are lacking.

In a closing discussion section, some important peripheral questions are addressed from a policy standpoint. We ask first whether the large projected Chinese automobile fleet would in fact be sustainable. Increases in production would be required to and beyond current U.S. levels (Cutter Information, 1992). It is noted that oil reserves in the P.R.C. are quite uncertain (Sinton *et al.*, 1992) and that the congestion/pollution problems implied could well inspire the adoption of alternative fuels for Chinese vehicles (Sathaye *et al.*, 1994). Alcohols, natural gas, and hydrogen are all mentioned as potential energy sources and electric designs are considered as well. Maintenance practices are cited as a key to emissions control (Zhang *et al.*, 1995). Projections of free tropospheric ozone production by subsonic aircraft are quoted for the middle of the next century (Brasseur *et al.*, 1996). The purpose is to provide some perspective on our surface transportation related computations. Finally, uncertainties in our manipulations are detailed. Ratios of the ozone precursor types (Yuhua *et al.*, 1995), idealized meteorology (Kotamarthi and Carmichael, 1990), and vehicle design assumptions (Cutter Information, 1992) all constitute potential sources of error. Moreover, the large fleet sizes treated in the text remain largely a speculation.

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