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TECHNICAL PAPER

An extended approach to calculate the ozone relative response factors used in the attainment demonstration for the National Ambient Air Quality Standards

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With the promulgation of the National Ambient Air Quality Standards (NAAQS or standard) for 8-hr ozone (O_3), the U.S. Environmental Protection Agency (EPA) issued modeling guidance that advocated the use of results from photochemical air quality models in a relative sense. In doing so, the EPA provided guidance on how to calculate relative response factors (RRFs) that can project current design value (DV) mixing ratios into the future for the purpose of determining the attainment status with respect to the O_3 standard. The RRFs recommended by the EPA represent the average response of the photochemical model over a broad range of O_3 mixing ratios above a specified cutoff threshold. However, it is known that O_3 response to emission reductions of limiting precursors (i.e., NO_x and/or VOC) is greater on days with higher O_3 mixing ratios compared to days with lower mixing ratios. In this study, we present a segmented RRF concept termed band-RRF, which takes into account the different model responses at different O_3 mixing ratios. The new band-RRF concept is demonstrated in the San Joaquin Valley (SJV) region of California for the 1-hr and 8-hr O_3 standards. The 1-hr O_3 analysis is relevant to work done in support of the SJV O_3 State Implementation Plan (SIP) submitted to the EPA in 2013. The 8-hr example for the future year of 2019 is presented for illustrative purposes only. Further work will be conducted with attainment deadline of 2032 as part of upcoming SIPs for the 0.075 parts per million (ppm) 8-hr O_3 standard. The applicability of the band-RRF concept to the particulate matter ($PM_{2.5}$) standards is also discussed.

Implications: Results of photochemical models are used in regulatory applications in a relative sense using relative response factors (RRFs), which represent the impacts of emissions reductions over a wide range of ozone (O_3) values. It is possible to extend the concept of RRFs to account for the fact that higher O_3 mixing ratios (both 1-hr and 8-hr) respond more to emissions controls of limiting precursors than do lower O_3 mixing ratios. We demonstrate this extended concept, termed band-RRF, for the 1-hr and 8-hr O_3 National Ambient Air Quality Standard (NAAQS or standard) in the San Joaquin Valley of California. This extension can also be made applicable to the 24-hr $PM_{2.5}$ and annual $PM_{2.5}$ standards.

Introduction

In 1979, the U.S. Environmental Protection Agency (EPA) promulgated a National Ambient Air Quality Standard (NAAQS or standard) of 0.12 ppm for 1-hr ozone (O_3). Here, we also use the alternate form of 124 ppb for this standard. Although the EPA revoked the 1-hr O_3 standard and has since adopted a more health-protective 8-hr O_3 standard of 0.075 ppm (or 75 ppb), subsequent litigation led to the reinstatement of portions of the implementation requirements under the revoked 1-hr O_3 standard. Because the San Joaquin Valley (SJV) region of California did not attain the 1-hr O_3 standard by the attainment deadline of 2010, a new State Implementation Plan (SIP) was prepared by the California Air Resources Board (CARB) and the San Joaquin Valley Air Pollution Control District (SJVAPCD), which was submitted to the EPA in 2013. The modeling protocol that describes the modeling process for this new SIP can be found elsewhere (CARB, 2013).

EPA advocated the use of modeling results in a relative sense, using relative response factors (RRFs), in the attainment demonstration of a given standard. It is important to note that the current RRF concept was developed by the EPA for the 8-hr O_3 and $PM_{2.5}$ standards (EPA, 2007) and there is no comparable concept for the 1-hr O_3 standard. Described here is an improved procedure that can be used to develop RRFs for both 1-hr and 8-hr O_3. We describe the development and application of this procedure in the context of modeling conducted in support of the 2013 SJV 1-hr O_3 SIP. We then describe its applicability to 8-hr O_3. It is

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important to note that this application for 8-hr O_3 (with a future year of 2019) is not associated with SIPs for the 75 ppb standard since SIP work is still under development with an attainment date in 2032. The applicability of this method to the 24-hr $PM_{2.5}$ and annual $PM_{2.5}$ standards is discussed in later sections. More detailed descriptions of the development of RRFs for $PM_{2.5}$ standards will be presented in the future after we conduct photochemical modeling to support the preparation of those SIPs.

A rigorous demonstration of attainment with respect to the standard for O_3 involves the projection of a reference year design value (DV) mixing ratio into the future (EPA, 2007). The DV for the 1-hr O_3 standard is the fourth highest 1-hr O_3 mixing ratio measured at a given monitor during 3 consecutive calendar years, while the 8-hr O_3 DV is the average of the annual fourth highest 8-hr O_3 mixing ratio over a consecutive 3-year period. This projection of this DV to the future is generally based on three grid-based photochemical air quality simulations. The first simulation, known as the base year or model performance simulation, involves simulating the measured O_3 mixing ratios for a recent year to evaluate the performance of the modeling system. The emissions inventory used for the base year simulation is day-specific to the extent possible for evaluating model performance. The second simulation, known as the reference year simulation, is often conducted with less day specificity in the inventory.

The EPA modeling guidance (EPA, 2007) defines "base case" as the base year used for model performance evaluation, and "baseline" as the reference year used in the DV calculation and the starting point for emissions projections. The base and the reference years need not be the same, but they can be. The third simulation involves repeating the reference year simulation for a future year, where the meteorology is held constant but the inventory has been forecasted to the future year. In the work presented here, both the base and reference years are 2007 and the future year is 2019. Here, we also used the same inventory for both the base and reference years since the day specificities in the 5-month base year and reference year inventories were very comparable. Thus, for the rest of this document, the terms base year and reference year are interchangeable. However, we keep both terms to acknowledge the fact that they can represent different years/inventories in other applications.

As mentioned earlier, the EPA modeling guidance (EPA, 2007) for the 8-hr O_3 standard recommends using photochemical air quality models in a relative sense to project the reference year DV into the future for a given monitoring location. This is accomplished by multiplying the reference year DV by an RRF specific to that location (EPA, 2007). In the calculation of the RRF, first the reference year model predictions above a predetermined threshold are averaged for a given monitor. Then the future predicted values for the same calendar days included in the reference year average are used to form the future year average. The ratio of the future year average to the reference year average is the RRF. A numerical example of this procedure is presented in the EPA modeling guidance (EPA, 2007).

Since all days in the reference year above a predetermined O_3 threshold level are used to calculate a single RRF for a given monitor, this RRF is representative of a broad range of O_3 values in the reference and future years. However, as our previous modeling experience shows and the EPA modeling guidance (EPA, 2007) demonstrates, in general, the higher the reference-year O_3 mixing ratios, the larger is the response to emissions reductions of limiting precursors. A recent EPA memorandum (EPA, 2012) also documents the enhanced response to emissions reductions at higher O_3 values.

The phenomenon of enhanced response of higher O_3 mixing ratios to emissions controls is also supported by observed trends in 1-hr and 8-hr daily maximum O_3. As a part of the health-based review process of the State Ambient Air Quality Standards for O_3, the California Air Resources Board (CARB) reported the historic (1980–2002) trends in annual percentiles of daily maximum 1-hr and 8-hr O_3 in the South Coast air basin (CARB, 2005). The 40th to 90th percentiles in increments of 10 and the maximum O_3 mixing ratios were calculated as 3-year averages from the annual distribution of the basin's daily maximum. The CARB (2005) report (see Figure 10-1 in that work) showed a consistent downward trend in the observed 1-hr O_3 for the maximum and different percentile levels, but the highest values showed the largest decrease (e.g., the relative percent reductions between the first [1981] and last [2001] year of the time series [i.e., Figure 10-1 of CARB, 2005] in annual daily maximum 1-hr O_3, and the 90th, 80th, 70th, 60th, 50th, and 40th percentiles of 1-hr O_3 mixing ratios are ~57, 54, 50, 46, 41, 34, and 29% respectively). Similar behavior is also seen in observed 8-hr O_3 trends (Figure 10-2 of CARB, 2005).

The EPA modeling guidance (EPA, 2007) suggests that this enhanced response of higher O_3 values to emission reductions may be suggestive of O_3 being more "controllable" at those levels. In other words, the contribution of local and regional sources to formation of O_3 is relatively larger on days with elevated O_3 levels, when compared to the low-/medium-range O_3 values. In addition, high O_3 days can also occur due to stagnation caused by high pressure systems. The higher temperatures can result in increased chemical reaction rates and enhanced emissions of highly reactive biogenic volatile organic compounds. This could lead to enhanced ozone production efficiency (OPE) (Zhou et al., 2014) on high O_3 days. It should be possible to further clarify the underlying causes for this trend with retrospective studies linking the enforced emission control strategies with the observed values.

The current form of the RRF concept does not allow for this enhanced response to emissions controls at the high end of the simulated/measured O_3 distribution and uses a single RRF value to represent a broad range of O_3 values in the reference and future years. For this reason, we use the term "single-RRF" when we refer to the current form. Due to its representation of a broad range of O_3 values, single-RRFs indeed represent an average response and could be too conservative in some cases. To better represent the enhanced response of higher O_3 mixing ratios to emissions controls, we have developed a segmented RRF method termed "band-RRF," which is a logical extension to the single-RRF method. The details of this method are provided in

the following sections for application to both the 1-hr and 8-hr O_3 NAAQS in the SJV region of California. We discuss other advantages of this new method in later sections.

The Modeling System

We first describe the modeling system that produced the simulated O_3 mixing ratios used to calculate the RRFs. The Weather Research and Forecasting (WRF; Skamarock et al., 2005) model was used to generate the meteorological fields for the 5-month simulation period of May through September 2007 (CARB, 2013). The initial and boundary conditions (IC/BCs) for WRF were prepared based on National Centers for Environmental Prediction (NCEP) Eta 212 grid (40 km) model output that is archived at the National Center for Atmospheric Research (NCAR). Initial conditions for WRF were updated at 6-hr intervals for the 36-, 12-, and 4-km grids. In addition, surface and upper air synoptic observations obtained from NCEP were also used to further refine the IC/BCs. The WRF model was nudged toward observed meteorological conditions by using the analysis nudging option of the Four Dimensional Data Assimilation (FDDA) for the 36-km grid only. The physics options used for WRF include the WSM 6-class graupel microphysics scheme, the Monin–Obukhov surface layer scheme, the unified Noah land-surface model, the YSU planetary boundary layer scheme, the Kain–Fritsch (new Eta) cumulus scheme for the 36- and 12-km domains, the RRTM long-wave radiation scheme, and the Dudhia short-wave radiation scheme. There were 31 vertical model layers telescoping from the surface to 50 mb.

Two sets of emissions inventories were prepared by CARB in consultation with the local air districts. The reference year inventory was prepared with emissions developed specifically for each day of the simulation period for sources such as motor vehicles, biogenics, wildfires, prescribed fires, agricultural burning, road dust, and ocean-going vessels. Since the same meteorology was used for both the reference year and the future year, the same day-specificity was included in the 2019 reference inventory as well. For model performance evaluation, a fully day-specific base year inventory is the most appropriate. However, in practice, it is very difficult to make an inventory fully day-specific for a long simulation period. Therefore, the only difference between the reference and base year inventories was the inclusion of day-specific agricultural burning and wildfires in the base year inventory. The reference year inventory had a more average representation of the agricultural burning and wildfires.

Since the difference between the two inventories was not significant, the same 2007 base year inventory was used as the 2007 reference year inventory. An emission forecast was developed for the future year of 2019. The anthropogenic emissions forecasts for 2019 were based on forecasted economic conditions, population growth, and implemented/planned emission controls. Biogenic emissions were generated using the MEGAN model version 2.04 with California-specific emission factor and plant functional type data (CARB, 2013). Biogenic emissions were kept constant between 2007 and 2019 because future inputs such as land use/land cover are highly uncertain. Detailed descriptions of the emission inventory preparation including temporal and spatial distributions as well as chemical speciation can be found elsewhere (CARB, 2013). The future 2019 total anthropogenic emissions of NOx and VOC were approximately 55% and 20% lower when compared to the corresponding base year 2007 totals.

The CMAQ model version 4.7.1 (Foley et al., 2010) was used to simulate O_3 formation in the SJV region of California. The following options were used: the SAPRC99 gas-phase chemical mechanism (Carter, 2000) with the EBI solver, the aero5 aerosol module, the unmodified piecewise parabolic method for horizontal and vertical advection, and the eddy diffusivity algorithm for vertical diffusion. Figure 1 shows the CMAQ modeling domains that were used in this work. The larger domain covering all of California has a grid size of 12 km and was used to provide boundary conditions for the smaller domain, which covers central California with a finer grid size of 4 km. The 31 vertical layers in the WRF simulation were mapped onto 15 vertical layers in CMAQ that extended telescopically from the surface to 50 mb, with the majority of layers falling within the planetary boundary layer. The boundary conditions for the larger 12-km domain were extracted from the global atmospheric chemical transport model for ozone and related chemical tracers (MOZART; Emmons et al., 2010; CARB, 2013).

To assess the model prediction skills in simulating O_3 values, a model performance analysis was carried out for the 2007 base case by including simulated O_3 values above a 60-ppb threshold and using statistical metrics recommended by the EPA for 1-hr O_3 (EPA, 1991). These metrics are peak (unpaired) prediction accuracy (±15–20%), overall bias (±5–15%), and gross error statistics (30–35%). The acceptable ranges indicated in the parentheses are approximate and were recommended by Tesche at al. (1990). Detailed model performance statistics are presented in the Supplemental Materials. In general, the fraction of days from the model that meets the performance criteria for 1-hr O_3 is highest in the lower elevation regions of the central (~58%) and southern (~56%) SJV. In the northern region and above 3000 ft in the SJV, the number of days meeting model performance criteria was generally lower (~13 and 36%, respectively) due to the lower O_3 mixing ratios and fewer monitoring stations in those areas. The 8-hr O_3 model performance statistics show similar behavior and are consistent with the corresponding1-hr O_3 statistics (Table S1 in Supplemental Materials).

Calculation of the Reference Year 1-Hour O_3 DVs

A key quantity that needs to be calculated a priori based on observations is the reference year 1-hr O_3 DV. This DV is then projected to a future year using RRFs. The form of the 1-hr O_3 standard has evolved over the years,and the current form requires that the expected number of days per calendar year with daily maximum 1-hr O_3 mixing ratio exceeding 124 ppb must be less than or equal to 1 per year over a 3-year period. If every day is monitored at a given location, then during a 3-year period the fourth highest daily maximum 1-hr O_3 mixing ratio becomes the DV for that location. If there are missing data in the observational record, then one can use the method advocated by Curran

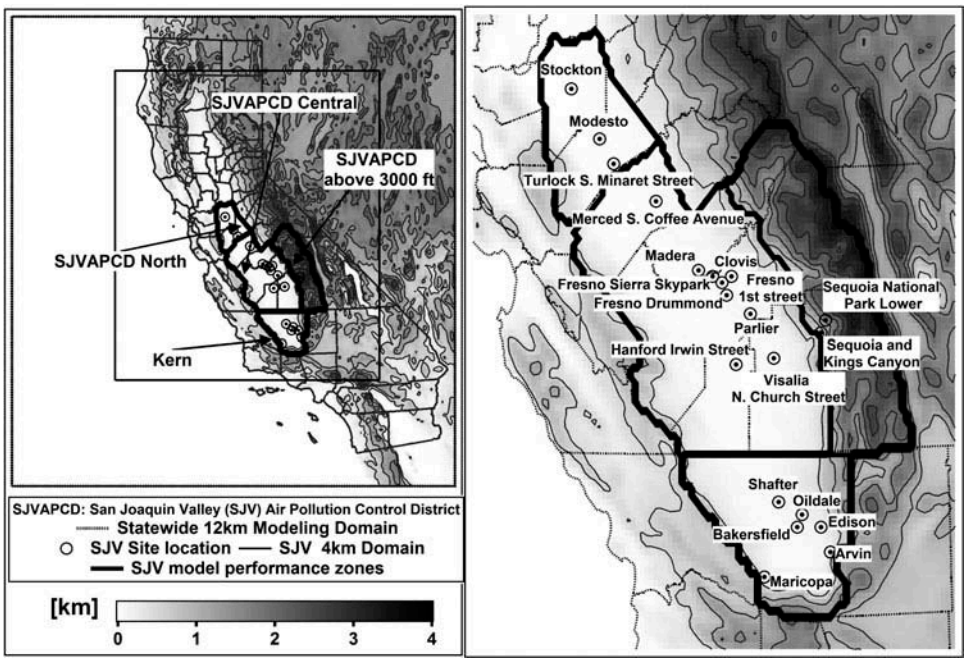

Figure 1. The ozone monitoring stations in the San Joaquin Valley (SJV) region of California. SJVAPCD stands for the San Joaquin Valley Air Pollution Control District. The outer box of the left panel is the California statewide 12-km modeling domain. The gray shaded and black line contours denote the gradients in topography (km), while the thick black lines show the boundaries of the SJV subregions used in model performance analysis (see Supplemental Materials for further details on model performance). Simulations for this study were conducted using the inner modeling domain at 4 km resolution that covers the entire Central Valley of California. The insert on the right shows a zoomed-in view of the site locations that were used for model performance evaluation. Table 1 lists only a subset of these sites (the 10 highest 1-hr and 8-hr O_3 DVs for the year 2007).

and Cox (1979), which assumes the same exceedance rate of the measured days for the unmeasured days.

In California, all major locations are monitored daily. Therefore, the fourth highest daily maximum 1-hr O_3 mixing ratio during a 3-year period is a very good representation of the DV. This methodology also allows for data incompleteness (i.e., missing data in the observational record) by allowing the data analyst to select the third (or even the second) highest daily maximum 1-hr O_3 mixing ratio as the DV if the data completeness criteria are not met. For these reasons, we use that form of the DV in this study. The 2007 1-hr O_3 DVs, calculated over the period of 2005–2007, for representative sites (see Figure 1 for location of sites) in the SJV region are shown in the second column of Table 1. The 2007 1-hr O_3 DVs for the SJV monitoring sites exhibit a large variability with the highest and lowest values seen at the Edison (135 ppb) and Madera (95 ppb and not shown in Table 1) sites respectively.

Methodology to Calculate the RRFs

The calculation of single-RRFs for the 8-hr O_3 standard is described in the EPA modeling guidance (EPA, 2007), and is summarized briefly here. First, the 8-hr daily maximum O_3 mixing ratios for both reference and future years are computed using the simulated 1-hr O_3 mixing ratios. Then, for each day and each monitor, the highest "nearby" (i.e., in a grid cell within a 15-km radius of the monitor) 8-hr daily maximum O_3 mixing ratio is selected from the reference and future year simulations. The mean 8-hr daily maximum O_3 mixing ratios from future and reference year simulations are computed for each nearby grid cell of a monitoring site.

A subset of reference year days with simulated 8-hr daily maximum O_3 above a predetermined threshold are selected next and the mixing ratios are averaged separately for the reference and future year for those days. This threshold is monitor specific and is determined using the following procedure outlined in the EPA modeling guidance (EPA, 2007). For the previous 84-ppb 8-hr O_3 standard, the starting threshold is set to 84 ppb and an RRF is calculated if there are at least 10 simulated days above the threshold (i.e., 84 ppb). However, if the number of simulated days above the threshold is less than 10, this threshold is lowered to 70 ppb (i.e., the absolute minimum threshold) until 10 days become available. In the event of having less than 10 days above 70 ppb, all the available days above 70 ppb are used for calculating the RRF provided there are at least 5 days. If a monitor does not have at least a minimum of 5 days above 70 ppb, then an RRF is not calculated for that monitor (EPA, 2007). The single-RRF is then computed as the ratio of the average future year and reference year 8-hr daily maximum O_3 mixing ratios. The EPA modeling guidance (EPA, 2007) provides a numerical example on pages 27–28.

The EPA modeling guidance (EPA, 2007) requires that the model performance criteria be met to evaluate prediction skills of the base year simulation and to build confidence in projecting the future values. However, the model performance statistics are not explicitly considered in the 8-hr O_3 single-RRF calculations, which are typically based on reference and future year simulations (Hogrefe et al., 2008). In the current application, we

Table 1. The 2007 and 2019 O_3 DVs for representative monitoring sites in the San Joaquin Valley of California; listed here are the top 10 2007 DV sites for both 1-hr and 8-hr O_3

	1-hr O_3				8-hr O_3		
Monitoring station	DV (ppb) ambient (2005–2007)[a]	DV (ppb) single (2007–2019)[b]	DV (ppb) band (2007–2019)[b]	Monitoring station	DV (ppb) ambient (2005–2007)[a]	DV (ppb) single (2007–2019)[b]	DV (ppb) band (2007–2019)[b]
Edison	135	111.9	108.1	Arvin	107	88.8	85.5
Arvin	131	105.5	103.1	Sequoia and King Canyon	103	86.2	84.7
Fresno 1st street	130	105.3	98.1	Edison	99	83.2	81.0
Clovis	125	100.9	96.1	Fresno 1st street	98	81.6	78.3
Fresno Sierra Skypark	124	100.9	92.1	Bakersfield	97	81.3	79.0
Parlier	121	95.4	92.6	Fresno Sierra Skypark	95	79.1	76.1
Sequoia and King Canyon	118	98.7	94.8	Sequoia National Park	95	81.0	81.6
Bakersfield	117	96.8	94.5	Visalia N. Church Street	95	80.4	78.7
Sequoia National Park	113	94.9	92.1	Clovis	93	77.1	75.0
Visalia N. Church Street	112	88.6	87.8	Parlier	93	76.7	74.3
Oildale	112	93.3	92.0	Oildale	91	74.5	73.1

Notes: [a]The three year period included in the ambient design value calculations. [b]The base year 2007 and future year 2019 used for air quality model simulations used in this study.

followed the RRF procedure outlined in the EPA modeling guidance (EPA, 2007) but have imposed an additional constraint that only the reference year simulated values that are within ±20% of the corresponding observed values be used in the single-RRF calculation. This criterion attempts to exclude simulated data for some days that have less than ideal model performance and also impacts the band-RRF calculations as discussed in later sections.

1-Hour O_3 RRF calculation methodology

Single-RRF. It is important to note that there is no RRF concept for 1-hr O_3 since the RRF concept was developed by the EPA for the 8-hr O_3 and $PM_{2.5}$ standards (EPA, 2007). In this study, the calculation of single-RRFs for the 1-hr O_3 standard follows the procedure described in the EPA modeling guidance (EPA, 2007) to calculate single-RRFs for the 8-hr O_3 standard with one notable exception of including the reference year simulated values that are within ±20% of the corresponding observed values.

The choice of the absolute minimum threshold for the 1-hr O_3 daily maximum value, which is used to select modeled reference year days as input to the RRF calculation, is subject to scientific judgment. The EPA modeling guidance (EPA, 2007) recommends using 70 ppb as the absolute minimum threshold for the 84 ppb 8-hr O_3 standard. An equivalent minimum threshold for the 124 ppb1-hr O_3 standard would be approximately 100 ppb. However, it must be noted that the 1-hr O_3 daily maximum values represent real values in the atmosphere and exhibit relatively larger variability than the 8-hr O_3 daily maximum values (which are average values). Thus, in the 1-hr O_3 RRF calculations, it may not be appropriate to use the equivalent minimum threshold for 1-hr O_3 daily maximum that is calculated based on the minimum threshold used for the 8-hr O_3 daily maximum values.

To evaluate the impact of the choice of minimum threshold value on the 1-hr O_3 single-RRF values, a series of RRF calculations was performed using fixed minimum thresholds ranging from 60 ppb to 100 ppb in 10-ppb increments. The two factors considered in selecting a suitable threshold were (1) encompassing the broad range of 2007 DVs exhibited by the SJV monitoring sites and (2) availability of a minimum number of 5 days to form a stable single-RRF (EPA, 2007). Based on the criteria already described, we chose 90 ppb as the minimum threshold for 1-hr O_3 single-RRF calculations. This choice depends on the nature of the O_3 problem in a given geographical location and should be reevaluated accordingly for applications outside of the SJV region of California.

We applied the procedure outlined in the EPA modeling guidance (EPA, 2007) to determine the number of days and the minimum threshold and subsequently calculate the single-RRF at each monitor in the SJV region for the 124 ppb 1-hr O_3 standard. The reference year (i.e., 2007) 1-hr O_3 DVs were then multiplied by the 1-hr O_3 single-RRFs to determine the future 1-hr O_3 DVs, which are shown in the third column of Table 1.

Band-RRF. We now describe the procedure to calculate 1-hr band-RRFs. We begin with the same data set used in the

calculation of single-RRFs that includes reference year simulated 1-hr O_3 mixing ratios falling within ±20% of the corresponding measured values. We then bin the reference year data into regular 5-ppb bands from 60 to 120 ppb. The selection of the width of the bands (5 ppb in this case) is subject to scientific judgment. If the bands are too wide, there will not be enough points for performing a linear regression to represent missing bands, which is described in the later sections. If the bands are too narrow, then the RRFs within each band will be based on very few days. We found 5 ppb to be a reasonable balance between these two extremes in this application.

It should be noted that in each bin the lower limit values are included but not the upper limit (e.g., bin 60–65 ppb includes values greater than or equal to 60 but less than 65). Even though it is important to spread high values (considering their enhanced response to emission reductions) into more bands, all values greater than or equal to 120 ppb were segregated into a single band due to the small number of simulated high values ≥120 ppb. Within each band, an RRF was calculated as a ratio of average future year mixing ratios to average reference year mixing ratios.

The calculation of band-RRFs is illustrated in Figure 2. The top left panel of Figure 2 shows the daily maximum 1-hr O_3 time-series plot for the Edison monitoring site located in the southern SJV region (see Figure 1). Edison had the highest reference year (i.e., 2007) DV of 135 ppb (based on measurements made during 2005–2007) in the SJV region. The agreement between the observations (solid black circles) and the simulation was satisfactory (~54% of the simulated days meet the model performance criteria outlined earlier in the modeling system section) for the highest value simulated within a 15-km radius of the monitor (solid black line).

The top right panel of Figure 2 shows the band-RRF values (y-axis) for each band (x-axis). The decrease in band-RRF values with increasing band number illustrates that the model is more responsive to emissions controls at higher O_3 values. For comparison, the single-RRF for this site is shown as a black dashed line. For some sites, the RRF values for certain bands (especially at the high end) may be missing because the model did not simulate mixing ratios within that band.

To represent the missing bands, we performed a linear regression of available RRFs starting from the 60 ppb bin and only when at least three bands with simulated 1-hr O_3 mixing ratio ≥90 ppb were available (solid black line). We chose this criterion to prevent the lower less responsive bands from dominating the fit. Based on an analysis for all the representative SJV monitoring sites (not shown here) that are included in the SIP, we selected the 60-ppb threshold (with at least three bands that have simulated O_3 mixing ratio ≥ 90 ppb) for the band-RRF regression. This choice increases the number of bands available

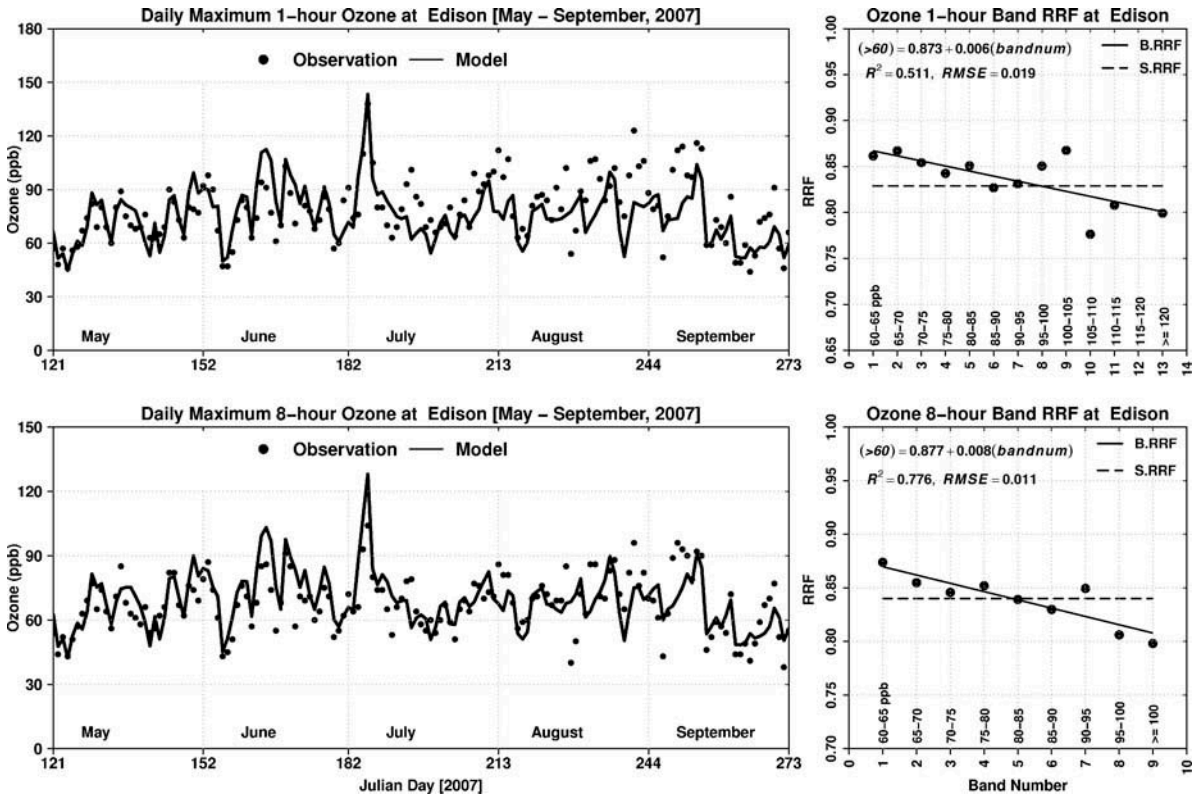

Figure 2. An illustration of the band-RRF procedure for the Edison monitoring site. The top left panel shows the time series of observed (black circles) and simulated (black line) daily maximum 1-hr O_3 for the simulation period (May–September 2007). The top right panel shows the scatter plot of binned band-RRF for 1-hr O_3 from 60 to 120 ppb (in 5-ppb increments) vs. the number of bands. The bottom left panel shows the time series of observed (black circles) and simulated (black line) daily maximum 8-hr O_3 for the simulation period (May–September 2007). The bottom right panel shows the scatter plot of binned band-RRF from 60 to 100 ppb (in 5-ppb increments) for 8-hr O_3 vs. the number of bands. S.RRF and B.RRF denote single- and band-RRFs, respectively.

for the regression and provides more stability. It also ensures that low mixing ratios will not dominate the regression. In addition, including only the reference year data that fall within ±20% of the measured values in the RRF calculations further constrains the band-RRF regression fit.

We have used the RRFs on the regression line for all bins instead of the actual band-RRF points when available, since the regression fit represents the average site specific RRF for that particular mixing ratio range. This approach also reduces the uncertainty caused by a band with very few data points (that are used in the RRF calculation for that particular band) and prevents it from having a disproportional impact on the future DV calculations.

The decrease in RRF with increasing band number raises an issue with respect to multiplying the reference year DV directly by the appropriate band-RRF. If the individual observations used to determine the reference year DV were to be projected into the future with their corresponding band-RRF and the fourth highest future value was selected as the future DV, it is possible that this future DV could be different from a future DV that was projected directly from the reference year. For example, the order of values when projected to the future year could be different from the order of values in the reference year. This possibility was recognized in a different context (24-hr $PM_{2.5}$) in the 2011 addendum to the 2007 EPA modeling guidance (EPA, 2011).

To account for this potential reshuffling of values, the new future DVs were calculated by multiplying the top 10 observed 1-hr daily maximum O_3 mixing ratios from the 3 years ending in the reference year (i.e., 2005–2007) by their corresponding band-RRF values. The future year values were then re-sorted and the value of the fourth highest 1-hr O_3 was selected as the future 1-hr O_3 DV for that monitor. The future 1-hr O_3 DVs, calculated using this procedure, are shown in the fourth column of Table 1 and are in general lower than the corresponding single-RRF DVs shown in third column of Table 1 (e.g., at the Edison monitoring site, the band-RRF-based future DV of 108.1 ppb is ~4 ppb lower than the corresponding single-RRF-based DV of 111.9 ppb).

8-Hour O_3 RRF calculation methodology

In this section, the procedure to calculate the 8-hr O_3 RRFs is described. The 8-hr O_3 DV is the average of the annual fourth highest daily maximum 8-hr O_3 mixing ratios over 3 consecutive calendar years (EPA, 2008). The 2007 8-hr O_3 DVs (based on the 2005–2007 measurement period) are shown in the sixth column of Table 1 for representative sites in the SJV region. These DVs are highly variable across the region and range from 77 ppb at Stockton (not shown in Table 1) to 107 ppb at Arvin Bear Mountain (see Figure 1 for site locations). The ordering (ranking) of the 2007 DVs for the 1-hr and 8-hr O_3 DVs is different. For instance, the highest 1-hr O_3 DV is at the Edison site, while this site is ranked third for the 8-hr O_3 DVs. This is expected, given the differences in the form of the two O_3 standards.

Single-RRF. We now discuss the single-RRF calculations for 8-hr O_3 values. In this study, we followed the procedure to calculate the single-RRFs for the 8-hr O_3 standard as described in the EPA modeling guidance (EPA, 2007) with one notable exception: We calculated the RRFs based on the subset of the data with the reference year simulated values that are within ±20% of the corresponding observed values (to maintain consistency with the 1-hr O_3 calculations described earlier). In this study, we have used 70 ppb as the minimum threshold value for 8-hr daily maximum O_3 values that are included in the RRF calculations (EPA 2007). This minimum threshold is based on the 84-ppb standard and may need to be reassessed in the future to reflect the current 75 ppb 8-hr O_3 standard. The future 8-hr O_3 DVs, calculated by multiplying the reference year (i.e., 2007) 8-hr O_3 DVs with the corresponding 8-hr O_3 single-RRF values, are shown in the seventh column of Table 1.

Band-RRF. We now describe the procedure of applying the band-RRF concept to the 8-hr O_3 standard using the same example of the Edison monitoring site used earlier in the 1-hr O_3 band-RRF section. The time series (bottom left panel of Figure 2) of the observed (solid black circles) and simulated "nearby" (i.e., in a grid cell within a 15-km radius of the monitor) daily maximum 8-hr O_3 values (black solid line) at Edison shows good agreement (~52% of the simulated days meet the model performance criteria outlined earlier in the modeling system section) and follows the similar trend seen in corresponding 1-hr O_3 values (top left panel of Figure 2). Further details of the model performance statistics for 8-hr O_3 are presented in the Supplemental Materials.

We applied an analogous procedure for 8-hr O_3 band RRF analysis (similar to the 1-hr O_3 band-RRF calculations described in previous sections) but with two notable exceptions. First, an RRF was calculated for each 5 ppb bin in the 60–100 ppb range, where the upper bound of 100 ppb is less than the corresponding 120 ppb used in the 1-hr O_3 calculations. Second, the bands with missing RRFs were represented by the band-RRF regression and this regression was applied only when at least three bands with simulated 8-hr O_3 mixing ratio $\geq$70 ppb were available. Note that this criterion was set to 90 ppb for the 1-hr O_3 band-RRF regression calculations. The bottom right panel of Figure 2 clearly shows that the simulated higher 8-hr O_3 values are more responsive to emission controls through the decreasing trend in RRF values with increasing band number (consistent with the corresponding 1-hr O_3 values).

To account for potential reshuffling of the annual fourth highest mixing ratio, as in the case of 1-hr O_3 described earlier, a larger number of days (10 days per year with a total of 30 days during 3 years) were projected to the future and subsequently used in the future year DV calculation. This is different from the 1-hr O_3 procedure, where the 10 highest measured O_3 mixing ratios from the entire 3-year measurement period were used in calculating the future 1-hr O_3 DV at each monitor. The top 10 daily maximum 8-hr O_3 mixing ratios from each of the 3 years (i.e., 2005–2007) were projected to the future using the corresponding band RRFs, re-sorted, and the fourth highest value was calculated at each monitor. The future 8-hr O_3 DV is then calculated as the three-year average of the annual fourth highest 8-hr O_3 mixing ratio at each monitor (eighth column of Table 1). These DVs are in general lower than the corresponding single-RRF DVs (seventh column of Table 1), and this is consistent with

the trend seen in the 1-hr O_3 future DVs. For instance, at the Edison monitoring site, the band-RRF-based future DV of 81 ppb is ~2 ppb lower than the corresponding single-RRF DV of 83.2 ppb.

The EPA modeling guidance (EPA, 2007) recommends simulating a full O_3 season for demonstrating compliance with the 8-hr O_3 standard using the RRF methodology. In this study, single- and band-RRFs are calculated based on modeling results from a single O_3 season (i.e., 2007 reference year) projected to the future, consistent with the EPA recommendation. Since the reference year DVs are based on 3 years of ambient O_3 mixing ratios, ideally 3 years should be simulated and used in the future year DV calculation. This would allow the use of year-specific regression curves to select appropriate band-RRFs for each of the top 10 values. However, it is currently not feasible to conduct three annual simulations within the context of a SIP, primarily due to computational limitations. Therefore, the standard practice in SIP preparation is to simulate a single O_3 season (e.g., http://www.dec.ny.gov/chemical/37012.html) or a shorter period in some cases (e.g., http://www.tceq.texas.gov/airquality/airmod/data/dfw8h2).

It is known in California that high O_3 mixing ratios occur within very specific meteorological regimes, and they occur repeatedly over time (Beaver and Palazoglu, 2009). Thus, it may be reasonable to assume that these meteorological regimes are represented in the RRFs developed for 1 year and they can be reasonably applied to high O_3 mixing ratios over the 3 years.

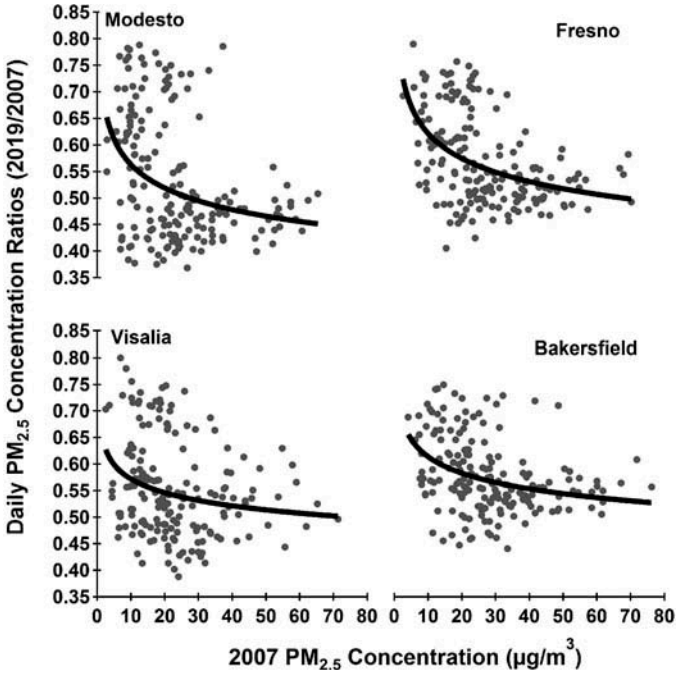

Figure 3. The total $PM_{2.5}$ daily concentration ratios (2019/2007) at Modesto, Fresno, Visalia, and Bakersfield monitoring sites in the SJV region. The solid line represents the power form of the regression. (See Chen et al. [2014] for further details on the simulated $PM_{2.5}$ concentrations.)

Extension of the Band-RRF Concept to $PM_{2.5}$

Before we discuss the extension of the band-RRF procedure to the $PM_{2.5}$ standards, we first demonstrate in Figure 3 that, as for O_3, high $PM_{2.5}$ concentrations respond more to emissions controls of the limiting precursors in the SJV region. The four panels in Figure 3 show daily 2019/2007 $PM_{2.5}$ concentration ratios as functions of 2007 simulated $PM_{2.5}$ concentrations for four representative locations in the SJV. The solid line is the nonlinear regression line generated using the power form. The simulated $PM_{2.5}$ concentrations used in Figure 3 are described in Chen et al. (2014).

The 24-hr $PM_{2.5}$ DV is based on the average of the annual 98th percentile of observed 24-hr $PM_{2.5}$ values over a period of 3 consecutive years (40 CFR Part 50, Appendix N). The procedure to test for attainment of this standard (EPA, 2011) already has some elements of the band-RRF method in it. For example, after determining the rank of the 98th percentile value for a given reference year, the top eight measured values per calendar quarter (32 values/year) are projected into the future, one component (e.g., nitrate, sulfate, ammonia, etc.) at a time, using quarter- and component-specific single-RRFs. The projected future components are then combined into a future total $PM_{2.5}$ concentration, sorted in descending order, and the value of the rank that corresponds to the 98th percentile in the reference year is selected as the future 98th percentile. The future DV is the average of future 98th percentile concentrations for three consecutive years. The only modification needed in this case is to introduce band-RRFs for each calendar quarter for each component. The same procedural steps that were described in the 1-hr O_3 standard can be followed in this case. Other processing steps (e.g., determining the speciation, calculating the particle bound water, etc.) would remain the same.

The demonstration for attainment of the annual $PM_{2.5}$ standard requires that quarterly averages of the reference year be used to form the annual average and that three consecutive years be averaged to form the annual DV (EPA, 2007). In the procedure to demonstrate attainment of the annual $PM_{2.5}$ standard, each quarterly average is projected to the future using quarter- and component-specific RRFs. Then the future DV is calculated using the same averaging procedure followed in the reference year (EPA, 2007). Again, the only modifications needed are to introduce band-RRFs for each calendar quarter for each component and to project all measurements into the future. The same procedural steps that were described for the O_3 standards can be followed to calculate the quarter- and component-specific RRFs. All the other processing steps would remain unchanged.

It may appear to be a daunting task to calculate band-RRFs, particularly for $PM_{2.5}$, where multiple components are involved. However, nearly all steps can be automated with spreadsheet software that is readily available. We are currently in the process of conducting photochemical model simulations for the future $PM_{2.5}$ SIPs. At the conclusion of those simulations we will provide operational details and numerical examples on how to apply band-RRFs to both 24-hr $PM_{2.5}$ and annual $PM_{2.5}$ standards.

Conclusion

In this study, we have developed and applied the band-RRF concept, which addresses the observed phenomenon that simulated high O_3 mixing ratios are, in general, more responsive to emission controls of limiting precursors than lower mixing ratios are. The concept of band-RRF is a logical extension to the currently used single-RRFs and can be applied to estimate varied model responses for different ranges of O_3 mixing ratios. Since the reference year DVs are typically based on 3 years of ambient O_3 mixing ratios, it would be ideal to simulate all 3 years and then calculate an individual day-specific RRF to estimate the future DV at each monitoring site. However, computational limitations do not make it viable to conduct three annual simulations within the context of a SIP. The band-RRF concept makes it possible to calculate the day-specific RRFs without simulating the three measurement years by estimating average site specific RRFs for different ranges of O_3 mixing ratios.

The band-RRF based future DVs are generally lower than those calculated with single-RRFs. However, there can be cases (e.g., 8-hr O_3 DV for the Sequoia National Park Site in this study) where the band-RRF procedure results in a higher DV if higher O_3 mixing ratios are less responsive to emissions controls (e.g., due to long-range O_3 transport). The single-RRF procedure, due to its averaging nature, may not be able to represent this situation accurately. The concept of the band-RRF is only marginally more complex than that of the single-RRF. It is readily extendable to any standard that uses RRFs and can be easily implemented using spreadsheet software.

The width of the bands should be selected carefully to prevent having too few points for regression analysis (if the bands are too wide) and calculating RRFs for a given band with too few points (if the bands are too narrow). However, as seasonal (May–September 2007 in this case) and annual air quality simulations become commonplace in regulatory modeling, the selection of the width of the bands would become less critical.

Here, we have presented an improved approach to using RRFs to account for the deficiencies of the photochemical model. However, using the modeling results in a relative sense may not be the only solution. Another potential strategy may be to correct the biases in the reference year using the observations (Kang et al., 2010, Zhang et al., 2012). In the simplest case, if it is assumed that the biases in the future year are identical to those in the reference year, then the same bias adjustment can be applied to the future year simulation. Such a bias-corrected future year could then be used directly to calculate the future DVs without resorting to RRFs.

Disclaimer

This paper has been reviewed by the staff of the California Air Resources Board and has been approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the California Air Resources Board, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

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Supplemental Material

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