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Abstract

Diesel exhaust is now one of the largest sources of nitrogen oxide (NO_x) emissions, and also contributes significantly to emissions of fine particulate black carbon (soot). The California Air Resources Board (CARB) recently adopted a rule to requiring that all in-use trucks operating in California meet current (2007) exhaust PM emission standards by 2014. This will require retrofit or replacement of older in-use engines, with emission reductions occurring on an accelerated schedule compared to what would be achieved by fleet turnover alone. A side effect of using diesel particle filters (DPF) to control exhaust PM emissions is that the NO_2/NO_x ratio in exhaust emissions may increase to ~35%, in comparison to lower values (~5%) typical of engines without particle filters. We evaluate the effects of increased deployment of DPF systems as well as NO_x controls on air quality in southern California using an Eulerian air quality model. Compared to a 2014 reference scenario without the retrofit program, we found black carbon concentrations decreased by 12±2% and 14±2% during the summer and fall, respectively, with corresponding increases in ambient ozone concentrations of 3±2% and 7±3%. NO_2 concentrations decreased by 2-4% despite the increase in primary NO_2 emissions – because total NO_x emissions were reduced as part of a parallel but more gradual program to retrofit NO_x control systems on in-use engines. However, in some cases, NO_2 concentrations were found to increase at locations with high diesel truck traffic. Increases in fine particulate nitrate of 3-7% were predicted near downtown Los Angeles.

Introduction

Heavy-duty diesel engines are an important source of air pollution on urban, regional, and national scales (Lloyd and Cackette, 2001a; Sawyer et al., 2000; Yanowitz et al., 2000). Although they account for only 2% of the on-road vehicle fleet, and 4% of the vehicle-km traveled, heavy-duty trucks are responsible for over half of nitrogen oxide (NO_x) and exhaust particulate matter (PM) emissions from on-road mobile sources (Kirchstetter et al., 1999). The importance of diesel engine emissions has been growing since 1990 due to success in controlling light-duty gasoline engine emissions, the failure of early efforts to control diesel NO_x emissions, and a threefold higher growth rate of diesel fuel sales compared to gasoline (EIA, 2006; Harley et al., 2005).

In contrast to earlier diesel emission control strategies that relied on combustion modifications and improved engine designs, current control strategies now typically include exhaust after-treatment (Johnson, 2006). Diesel particle filters (DPF) include a platinum catalyst upstream of the particle filter; the catalyst promotes oxidation of nitric oxide (NO) present in the exhaust to nitrogen dioxide (NO_2). NO_2 in turn is used as an oxidizing agent to remove carbon particles from the filter to prevent it from plugging. Urea-based selective catalytic reduction (SCR) is a possible approach to controlling NO_x emissions from heavy-duty engines. A reason for using urea instead of ammonia is that urea can be stored in liquid form and is safer and easier to handle. Controls such as DPF and SCR systems have not yet been widely deployed on diesel engines in the U.S. Nationally, new heavy-duty diesel engines must meet more stringent exhaust PM and NO_x emission limits starting with the 2007 and 2010 engine model years, respectively.

However, because of the long service life and slow rate of turnover for heavy-duty engines, the California Air Resources Board (CARB) has adopted an in-use emissions control rule that requires heavy-duty vehicle owners to retrofit or replace older engines (CARB, 2008a). This program is intended to accelerate reductions in diesel exhaust emissions that would otherwise occur more gradually due to fleet turnover, as shown in Figure 1. As a result, on-road heavy-duty diesel trucks operating in California are expected to meet current new-engine exhaust PM emission standards by 2014. The adoption of best available control technology for NO_x will proceed at a slower pace. It is estimated that by 2014, half of the in-use trucks operating in California will meet current new-engine emission standards for NO_x (CARB, 2008a).

While diesel emission regulations are technology neutral, and various approaches are possible, SCR systems are a viable option for retrofit on in-use engines to reduce NO_x emissions (CARB, 2008a). Other approaches to diesel NO_x emission control include lean NO_x traps (LNT) that react NO_x with unburned hydrocarbons, and NO_x storage catalysts that adsorb NO_x chemically under lean conditions, and then desorb NO_x periodically under rich conditions (Johnson, 2004). Side effects of the new emission controls may include reduced engine efficiency and changes in emissions of other pollutants such as nitrogen dioxide (NO_2), nitrous acid (HONO), and ammonia (NH_3) (Jeong et al., 2008; Johnson et al., 2009). Another concern relating to use of DPF systems is the possibility of increased ultrafine particle number emissions (Lloyd and Cackette, 2001; Holmen and Ayala, 2002; Johnson, 2004).

To date the main concern about DPF systems appears to be increased primary NO_2 emissions. For example, Carslaw (2005) and Carslaw et al. (2006) showed that at many sites throughout London, NO_2 concentrations did not decrease along with NO_x between 1997-2003, and the increase in the measured NO_2/NO_x ratio could be linked to the use of DPFs and an increase in the fraction of light-duty diesel vehicles. Jenkin et al. (2008) reported a 7% increase in mean O_3 mixing ratios when modeling the effects of increases in NO_2/NO_x emission ratios in southern England.

The diesel engine replacement/retrofit program in California will lower exhaust PM emissions, but will also increase the NO_2/NO_x emission ratio from <10 to ~35% over a relatively short time period (i.e., by 2014). Since the schedule for reducing diesel NO_x emissions is more gradual compared to that for exhaust PM, increases in concentrations of NO_2 and related pollutants may be observed. Therefore the objective of this study is to assess the impacts of diesel engine emission controls on ambient air quality, by comparing future emission scenarios for 2014 with and without the accelerated diesel engine replacement/retrofit program. To make these comparisons, an Eulerian photochemical air quality model is applied to southern California for both summer (high ozone) and fall (high PM and NO_2) conditions. Air quality endpoints assessed here include O_3 and NO_2 in the gas phase, as well as fine particulate nitrate and elemental carbon mass.

Methods

Air Quality Model. The Community Multiscale Air Quality model (CMAQ; Byun and Schere, 2006) version 4.6, with the SAPRC99 gas-phase chemical mechanism (Carter, 2000) is used to evaluate air quality for baseline (2005) and two alternative future (2014) emission scenarios. The choice of future year (2014) in this study was determined by the schedule for completing installation of particle filters on (or replacing) all older on-road heavy-duty diesel engines in California. Processes relating to secondary particle formation were modeled using the AE4 aerosol module (Binkowski and Roselle, 2003). Gridded hourly meteorological fields were developed using the Mesoscale Meteorological model (MM5) version 3.6.1 (SCAQMD, 2007a). Both meteorological input data and pollutant inflow boundary conditions for the air quality model are further described by Millstein and Harley (2009).

The modeling domain (65 × 40 grid cells with 5 km horizontal resolution) is centered on the Los Angeles area in southern California, with the western boundary over the Pacific Ocean and the eastern boundary located in the Mojave Desert. The northern edge of the domain crosses north of Santa Barbara while the southern limit of the domain passes just north of San Diego (see Figure 2). Fifteen vertical model layers were used in a telescoping scheme, with the lowest layer being 36 m thick, and the topmost layer ending at a height of ~15 km above sea level. Two simulation periods are considered in this study: a summer high-ozone period running from 6 July through 29 August 2005, and a fall period extending from 30 October through 7 December 2005. During these time

periods in 2005, wildfires did not have a major impact on air quality within the modeling domain.

Baseline Emissions. Anthropogenic emission estimates for 2005 were developed by the South Coast Air Quality Management District and the California Air Resources Board, as described in the 2007 Air Quality Management Plan for the South Coast Air Basin (SCAQMD, 2007b). Baseline emissions from diesel-powered construction equipment were multiplied by 0.20 and 0.32 for NO_x and exhaust PM, respectively, to account for overstated engine activity in this sector in the baseline inventory (Millstein and Harley, 2010). The spatial distribution of construction equipment emissions was also adjusted as these activities had moved on to new locations relative to those specified in the baseline inventory. The downward revisions to construction emissions lead to increased relative importance of on-road diesel engine emissions that are the focus of the present study.

The baseline chemical composition of NO_x emissions from on-road vehicles was revised to match the 5 and 1% NO_2/NO_x ratios reported for heavy-duty diesel and light-duty gasoline vehicles, respectively, in recent California highway tunnel experiments (Ban-Weiss et al., 2008a). Additionally, weekend emission estimates for selected source categories were scaled to match the weekday-weekend source activity patterns reported by Chinkin et al. (2003) for southern California.

Baseline anthropogenic emissions for fall 2005 and summer/fall 2014 simulation periods were estimated by scaling summer 2005 emission estimates using ratios appropriate for

each pollutant and source category; these scaling factors were derived from tabulated seasonal planning inventories for 2005 and 2014 (SCAQMD, 2007b). Biogenic emissions were estimated using the BEIGIS model (Scott and Benjamin, 2003); separate day-specific estimates were available for each day of 2005 (Scott, 2009). Biogenic emissions were unchanged in the 2014 simulations.

The forecasts of future anthropogenic emissions in southern California reflect the effects of population growth and all emission control rules adopted prior to 2006. Baseline emission forecasts for 2014 do not include the effects of the new rule requiring in-use diesel engine retrofits. The effects of other recently adopted rules requiring emission reductions at ports (CARB, 2006) and reductions of greenhouse gas emissions (CARB, 2008b) are also not included here. The 2014 emission inventory factors in growth in the number of trucks on the road, as well as fleet turnover which acts by replacing older trucks with new ones that meet current new-engine emission standards for PM and NO_x. Thus significant (~50%) reductions in diesel truck emissions are forecast to occur between 2005 and 2014 even without the in-use engine retrofit/replacement rule, as shown in Figure 1.

Emissions for 2014 Retrofit Scenario. Additional reductions in PM and NO_x emissions due to accelerated retrofits and engine replacements, beyond what can be expected from fleet turnover, have been estimated by Jaw et al. (2009) and are shown in Figure 1. Emissions of NO_x from on-road diesel vehicles are reduced by 22% relative to the baseline 2014 emission scenario. Similarly, exhaust PM emissions are reduced by 64%.

These scaling factors were derived from the ratio of 2014 emissions for the two cases shown in Figure 1, with small adjustments to account for emissions from lighter vehicles (below 6 350 kg = 14 000 lb gross vehicle weight) that will not be subject to retrofits. Minor contributions to total CO and VOC emissions in the 2014 baseline inventory from on-road diesel engines were left unchanged in the retrofit scenario.

The NO_2/NO_x emission ratio for on-road heavy-duty diesel engines was set at 5% (per Ban-Weiss et al., 2008a) in the baseline 2014 emission scenario. The ratio was increased to 35% (Herner et al., 2009) in the 2014 retrofit scenario, reflecting universal use of DPF systems to control exhaust PM emissions. Some increase in NO_2/NO_x emission ratio may occur by 2014 even without the accelerated retrofit program, though this effect on fleet-average emissions will be small because any new DPF-equipped trucks must also meet low-NO_x emission standards that apply to new engines. Retrofit of DPF systems on older engines with higher NO_x emission rates has more potential to increase the fleet-average NO_2/NO_x emission ratio.

Results and Discussion

As shown in Figure 1, in the baseline emissions scenario, both NO_x and $PM_{2.5}$ are forecast to decrease with a characteristic time (half-life) of 7-8 years, reflecting the combined effects of growth in travel and fleet turnover in the on-road heavy-duty vehicle sector. This corresponds to a 9-10% per year reduction in emissions of both pollutants, which is a higher rate than has been achieved in past years. For example, Ban-Weiss et al. (2008b) found that heavy-duty diesel NO_x and PM emission factors measured on-road

decreased between 1997 and 2006 by 3 and 5% per year, respectively, and these numbers do not account for growth in diesel fuel use over the same period which offset some of the reductions. Also growth in diesel truck travel and fleet turnover are both likely to have been affected by the major downturn in the economy that has occurred since 2007. In any case, the retrofit rule is likely to speed up the rate of diesel engine emission reductions significantly, and may have a larger impact than shown in Figure 1 if the baseline emission forecasts are overly optimistic. After 2014, growth in vehicle travel leads to small increases in PM emissions in the retrofit scenario, and the difference in PM emissions between the two scenarios therefore decreases in later years. In the retrofit scenario, NO_x reductions occur at a slower pace than for PM reductions, as shown in Figure 1. Exhaust PM and NO_x emissions from the affected diesel engines are reduced in the retrofit scenario by 68 and 25%, respectively, below 2014 baseline values.

Table 1 shows domain-wide anthropogenic emissions by scenario and season. While the reductions in on-road diesel engine emissions discussed above are significant, especially for exhaust PM, the overall reductions in anthropogenic emissions between the 2014 baseline and retrofit scenarios are less dramatic. In the 2014 baseline scenario (without retrofits), on-road diesel vehicle emissions account for 26 and 23% of total NO_x and EC emissions, respectively, shown in Table 1. For the 2014 retrofit scenario, diesel vehicles are responsible for 21% of total NO_x emissions, and <10% of total EC emissions. Note in Table 1 that forecast changes in emissions between 2005 and 2014 reflect increased emphasis on controlling diesel engine emissions, and therefore include greater relative reductions in NO_x compared to CO and VOC. In contrast, through the 1990s, mobile

source emissions of CO and VOC decreased more rapidly than NO_x (Kirchstetter et al., 1999; Parrish et al., 2002).

Air quality model predictions for 2005 were compared to routine surface observations for gaseous pollutants, and to observations from 11 special study sites from the Multiple Air Toxics Exposure Study (MATES-III; SCAQMD, 2008), where speciated 24-h average fine particle concentrations were measured every third day. The network of special study $PM_{2.5}$ observation sites used for model evaluation is shown in Figure 2. As shown in Table 2, there are no large systematic biases in model predictions of the pollutants examined here for 2005 (fine particulate EC and nitrate; O_3 and NO_2 in the gas phase), though the possibility of compensating errors cannot be excluded.

More detailed comparisons of model predictions with observations are shown in Figure 3 on a site by site basis. At most locations, there is reasonable agreement for both seasons and all pollutants considered here. As expected, absolute EC and NO_2 concentrations are higher during fall, whereas O_3 concentrations are higher during summer. EC concentrations are over-predicted during the summer at Compton and North Long Beach; both sites are located close to the ports of Los Angeles and Long Beach. Fall season predictions at these locations match the observations more closely. EC concentrations at Fontana are significantly under-predicted in both seasons. These under-predictions are pervasive, not isolated to a few days or extreme events. A search of satellite imagery of the surrounding area revealed a large inter-modal freight facility located 1 km to the west; that facility is very likely to be contributing to the elevated EC concentrations observed at

Fontana. Diesel NO_x emissions from this facility are also affecting concentrations of O_3 and NO_2 observed at Fontana: in both summer and fall there are signs of ozone titration (and therefore decreased O_3 and increased NO_2) apparent in Figure 3. Similar effects are apparent for EC, O_3, and NO_2 concentrations measured at Burbank during the fall but not summer. The Burbank site is located about 500 m away from an interstate highway (I-5) that is a major truck route in and out of the Los Angeles basin. For both Fontana and Burbank, the 5 km horizontal resolution of the emission inventory and the air quality model are not sufficient to resolve plume-scale dispersion near major freight-handling facilities and highways, even if emissions from these sources were known with high precision.

Both 2014 scenarios yield lower particulate EC and nitrate, and higher O_3 concentrations relative to 2005, in response to changes in PM and NO_x emissions. Relative to the 2014 baseline, reductions of 12±2% and 14±2% in average EC concentrations are predicted for the diesel retrofit scenario, in summer and fall respectively. Changes to other pollutants were small relative to the changes in EC. As shown in Table 2, the reductions in EC due to the retrofit program are smaller than reductions that are expected due to changes in emissions from all sources between 2005 and 2014 without any retrofits.

Decreases in EC are the largest and most likely to be observable ambient effect of the in-use diesel engine retrofit rule. Direct on-road measurements of diesel engine emissions over the next 5 years in California should be able to detect changes in exhaust PM emissions as well as in NO_x mass and speciation.

Figure 4 shows predicted spatial distributions of daily average (or 8-h average in the case of ozone) pollutant concentrations for the 2014 baseline emission scenario. Figure 4 also shows further changes to these concentrations that may result from diesel retrofits. Starred locations shown in Figure 4 are Los Angeles and Riverside/Rubidoux further to the east. The largest absolute reductions in EC from the in-use diesel retrofit program occur around central Los Angeles and near the port where truck traffic is high. These locations not only have high average EC levels but also relatively high population density. This suggests prospects are good for reducing population exposure to diesel exhaust PM emissions in densely populated areas. The undesirable effects of diesel retrofits on O_3 concentrations are largest near central Los Angeles, an upwind VOC-limited region where NO_x reductions can lead to O_3 increases. Larger relative increases in ozone are forecast to occur during fall when absolute O_3 concentrations are lower (see Table 2 and Figure 3). Although increases in O_3 concentrations also are predicted in densely populated areas, the O_3 increases are smaller in a relative sense for both seasons, when compared to expected decreases in EC.

Changes to NO_2 concentrations reported in Table 2 require careful interpretation as there are competing underlying effects. During summer, NO_2 concentrations decrease by 4±3%. This is because by 2014 about half of all in-use diesel trucks are supposed to have been retrofitted with NO_x after-treatment devices, thus reducing total NO_x emissions. This reduction in total NO_x offsets the effect of DPF retrofits on older trucks, which lead to increased primary NO_2 emissions as discussed earlier. During the fall when

photochemical activity levels are lower, small increases in average NO_2 concentrations are seen near central Los Angeles and the port area.

Figure 5 shows average weekday diurnal profiles of NO_2 concentrations for both 2014 emission scenarios and seasons at Los Angeles and Rubidoux. The diurnal plots show that the differences between NO_2 concentrations in the retrofit scenario and the base case are larger at night during both seasons. However, peak NO_2 levels near Los Angeles are not lower under the retrofit program and increased exposure to direct NO_2 emissions from DPF-equipped engines remains as a possible concern. At the air basin scale, retrofitting of NO_x control equipment on in-use diesel engines plays an important role in mitigating the effect of increased NO_2 emissions due to retrofit of diesel particle filters on older engines.

Both 2005 and 2014 emission inventory estimates are uncertain. The most important uncertainties relevant to the present study are the changes in on-road diesel emissions that will occur by 2014 with and without the retrofit program. Emissions from other source categories (e.g., on-road gasoline engines, off-road mobile sources, road/soil dust emissions in the fine mode, biomass burning) also help to determine the relative importance of on-road diesel engine emissions and their control. Another important issue, not addressed here, is the longer-term durability of new and retrofit emission control systems on in-use diesel engine emissions beyond 2014 when most of the installed emission control systems will still be relatively young.

Summary and Conclusions

This study investigated effects on air quality of a control strategy that accelerates control of emissions from on-road heavy-duty diesel engines. In-use trucks will be required to meet current new-engine emission standards on a schedule that ranges from 2012 to 2023 depending on pollutant and model year. By 2014, all heavy-duty diesel trucks are expected to meet stringent PM emission standards, and about ~50% of in-use trucks will be required to have low NO_x emissions as well. The likely retrofit control system for exhaust PM, known as a diesel particle filter or particle trap, typically oxidizes NO present in the exhaust to NO_2 to help oxidize carbon particles that accumulate on the exhaust filter. As a result, the ratio of NO_2/NO_x emissions from diesel trucks equipped with particle traps can increase dramatically. The increase in primary NO_2 emissions may degrade air quality in some respects, as an undesired side-effect of controlling diesel exhaust PM emissions.

In this study we found that if retrofits of in-use diesel engines for control of both exhaust PM and NO_x emissions proceed as planned, significant reductions in atmospheric concentrations of diesel exhaust particulate matter will be realized, with a possible accompanying but smaller increase in ambient ozone. NO_2 concentrations should generally decrease along with decreased total NO_x emissions from diesel engines, despite a higher NO_2/NO_x ratio in primary emissions.

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Table 1: Air pollutant emission estimates by year, season, and scenario in southern California[1] (metric tons per day, t/d).

Year	Scenario	CO	NO_x	NMOC[2]	NH_3	$PM_{2.5}$	EC[3]
2005	Summer Baseline	3949	990	1285	111	121	23
2005	Fall Baseline	3784	1042	852	113	124	24
2014	Summer Baseline	2358	632	1059	93	110	17
2014	Fall Baseline	2198	668	636	93	114	18
2014	Summer Retrofit	2358	597	1059	93	106	15
2014	Fall Retrofit	2198	632	636	93	110	16

[1] On-grid emissions for typical weekday conditions for the southern California modeling domain shown in Figure 2.

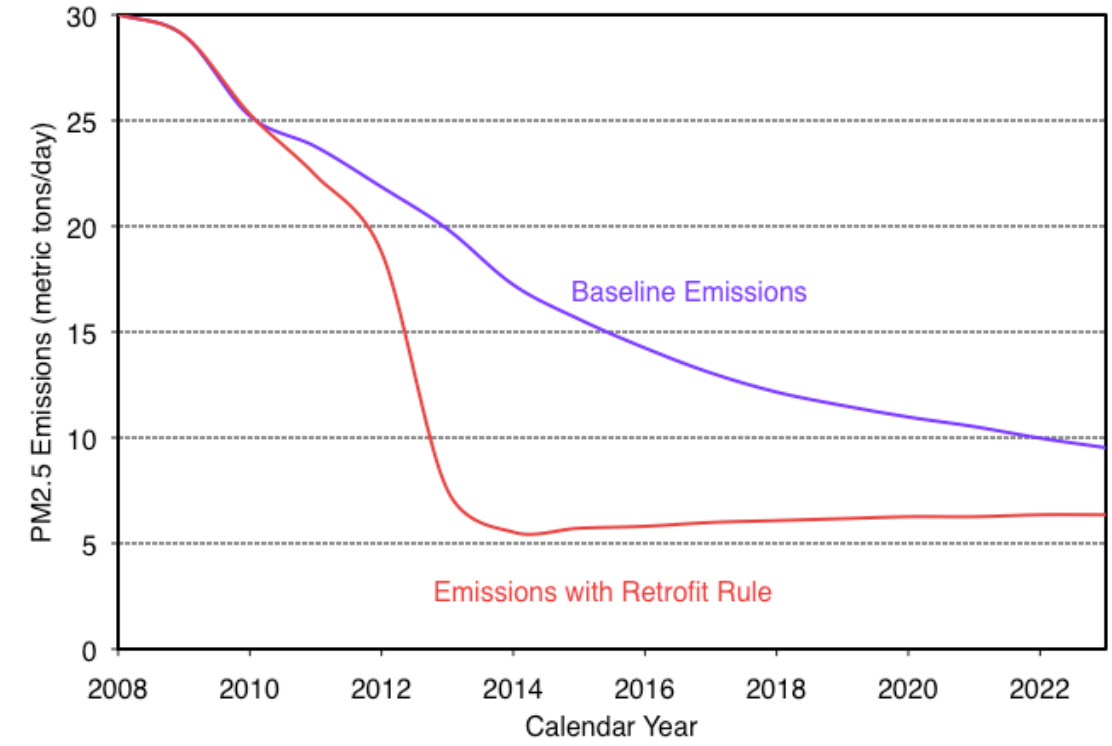
[2] The reported values for non-methane organic compounds (NMOC) include anthropogenic emissions only. Biogenic VOC contribute an additional 395±97 and 118±42 t/d in summer and fall, respectively.

[3] Elemental carbon (EC) emissions are also included as part of fine particle ($PM_{2.5}$) emissions in the previous column.

Table 2: Mean normalized differences (%) in concentrations of fine particulate elemental carbon and nitrate, and in ozone and nitrogen dioxide in the gas phase.

	EC	NO_3^-	O_3	NO_2
2005 baseline vs. observed				
Summer	+13±36	–20±23	+16±12	+23±32
Fall	–4±29	+10±29	+14±32	–6±25
2014 vs. 2005 baseline				
Summer	–23±3	–15±11	+9±8	–32±8
Fall	–23±2	–2±5	+36±14	–26±8
2014 retrofit vs. baseline				
Summer	–12±2	+2±2	+3±2	–4±3
Fall	–14±2	+6±2	+7±3	–2±3

Figure 1. On-road heavy-duty diesel engine exhaust particulate matter and nitrogen oxide emission trends (California state totals), with and without a new rule to accelerate retrofit/replacement of older engines. Source: Jaw et al. (2009).



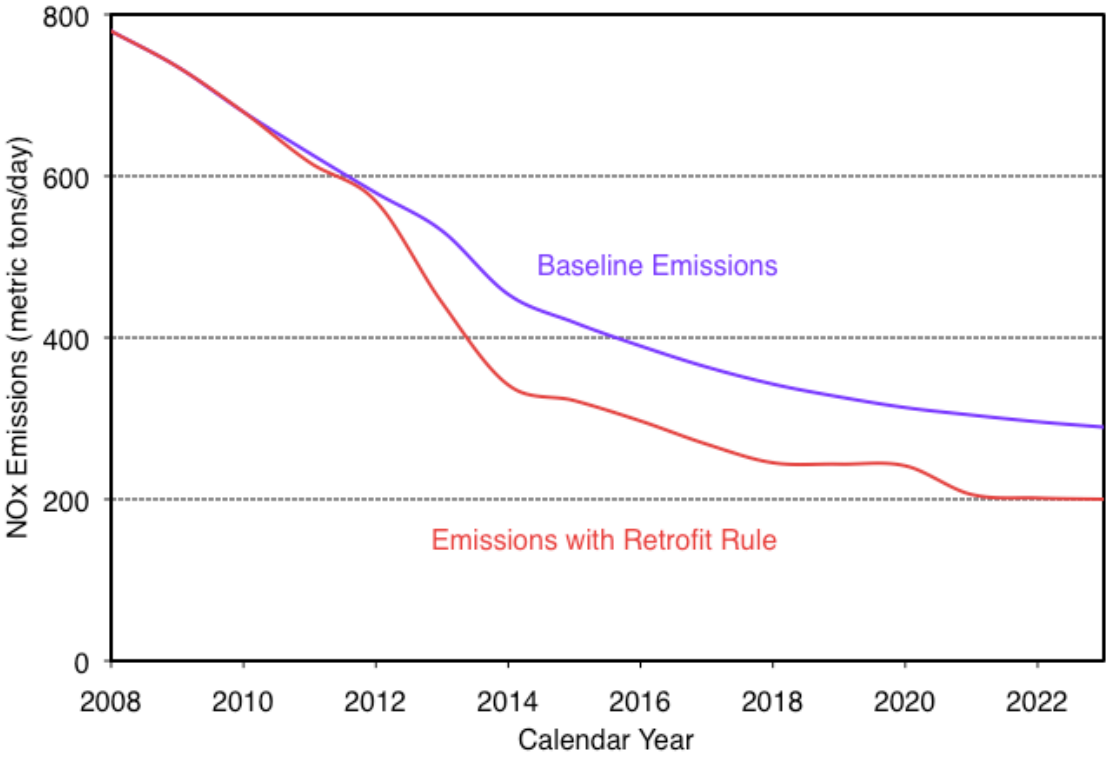


Figure 2. Southern California study domain, including MATES-III surface observation sites.

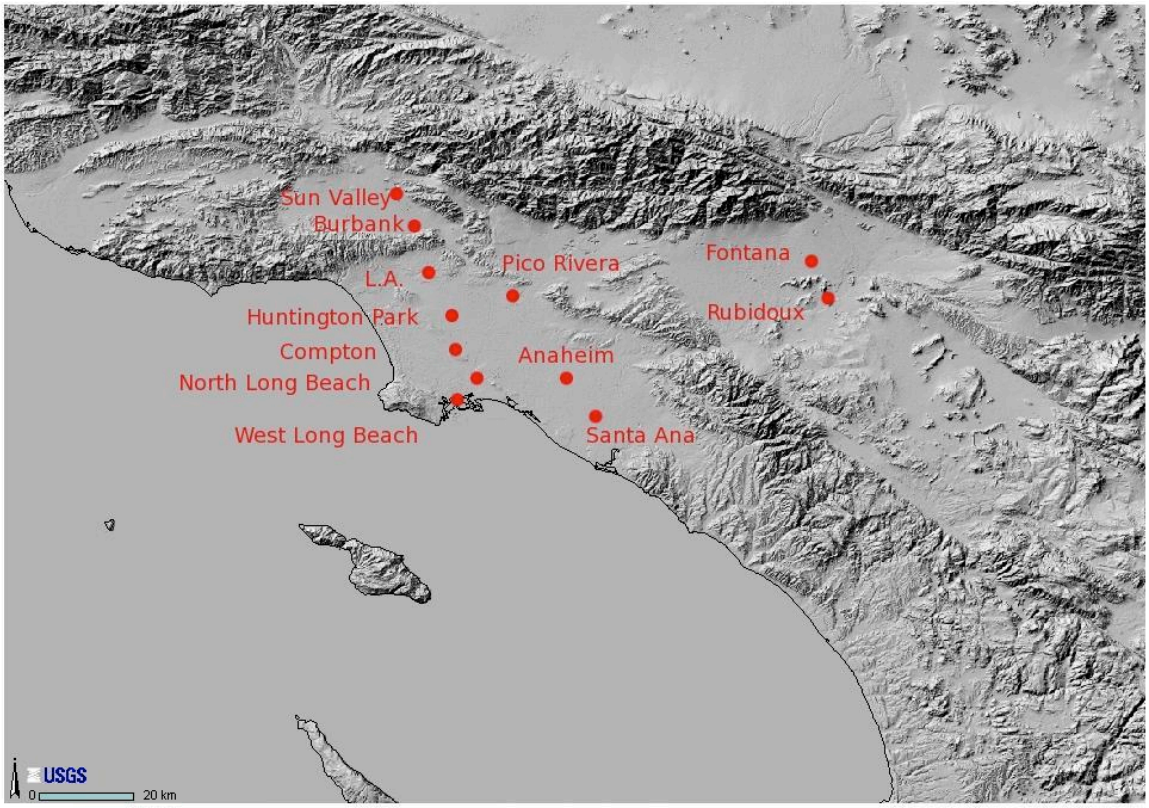
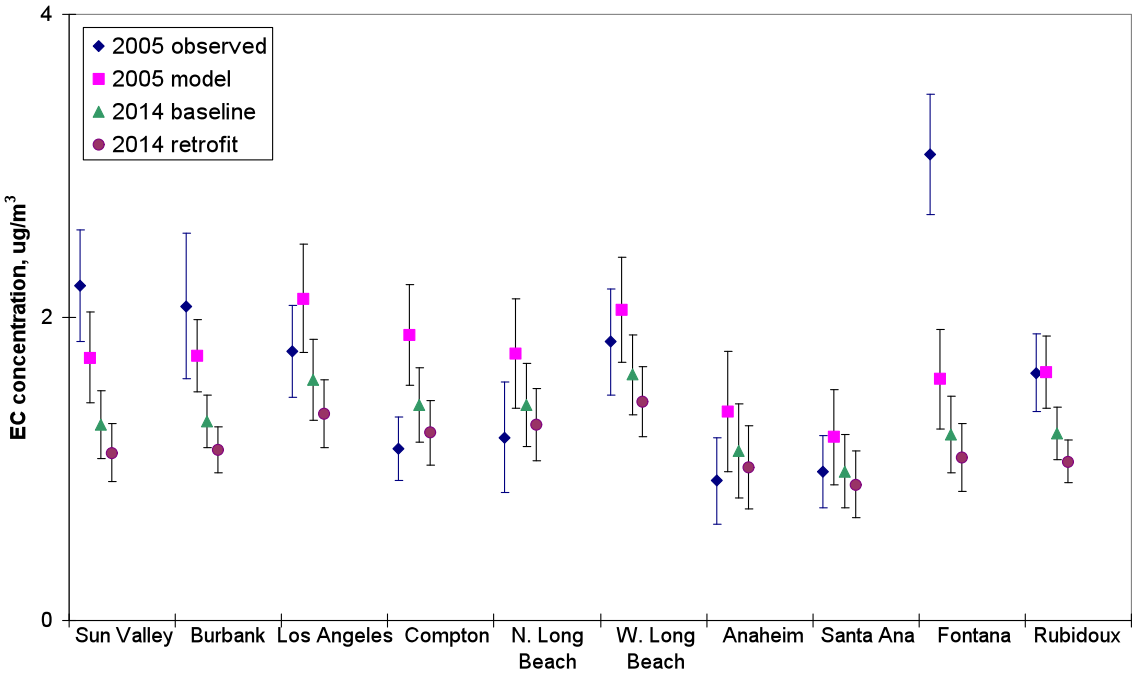


Figure 3a. Average weekday (μ ± σ) observed and modeled elemental carbon (EC) concentrations. All values shown are 24-h averages. Measurement site locations are shown in Figure 2. Top panel: summer. Bottom panel: fall.



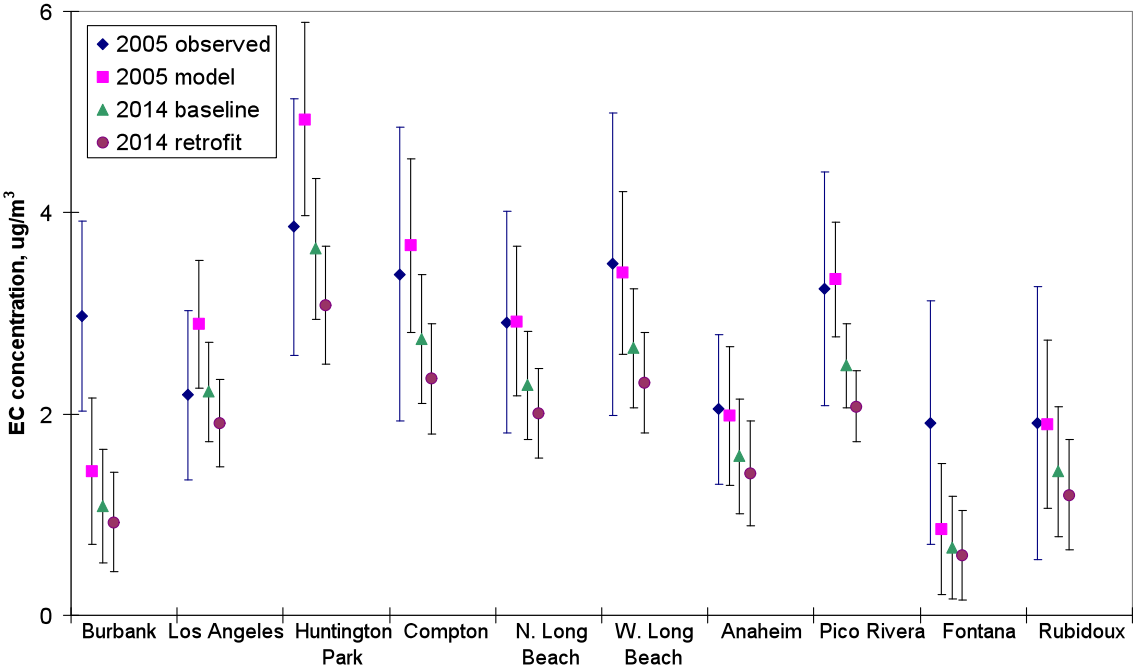
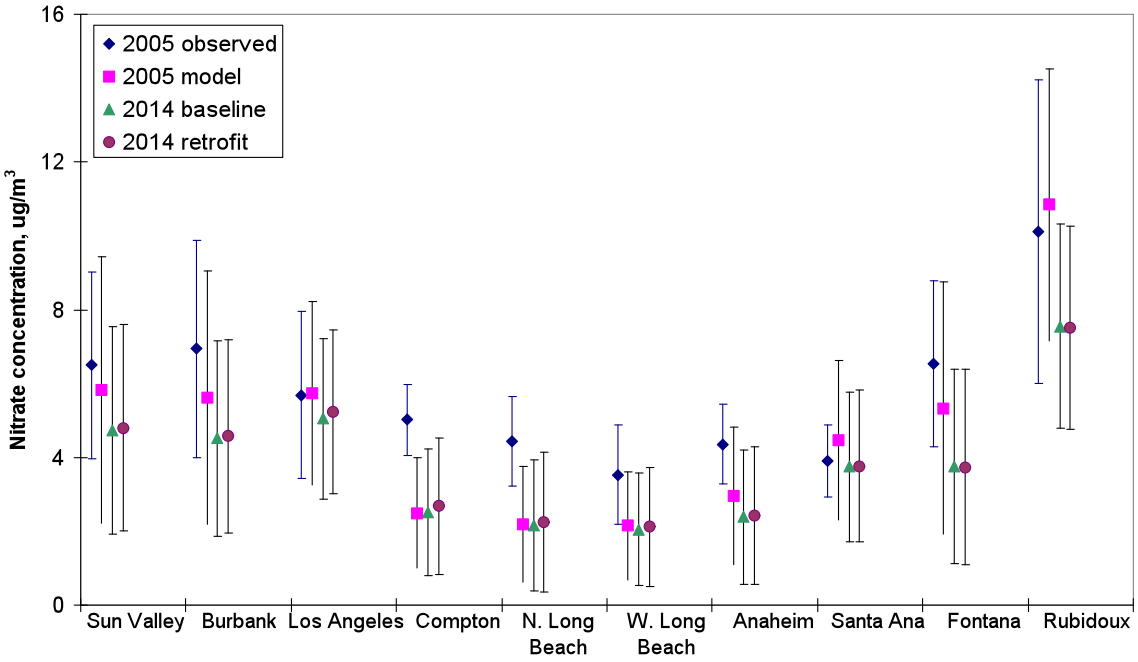


Figure 3b. Average weekday (μ ± σ) observed and modeled fine particulate nitrate concentrations. All values shown are 24-h averages. Measurement site locations are shown in Figure 2. Top panel: summer. Bottom panel: fall.



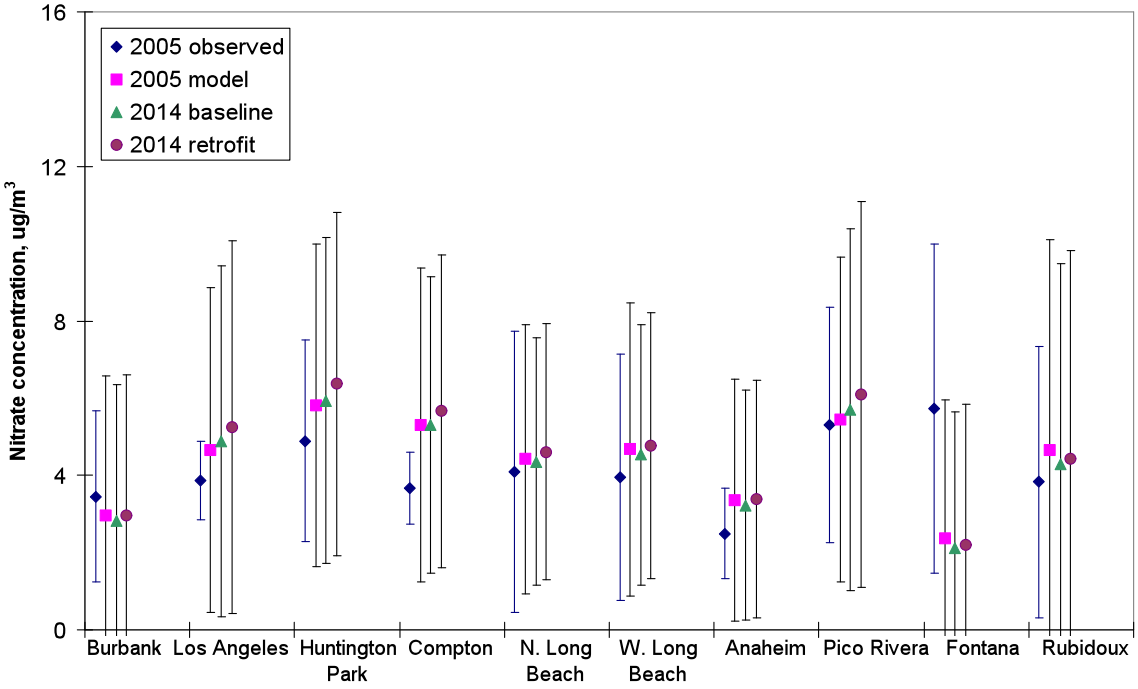


Figure 3c. Average weekday (μ ± σ) observed and modeled ozone (O_3) concentrations. All values shown are 8-h averages for the period 10 AM-6 PM local time. Measurement site locations are shown in Figure 2. Top panel: summer. Bottom panel: fall.

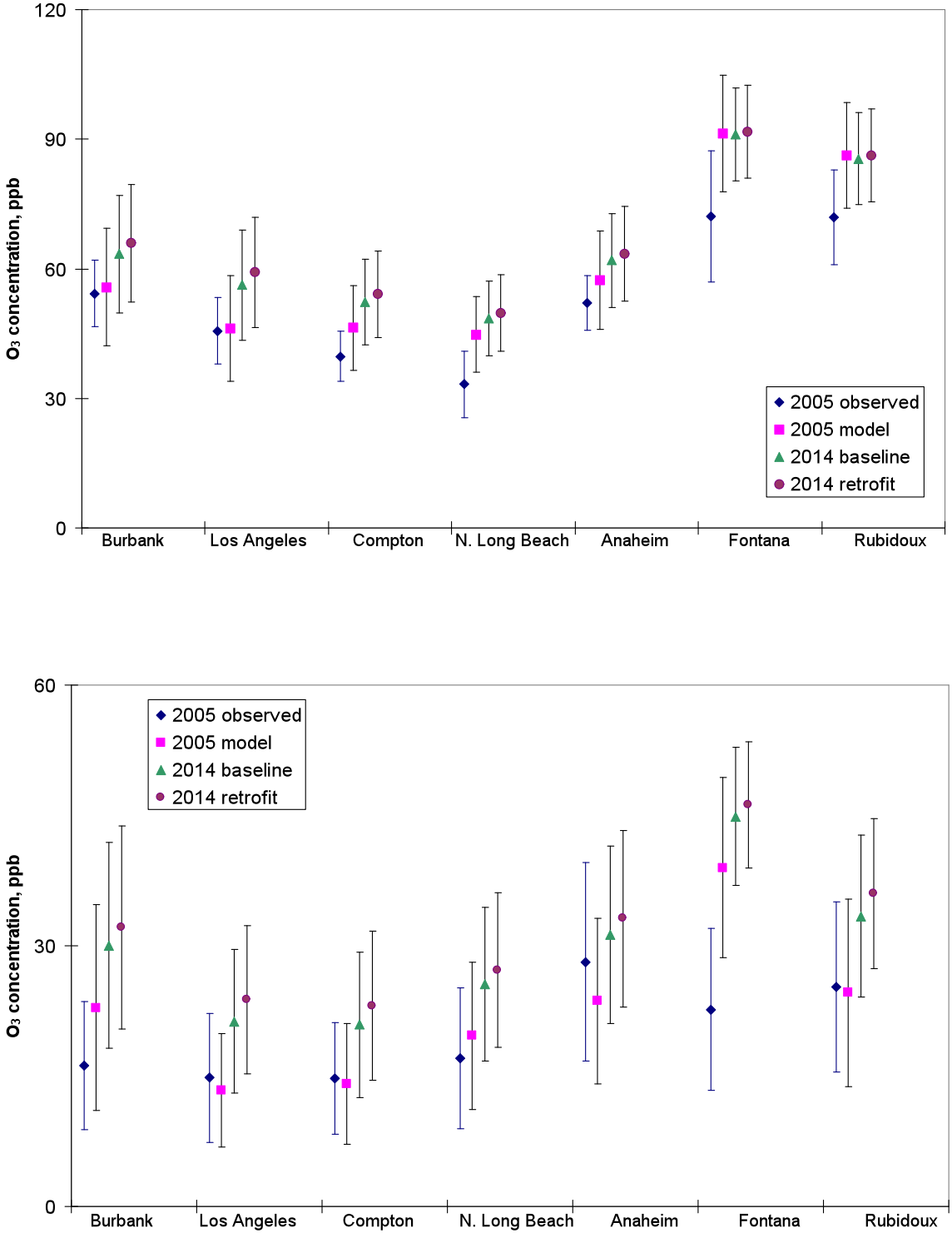


Figure 3d. Average weekday (μ ± σ) observed and modeled nitrogen dioxide (NO_2) concentrations. All values shown are 24-hour averages. Measurement site locations are shown in Figure 2. Top panel: summer. Bottom panel: fall.

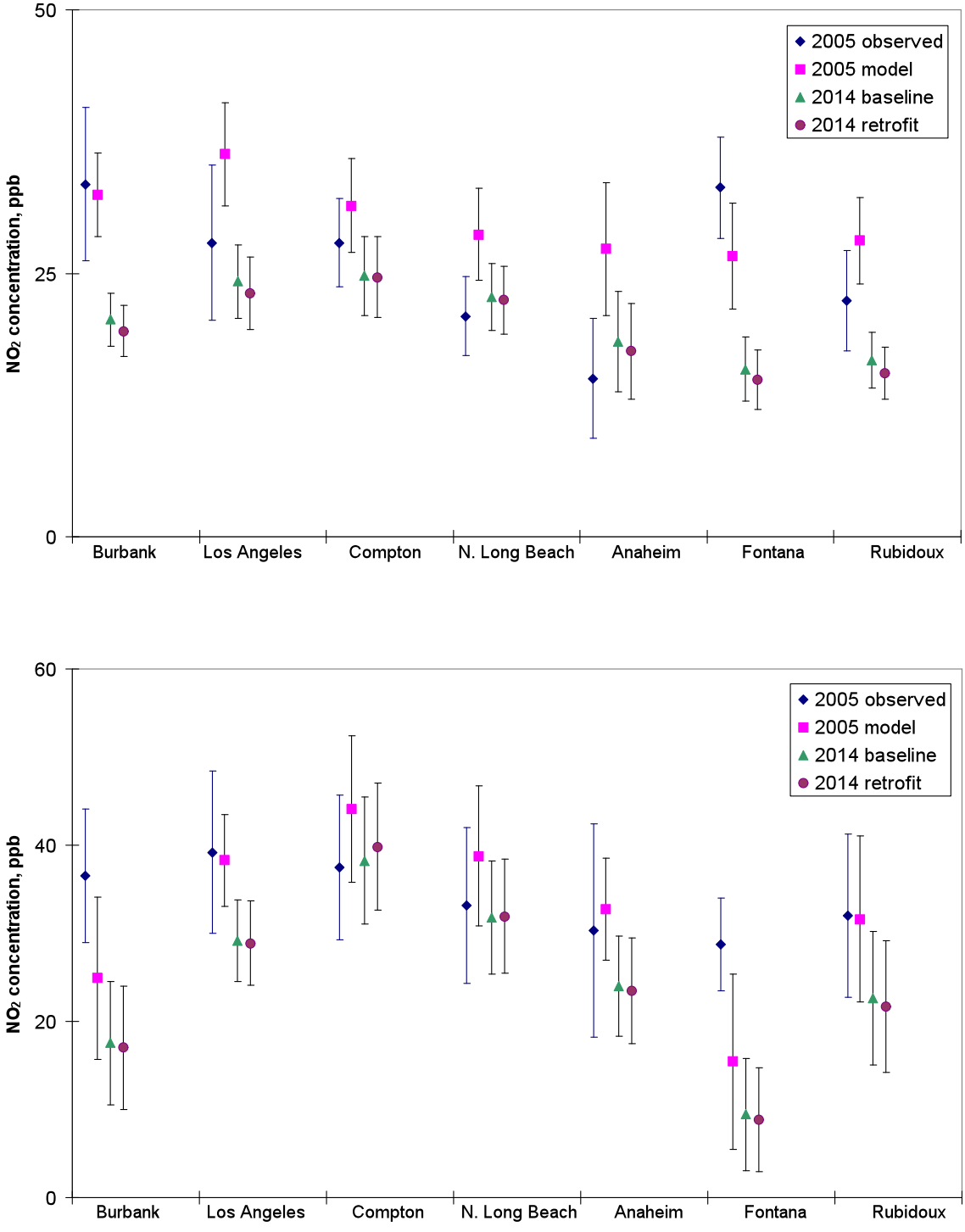


Figure 4a. Left panel: Average modeled summer concentrations of EC, NO_2, and O_3. O_3 concentrations were averaged over weekday hours 10 am – 6 pm local time. All weekday hours were used for EC and NO_2. Right panel: Difference between 2014 retrofit and baseline scenarios. The starred locations are central Los Angeles and Riverside/Rubidoux to the east.

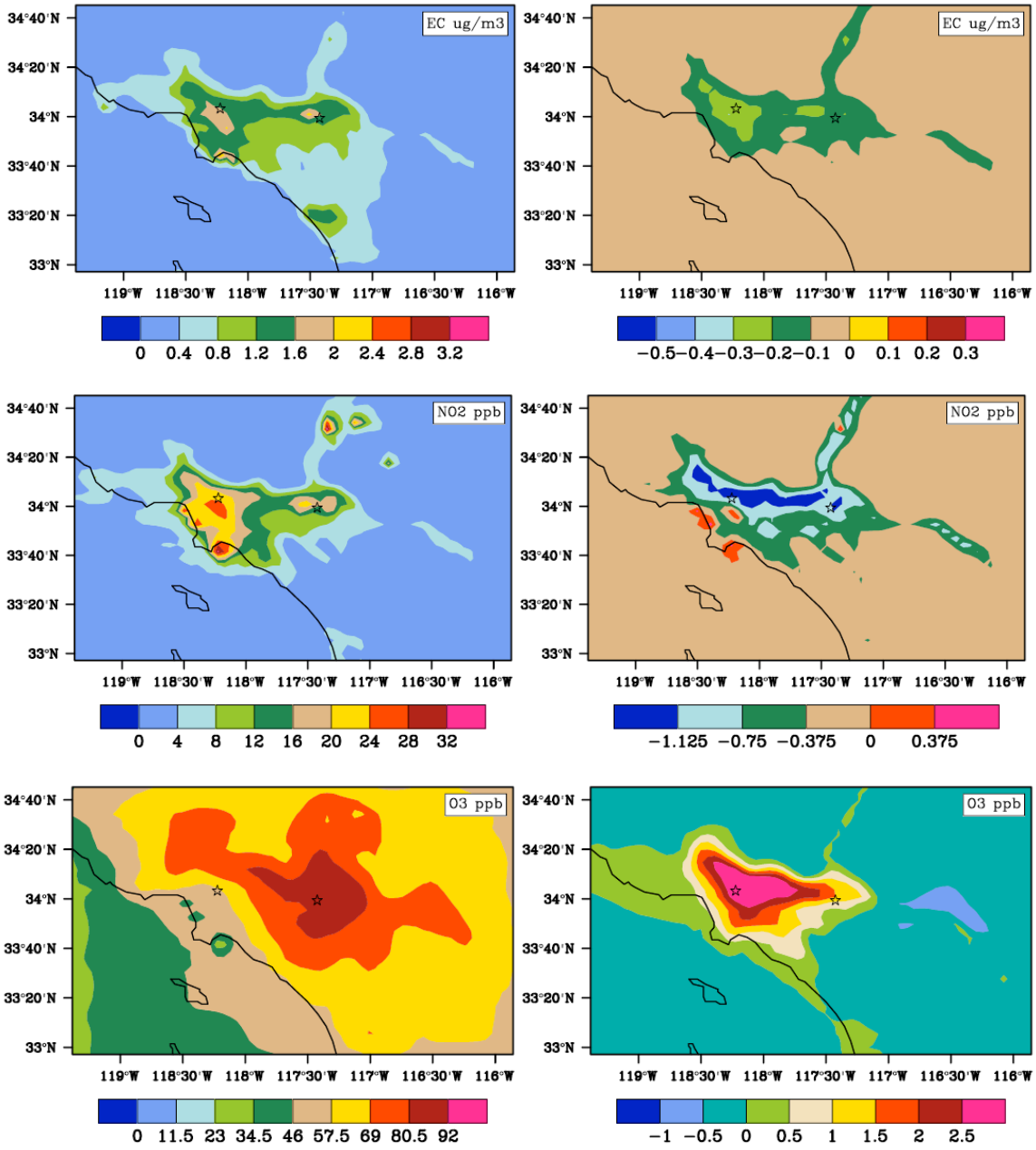


Figure 4b. Left panel: Average modeled fall concentrations of EC, NO_2, and O_3. O_3 concentrations were averaged over weekday hours 10 am – 6 pm local time. All weekday hours were used for EC and NO_2. Right panel: Difference between 2014 retrofit scenario and the 2014 baseline scenario. The starred locations are central Los Angeles and Riverside/Rubidoux.

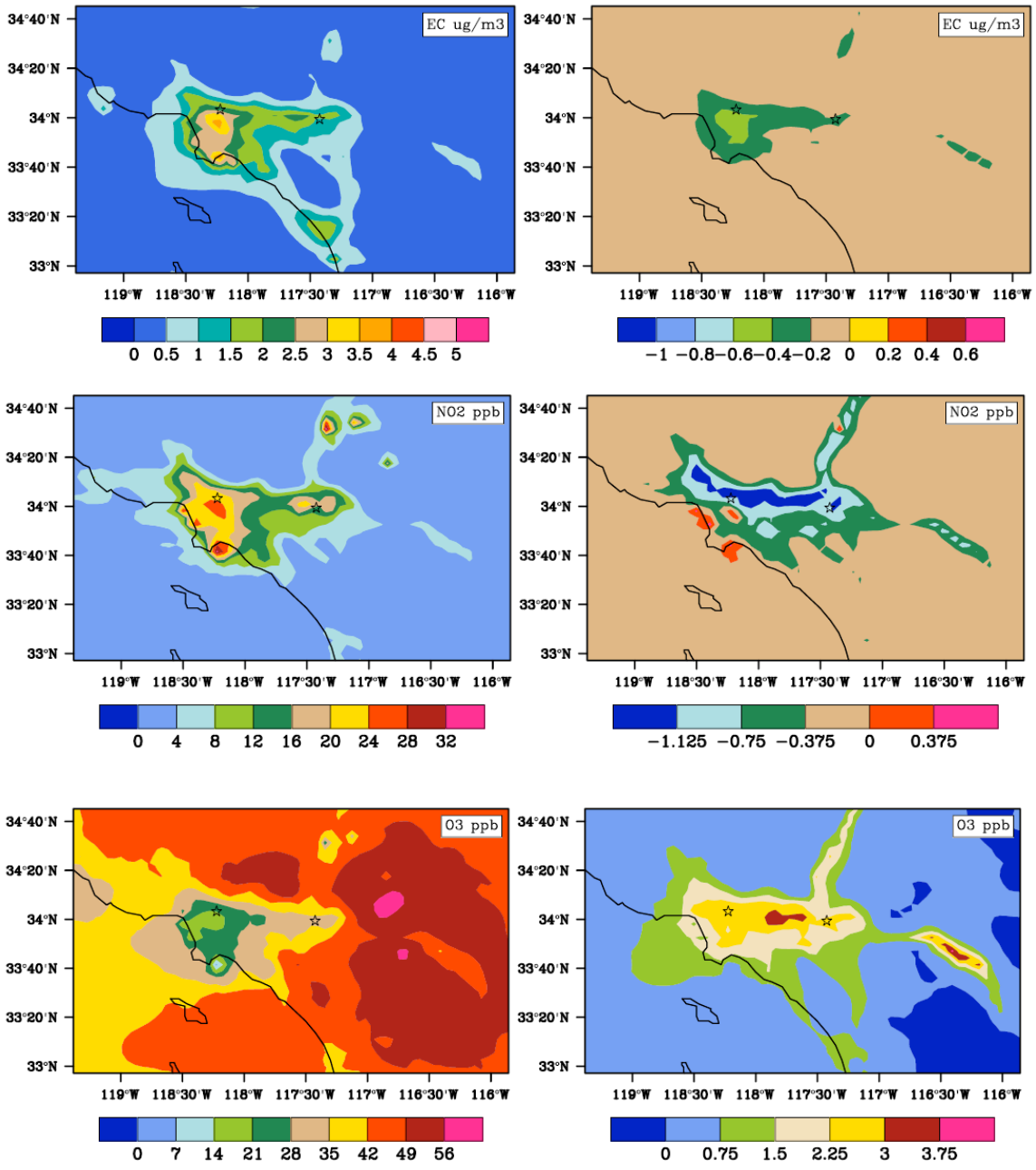


Figure 5. Average weekday diurnal NO_2 concentration profiles for summer (top) and fall (bottom). Blue: 2014 baseline scenario; Red: 2014 retrofit scenario. Left: central Los Angeles. Right: Riverside.