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Comparison of background ozone estimates over the western United States based on two separate model methodologies

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HIGHLIGHTS

- Two modeling approaches have been applied to estimate background ozone levels.
- Background ozone estimates are generally independent of the model methodology used.
- Seasonal mean background ozone can approach 40–45 ppb at sites in the western U.S.
- On high days, ozone in the rural western U.S. can average 60–80 percent background.

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ABSTRACT

Two separate air quality model methodologies for estimating background ozone levels over the western U.S. are compared in this analysis. The first approach is a direct sensitivity modeling approach that considers the ozone levels that would remain after certain emissions are entirely removed (i.e., zero-out modeling). The second approach is based on an instrumented air quality model which tracks the formation of ozone within the simulation and assigns the source of that ozone to pre-identified categories (i.e., source apportionment modeling). This analysis focuses on a definition of background referred to as U.S. background (USB) which is designed to represent the influence of all sources other than U.S. anthropogenic emissions. Two separate modeling simulations were completed for an April–October 2007 period, both focused on isolating the influence of sources other than domestic manmade emissions. The zero-out modeling was conducted with the Community Multiscale Air Quality (CMAQ) model and the source apportionment modeling was completed with the Comprehensive Air Quality Model with Extensions (CAMx). Our analysis shows that the zero-out and source apportionment techniques provide relatively similar estimates of the magnitude of seasonal mean daily 8-h maximum U.S. background ozone at locations in the western U.S. when base case model ozone biases are considered. The largest differences between the two sets of USB estimates occur in urban areas where interactions with local NO_x emissions can be important, especially when ozone levels are relatively low. Both methodologies conclude that seasonal mean daily 8-h maximum U.S. background ozone levels can be as high as 40–45 ppb over rural portions of the western U.S. Background fractions tend to decrease as modeled total ozone concentrations increase, with typical fractions of 75–100 percent on the lowest ozone days (<25 ppb) and typical fractions between 30 and 50% on days with ozone above 75 ppb. The finding that estimates of background ozone are not strongly dependent on the technique applied lends credibility to this and earlier modeling work.

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1. Introduction

It is well-established that surface ozone levels measured across the United States can be influenced by background ozone

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concentrations (Fiore et al., 2002, 2003; Zhang et al., 2011). The main sources of background ozone include natural emissions of ozone precursors (e.g., biogenic methane and volatile organic compounds [VOCs], as well as oxides of nitrogen [NO_X] from lightning and other natural processes), wildfires, transport of naturally occurring ozone from the stratosphere to the troposphere, and transport of anthropogenic ozone and ozone precursors from upwind regional and/or international locations. The first step in implementing an efficient plan for improving air quality is to develop a conceptual model of all the processes that lead to high concentrations of atmospheric pollutants within an airshed. As such, accurate estimates of the relative contribution of background ozone to observed ozone levels will be a key element in the development of air quality management plans for attainment of the ozone National Ambient Air Quality Standards (NAAQS), especially in the western U.S. where background influences tend to be greater.

The first ozone NAAQS was established in 1971 by the Environmental Protection Agency pursuant to its authority in the Clean Air Act to protect against the health and ecosystem risks associated with ambient oxidants (36 FR 8186). The earliest attempts to mitigate the ozone problem in the U.S. focused on local hydrocarbon controls in highly urbanized cities. While there was progress in reducing ozone, many of these urban areas failed to meet their NAAQS attainment goals in the mid-1970s. This prompted questions about the contribution of "background" processes (i.e., any sources outside the local region) to high ozone observations and how their impacts should be considered in the design of effective air quality control plans (Stasiuk and Coffey, 1974; Singh et al., 1977). Observational studies during the 1970s and 1980s began to quantify the local background that was associated with regional transport of ozone and ozone precursors in the eastern U.S. (Wolff et al., 1977; Clarke and Ching, 1983). By the early 1990s, most ozone control planning efforts were aimed at reducing regional NO_X emissions and local VOC emissions. Over the intervening decades, peak ozone concentrations improved considerably in California and the eastern U.S., but less so in rural portions of the western U.S. (Cooper et al., 2012; Simon et al., 2014), where ozone levels were generally lower. The increasing stringency of the ozone NAAQS, as needed to ensure adequate protection of public health, combined with the relatively unchanged ozone levels in the inter-mountain western U.S. have prompted increased concern about the role of background ozone levels in this region.

Initially, background ozone estimates were derived entirely from measurement studies, either by using direct measurements at relatively remote monitoring sites or via inference according to co-located measurements of ozone, NO_X, NO_Y, and CO (Trainer et al., 1993; Altshuller and Lefohn, 1996). While some rural monitoring locations are affected substantially by background sources and may be suitable for comparisons with model results (McDonald-Buller et al., 2011), several analyses have shown that even the most remote ozone monitoring locations in the United States are periodically (or persistently) affected by U.S. manmade emissions (Parrish et al., 2009; Wigder et al., 2013). Further, most routine ozone monitoring sites across the U.S. do not have the co-located NO_X, NO_Y, and CO measurements that are considered necessary to support inferential, observation-based, determinations of background contributions and monitoring coverage can be sparse in some locations. Because background contributions cannot be easily quantified using monitoring data, photochemical grid models have been widely used to characterize the contribution of background ozone to observed concentrations.

Over the past 10–15 years, several photochemical modeling analyses have been conducted to estimate the role of background sources on U.S. ozone levels. Many of these modeling analyses estimated a specific background metric which was intended to represent the ozone levels that would remain after North American anthropogenic emissions have been removed, or "zeroed out". This metric is generally referred to as North American background (NAB), although older analyses referred to it as policy-relevant background. One of the first analyses to attempt to quantify NAB was conducted by Fiore et al. (2003) using a zero-out methodology and the GEOS-Chem global model (Bey et al., 2001) with a relatively coarse horizontal grid resolution ($2.0 \times 2.5°$). The modeling concluded that springtime afternoon ozone concentrations (1300–1700 local time) could occasionally reach 40–50 ppb at high altitude sites in the western U.S. without any contribution from North American anthropogenic emissions. Background concentrations in the eastern U.S. were substantially lower. Summer season average NAB concentrations were lower, generally ranging from 15 to 35 ppb. Subsequent zero-out GEOS-Chem modeling (Wang et al., 2009), conducted with a finer horizontal grid resolution ($1.0 \times 1.0°$), essentially confirmed the NAB estimates from the earlier study and separately estimated that anthropogenic emissions from Canada and Mexico could contribute an additional 3 ppb to mean ozone levels across the U.S., with larger impacts at near-border sites. A third set of zero-out modeling was conducted by Zhang et al. (2011) using an updated version of the GEOS-Chem model and still finer horizontal grid resolution ($0.5 \times 0.67°$). This analysis concluded that seasonal mean North American background levels generally ranged from approximately 20–35 ppb at low-altitude U.S. sites, but could exceed 45 ppb at certain high-altitude locations in the western U.S. Lin et al. (2012a,b) utilized a separate global high-resolution model (GFDL AM3, approximately 50×50 km^2) to estimate springtime NAB levels at high-elevation western U.S. sites and concluded that April–June mean NAB values could be approximately 50 ppb at these sites. The Lin et al. (2012a,b) study was one of the first background assessments to use a bias-correction technique to adjust the background estimates for ozone biases inherent in the base case model predictions. Fiore et al. (2014) compared background estimates between the GEOS-Chem and GFDL AM3 models and concluded that the varying model estimates of background magnitude resulted primarily from differences in the treatment of stratospheric–tropospheric exchange, wildfire emissions, lightning NO_X emissions, and isoprene oxidation chemistry between the modeling systems. This finding highlighted the need for targeted, process-level analyses to reduce error in model estimates of the contributions of these source categories when developing estimates of background ozone.

Emery et al. (2012) extended the traditional methodology for estimating North American background by conducting zero-out simulations with a coupled system, which used both global GEOS-Chem modeling and regional scale (12 km) CAMx modeling simulations informed by the boundary conditions derived from the coarser-scale global simulation. The coupled global-regional modeling yielded slightly higher estimates of North American background (25–50 ppb) than what had been previously been estimated by comparable stand-alone global modeling. These increases were partially attributed to the higher resolution in the regional modeling. Lefohn et al. (2014) further advanced the traditional approach for characterizing background ozone by utilizing a coupled global-regional modeling system that included the CAMx ozone source apportionment technique (Dunker et al., 2002; ENVIRON, 2013) to track the contribution of background sources to total ozone within the simulation. Because historically the NAB definition had been inherently linked to zero-out modeling, Lefohn et al. (2014) introduced a new metric called "emissions-influenced background" (EIB) which represented the combined influence of natural sources and sources of ozone from outside the modeling domain on total modeled ozone, as well as combined chemical interactions between the manmade and background sources. The analyses concluded that EIB could comprise a substantial fraction of the total ozone (e.g., greater than 70%) at high-elevation sites in the western U.S.

Characterizing the contribution of background is particularly important in the western U.S. where the influence of background has been shown to be more substantial than in other parts of the U.S. (Zhang et al., 2011; Emery et al., 2012; Lin et al., 2012a,b; Fiore et al., 2014). While ambient monitoring data can supply limited information about source contributions to observed ozone (e.g., Oltmans et al., 2008; Parrish et al., 2009), it is often necessary to supplement observational assessments of background ozone with photochemical modeling estimates. Given the potential importance of background ozone in parts of the rural western U.S., the motivation of this study was to estimate background for the western U.S. with two distinct modeling techniques that have fundamentally different approaches for estimating the influence of background sources and processes. This paper summarizes the results of modeling conducted for a 2007 warm season period (April–October) using two distinct methodological approaches (zero-out and source apportionment) to estimate the influence of ozone sources other than U.S. anthropogenic emissions of NO_X, carbon monoxide (CO) and VOC on ozone levels in the western U.S. This definition of background is often referred to as U.S. background (USB). We believe this is the first comparative study of zero-out and source apportionment estimates of background ozone. Section 2 of the paper describes the different goals of the two methodologies as well as the coupled global-regional modeling used in each of the modeling configurations. A summary of the base year model performance evaluations is provided in Section 3. The remainder of that section provides estimates of seasonal mean USB ozone and USB ozone on days with base ozone levels that approach or exceed the 2008 ozone NAAQS of 75 ppb.

The 2007 modeling discussed in this paper was originally used as part of EPA's review of the ozone NAAQS. There are a variety of regulatory documents that discuss background ozone as part of EPA's November 2014 ozone NAAQS proposal (Federal Register, 2014; EPA 2014a, EPA 2014b, EPA 2014c). The combination of results from these two separate model characterizations has improved our understanding of background ozone and informed discussions of ways to effectively account for the impacts of USB ozone in Federal, State, and local efforts to attain current and/or future ozone standards.

2. Description of model methodologies

One of the key goals in any consideration of background ozone is to determine what portion of the ozone observed at any given location in the U.S. is due to sources other than domestic anthropogenic emissions (i.e., USB). There are several challenges associated with assessing USB ozone. First, the models that are used to estimate USB have known biases and errors in replicating observed ozone levels. While efforts are made to minimize these errors when the model simulations are configured, different models can produce differing estimates of background depending upon the inputs used (e.g., lateral boundary conditions, wildfire emissions) and built-in model science (e.g., vertical advection, chemistry). Because of the limited availability of ozone and precursor measurements, it is difficult to identify what portion of the total model error is associated with the simulation of background sources and processes, versus the error that may be caused by other aspects of the modeling. Before attempting to quantify USB, it is important to estimate how well the model does at reproducing observed ozone levels at locations and periods that are conducive to high background levels. Section 3.1 of this paper presents the model evaluation results for the 2007 modeling simulations used in this analysis and assesses whether any correlation exists between model error and model estimates of background. While the biases in seasonal mean ozone were generally low across the western U.S., this analysis employs a simple bias correction to constrain the USB estimates with observed ozone concentrations. This correction is described in more detail in Section 3.2.

A second challenge associated with estimating USB ozone is that levels can vary sharply as a function of location, season, episodic emissions events, and synoptic meteorological patterns. As such, it can be difficult to succinctly summarize background levels over a broad region like the western U.S. This paper estimates USB levels during the April–October (i.e., warm season) period when ozone concentrations and background influences are typically greatest over the western U.S. In terms of spatially summarizing the data, USB estimates are provided for each grid cell that contains an ozone monitoring site in the western U.S. that collected data during 2007 and met certain data completeness criteria.

An additional challenge in estimating any definition of background ozone (NAB, USB, EIB, etc.) is that there are can be multiple approaches for utilizing global-regional modeling systems to quantify the contribution of background to total ozone. These techniques generally fall into two different categories: source sensitivity and source apportionment. Each of these model assessment techniques addresses fundamentally different questions about the characteristics of the background. As described in the introduction, most of the previous assessments of background ozone have utilized an emissions "zero-out" sensitivity approach. These types of sensitivity modeling simulations are frequently used to support air quality planning by estimating how air quality levels would respond to reductions in emission loading (EPA 2005, EPA 2009). Zero-out modeling is designed to estimate the amount of ozone that would remain if some set of emissions were completely removed (e.g., manmade emissions within the U.S.). While zero-out scenarios of all U.S. manmade emissions are inherently unrealistic, they can provide an estimate of ozone production caused by manmade emissions within the U.S. A limitation of zero-out sensitivity approaches is that multiple model simulations are required to zero-out each source category individually, and these simulations do not reliably quantify process level contributions to background ozone under realistic ambient conditions because they would not account for the non-linear chemical interaction among different sources of ozone.

Source apportionment methods address the limitation of zero-out sensitivity approaches by using reactive tracers to estimate the contribution of each process-level source of ozone in a single model simulation that realistically treats the interaction of multiple sources (Dunker et al., 2002; ENVIRON, 2013; Lefohn et al., 2014). This approach has also been used to guide air quality planning by estimating the contributions of certain categories of emissions to ozone so that agencies can more effectively identify emissions control strategies to attain the NAAQS (EPA 2005; Kemball-Cook et al., 2009; EPA 2011a; Fann et al., 2013). Thus, source apportionment methods provide a means of estimating the contributions of multiple user-identified source categories, including boundary conditions and other background sources, to ozone formation in a single model simulation. The category-specific contributions to the resulting air quality are estimated using reactive tracer species to track the fate of ozone precursor emissions (VOC and NO_X) and the ozone formation caused by these emissions. Source apportionment modeling (SAM) is designed to determine source-specific contributions to modeled ozone which account for nonlinear chemistry (production and destruction reactions), transport, and deposition (Environ, 2013; Kwok et al., 2014).

Because the term USB has traditionally been defined from a zero-out simulation, we need a new term to refer to the amount of ozone that is formed from sources other than U.S. anthropogenic emissions, when estimated via the apportionment technique. In this paper, we will use the term "apportionment-based USB" (USB_{AB}) to represent that concept. The remainder of the paper will describe, present, and summarize the results from both methodologies over a recent simulation period and directly compare how the two separate approaches estimate USB and USB_{AB} ozone levels over the western states.

2.1. Global modeling and boundary conditions used in regional modeling

Both of the regional scale model simulations used in this analysis utilized lateral boundary conditions from the GEOS-Chem chemical transport model (Bey et al., 2001) that were post-processed to provide hourly values. Version 8-03-02 of GEOS-Chem was exercised over a global 2.0 × 2.5° grid, as described more fully in Henderson et al. (2014). The emissions estimates used in the 2007 base year global modeling were aggregated from a variety of sources, starting with the global Emissions Database for Global Atmospheric Research (EDGAR) emission inventory. These initial estimates were then improved by utilizing various area-specific inventories, such as the 2005 National Emissions Inventory (NEI) for the U.S. portions of the domain, and available inventories for Asia, Canada, Europe, and Mexico. In addition to the anthropogenic estimates, emissions were specified for a variety of natural sources including: lightning NO, soil NO_x, wildfires, and biogenic VOC emissions. The wildfire data is from the Global Fire Emissions Database (GFED) (Van der Werf et al., 2006). The biogenic VOC estimates were simulated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012). The meteorological data is based on the Goddard Earth Observing System Model, Version 5 (GEOS-5) analysis fields. Henderson et al. (2014) also provides a limited evaluation of the GEOS-Chem modeling with respect to its ability to provide accurate lateral boundary conditions of ozone to finer-scale regional simulations during sample cool-season and warm-season months. Using satellite retrievals from the Tropospheric Emissions Spectrometer (TES), it was concluded that the GEOS-Chem ozone prediction biases and errors are generally within TES uncertainty estimates during the warm season. In addition to the base 2007 GEOS-Chem simulation, a separate "zero-out" sensitivity run was also conducted in which all anthropogenic emissions of NO_x and VOC within the United States were removed, and methane levels were set to pre-industrial levels in the global model.

2.2. Regional modeling with CMAQ and CAMx

Lateral boundary conditions extracted from the global model were used as inputs for two regional modeling applications at 12 km horizontal resolution: 1) base and zero-out model simulations that were conducted with the CMAQ model and 2) a regional source apportionment model simulation that was conducted with the CAMx model. The two different models were configured to be as similar as possible. Both the CMAQ zero-out and CAMx source apportionment simulations used a domain that included the 48 contiguous states and small portions of Canada and Mexico, as shown in Fig. 1. Each modeling exercise employed identical 24 vertical layers which extended from the surface to the 50 millibar pressure level (approximately 17 km). The depth of the surface layer was approximately 38 m in both sets of simulations. The meteorological inputs for the regional modeling were derived from an offline simulation of the Weather Research and Forecasting Model (WRF), version 3.1, as described in EPA (2011b). The emissions inputs into CMAQ and CAMx were the same as used for the 2nd draft Health Risk and Exposure Assessment for Ozone (EPA, 2014) and are fully described in Appendix 4-B to that document. The foundation of these emissions estimates was version 2 of the 2008 National Emissions Inventory. For both sets of simulations, the base year modeling was configured to capture average emissions conditions for wildfires and electrical generating units (EGUs) in order to avoid year-specific anomalies in estimated seasonal

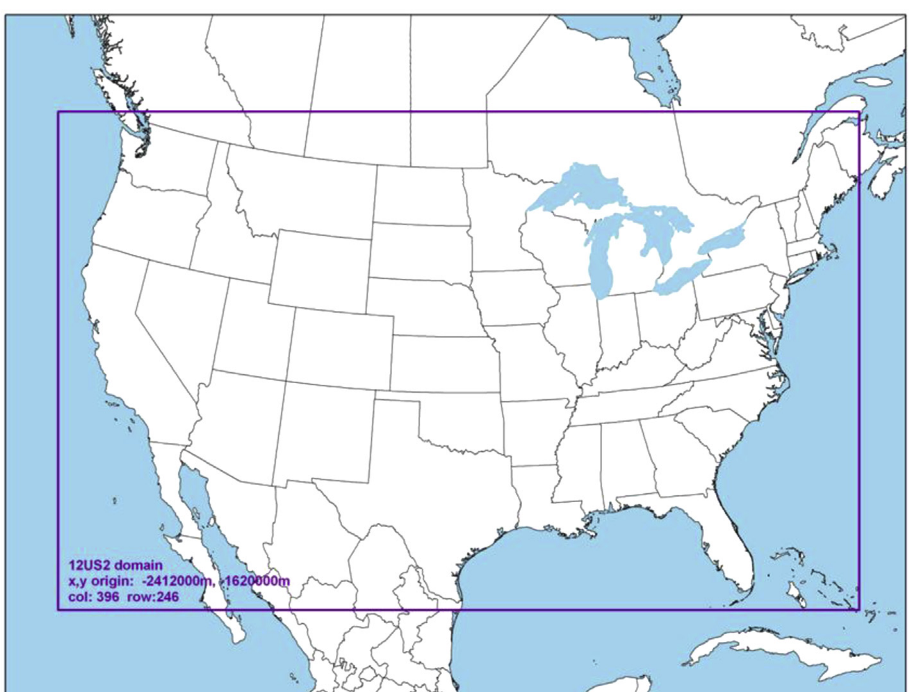

Fig. 1. Map of the modeling domain used in the regional 2007 CMAQ and CAMx modeling.

mean background levels. As such, wildfire data from 2003 to 2010 were averaged together into a typical-day model emissions inventory. EGU emissions were temporalized based on average temporal profiles from three years of data. Biogenic emissions were derived from the Biogenic Emission Inventory System (BEIS) model (Pierce et al., 1998) using the 2007 WRF meteorology as inputs. This version of BEIS has been shown to favorably compare with biogenic VOC measurements made at a high isoprene emitting region of the central United States (Carlton and Baker, 2011).

Two separate CMAQ scenarios were modeled: 1) a 2007 base case simulation and 2) a 2007 USB run with U.S. anthropogenic ozone precursor emissions removed in both the global and regional model simulations. A single CAMx scenario, the 2007 base case simulation, was modeled with source apportionment. Eleven separate source categories were tracked in the source apportionment analysis: five boundary condition terms and six in-domain sectors. The five boundary condition terms included the north, east, south, and west edges, as well as the contribution from the top boundary. The six in-domain sectors consisted of: U.S. anthropogenic emissions, Gulf of Mexico point source emissions, ocean-going marine vessel emissions from outside State boundaries, multi-year average wildfire emissions, biogenic emissions, and Canadian/Mexican emissions inside the modeling domain. For the source apportionment modeling, USB was defined as the sum of the categories noted above except the U.S. anthropogenic emissions and the Gulf of Mexico point source emissions. The apportionment tools in CAMx utilized here to estimate the contribution of background sources have been widely used (EPA 2005, EPA 2011a, Fann et al., 2013). The CAMx Anthropogenic Precursor Culpability Assessment (APCA) approach is used for this analysis (Environ, 2013). Here, the APCA tool attributes ozone production to the biogenic category whenever ozone is determined to result from a combination of both biogenic NO_x and VOC emissions, otherwise contribution is attributed to other categories even when ozone may be limited by biogenic emissions (ENVIRON, 2013).

3. Model results

3.1. Model performance evaluation

For the purposes of this evaluation, given the focus on warm-season estimates of background ozone, we assessed the ability of CMAQ and CAMx to reproduce observed daily maximum 8-h (MDA8) ozone values for the period April to October 2007. As noted above, because this analysis is interested in typical background levels across the western U.S., the base year modeling in this analysis used climatological monthly-average wildfire and typical-day EGU emissions. These inputs are not intended to capture discrete events that occurred in 2007, so perfect correlation between the daily observations and model predictions should not

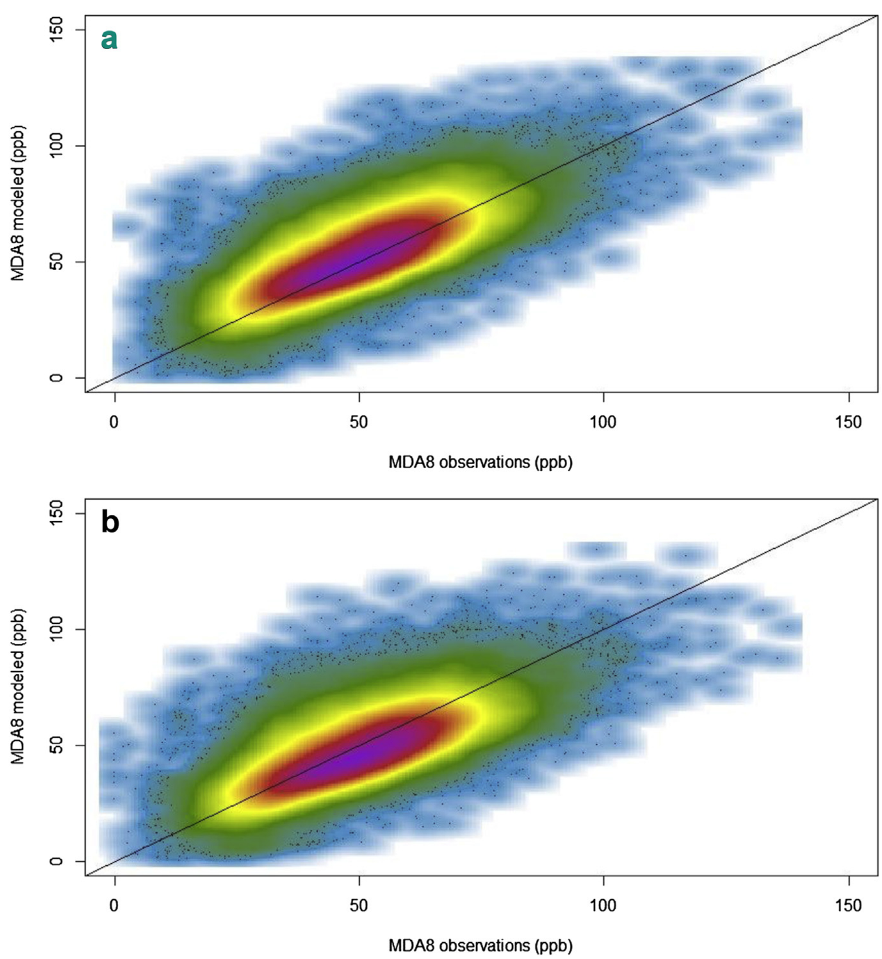

Fig. 2. Density scatterplot comparing CMAQ daily peak 8-h ozone predictions against observed 8-h ozone peaks paired in space and time for all western U.S. sites during April–October 2007. b. Density scatterplot comparing CAMx daily peak 8-h ozone predictions against observed 8-h ozone peaks paired in space and time for all western U.S. sites during April–October 2007.

be expected. Fig. 2a, b are density scatterplots of the observed and predicted daily 8-h ozone peaks paired in space and time for the 2007 base CMAQ and CAMx simulations. The observed data were retrieved from the EPA Air Quality System (AQS) database, as well as from the Clean Air Status and Trends Network (CASTNET). As can be seen, the majority of pairs line up along the 1:1 line. There is a tendency for the models to overestimate site-days with low 8-h ozone peaks, and to underestimate the site-days with higher peak ozone values. Fig. 3a, b depict the spatial bias patterns in both simulations of MDA8 ozone at all western U.S. sites that measured valid ozone data for at least 75 percent of the days during the April–October period. There are similarities and differences in the MDA8 ozone model performance between the CMAQ and CAMx base simulations. Both models exhibit MDA8 ozone overestimations along the southern California coastline and underestimations within the southern portions of the San Joaquin

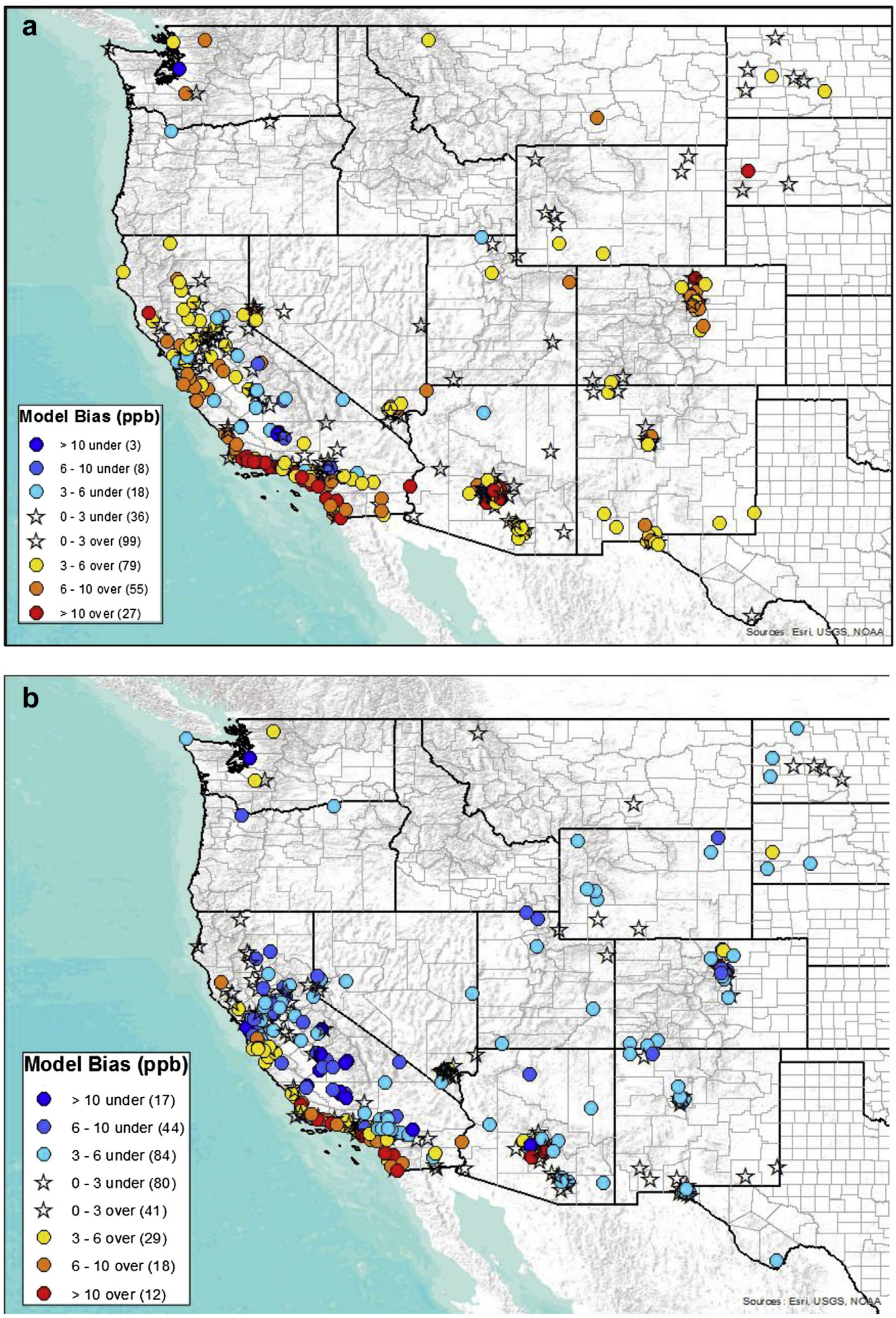

Fig. 3. Site-specific seasonal mean (April–October) maximum daily 8-h ozone biases in the 2007 CMAQ base simulation. Only sites with 75% data capture in 2007 are included. b. Site-specific seasonal mean (April–October) maximum daily 8-h ozone biases in the 2007 CAMx base simulation. Only sites with 75% data capture in 2007 are included.

Valley. While both models are relatively unbiased, CMAQ has a tendency to overestimate seasonal mean MDA8 ozone concentrations, with an aggregate mean MDA8 ozone bias of 3.3 ppb. Conversely, CAMx has a tendency to underestimate seasonal mean MDA8 ozone concentrations, with an aggregate mean MDA8 bias of −2.0 ppb. For the most part, the models reasonably estimate (e.g., most sites within ± 6 ppb) 2007 mean MDA8 ozone levels over the intermountain western U.S. where background influence is expected to be largest. These ozone bias and error values are equivalent or better than typical research grade modeling studies in the

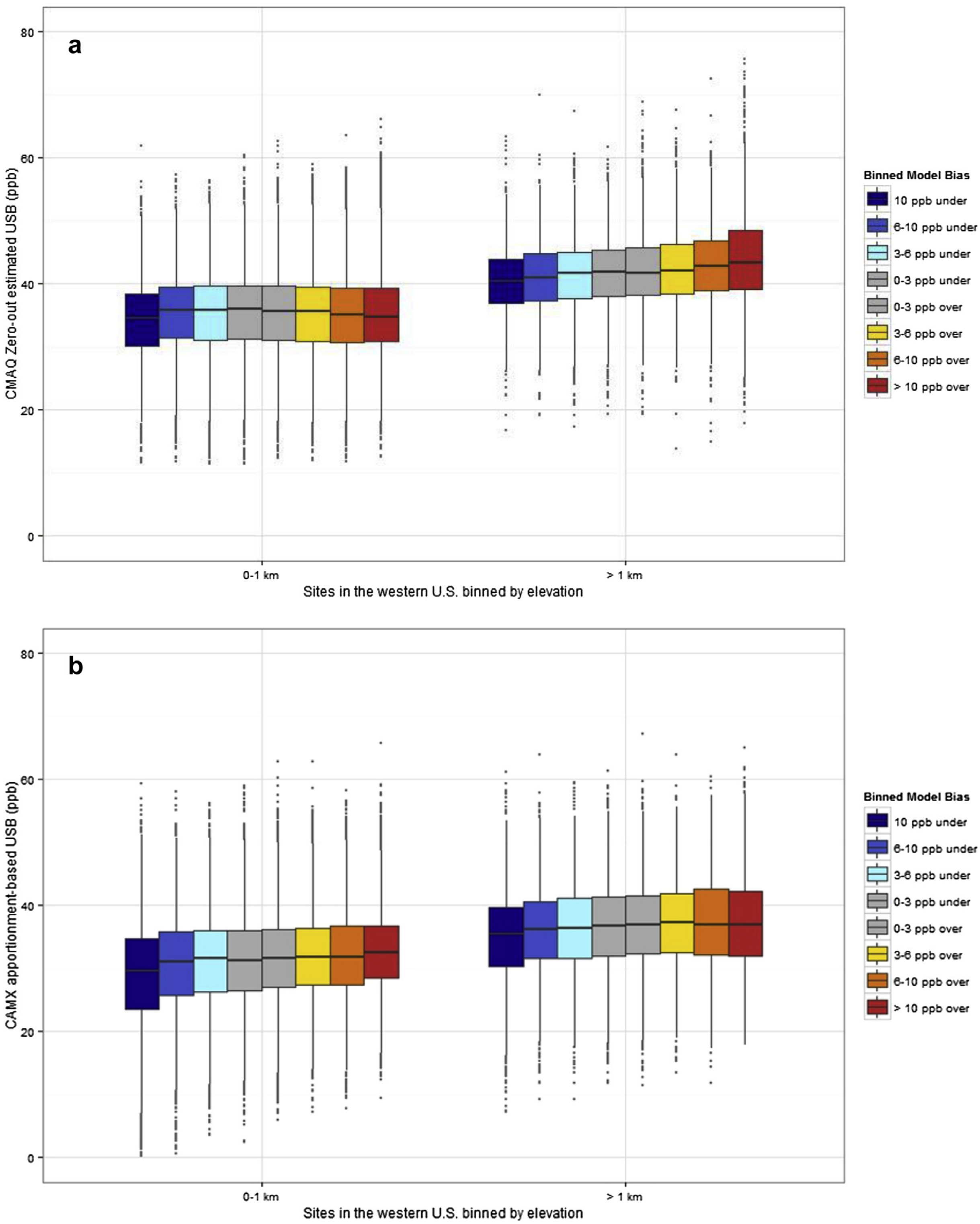

Fig. 4. Comparison of CMAQ estimations of MDA8 USB ozone for eight separate bins of model bias and two separate bins of site elevation. See Section 3.3 for a description of the box and whiskers. b. Comparison of CAMx estimations of MDA8 apportionment-based USB ozone for eight separate bins of model bias and two separate bins of site elevation. See Section 3.3 for a description of the box and whiskers.

peer-reviewed literature (Simon et al., 2012).

Beyond the basic statistical summaries in the previous paragraph, it is important to attempt to diagnose the ability of the models to account for background ozone within the simulations, even without direct observations of this quantity. To determine whether model bias and error may influence the characterization of background ozone, we assessed the relationship between daily model MDA8 ozone biases and same site-day estimates of USB and USB_{AB}. Fig. 4a, b shows the April–October distributions of estimated MDA8 USB and MDA8 USB_{AB} as a function of the site elevation and as a function of the day- and site-specific MDA8 model bias. Ideally, we would see no relationship between the

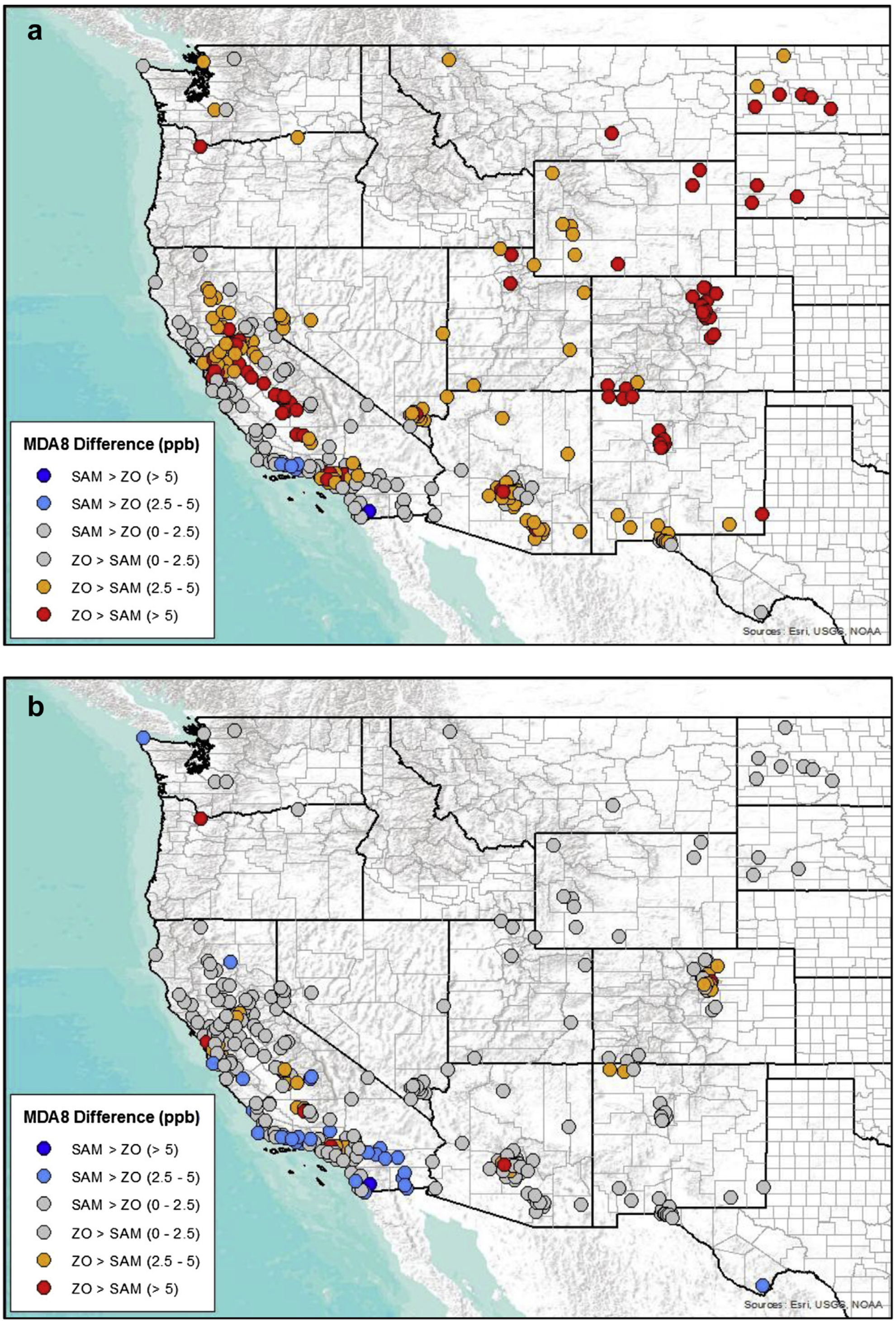

Fig. 5. Difference (ppb) in unadjusted April–October mean MDA8 USB ozone vs mean MDA8 USB_{AB} ozone at monitoring locations across the western U.S. Brighter colors indicate sites where zero out (ZO) estimates of USB exceed source apportionment (SAM) estimates of USB_{AB}. b. Difference (ppb) in bias-adjusted April–October mean MDA8 USB ozone vs. bias-adjusted mean MDA8 USB_{AB} ozone at monitoring locations across the western U.S. Brighter colors indicate sites where zero out (ZO) estimates of USB exceed source apportionment (SAM) estimates of USB_{AB}. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

background estimates and model bias. This would suggest that model estimates of background are not a function of fundamental artifacts in the modeling related to background processes. Fig. 4a indicates that little relationship exists between CMAQ-estimated USB and CMAQ biases at lower-elevation sites (i.e., sites located less than 1 km above sea level). However, Fig. 4a does indicate that there is a weak relationship between CMAQ-estimated USB and model performance for sites above 1 km. Note that the median USB values for the days in which MDA8 ozone is overestimated by more than 10 ppb days are approximately 4–5 ppb higher than the USB estimates that occur on days with greatest underestimation. This implies that part of the model error at high elevation sites in the western U.S. may be related to overestimates of background ozone in the CMAQ model. Conversely, as shown in Fig. 4b, there appears to be very little relationship between model bias and model estimates of background in the CAMx source apportionment modeling.

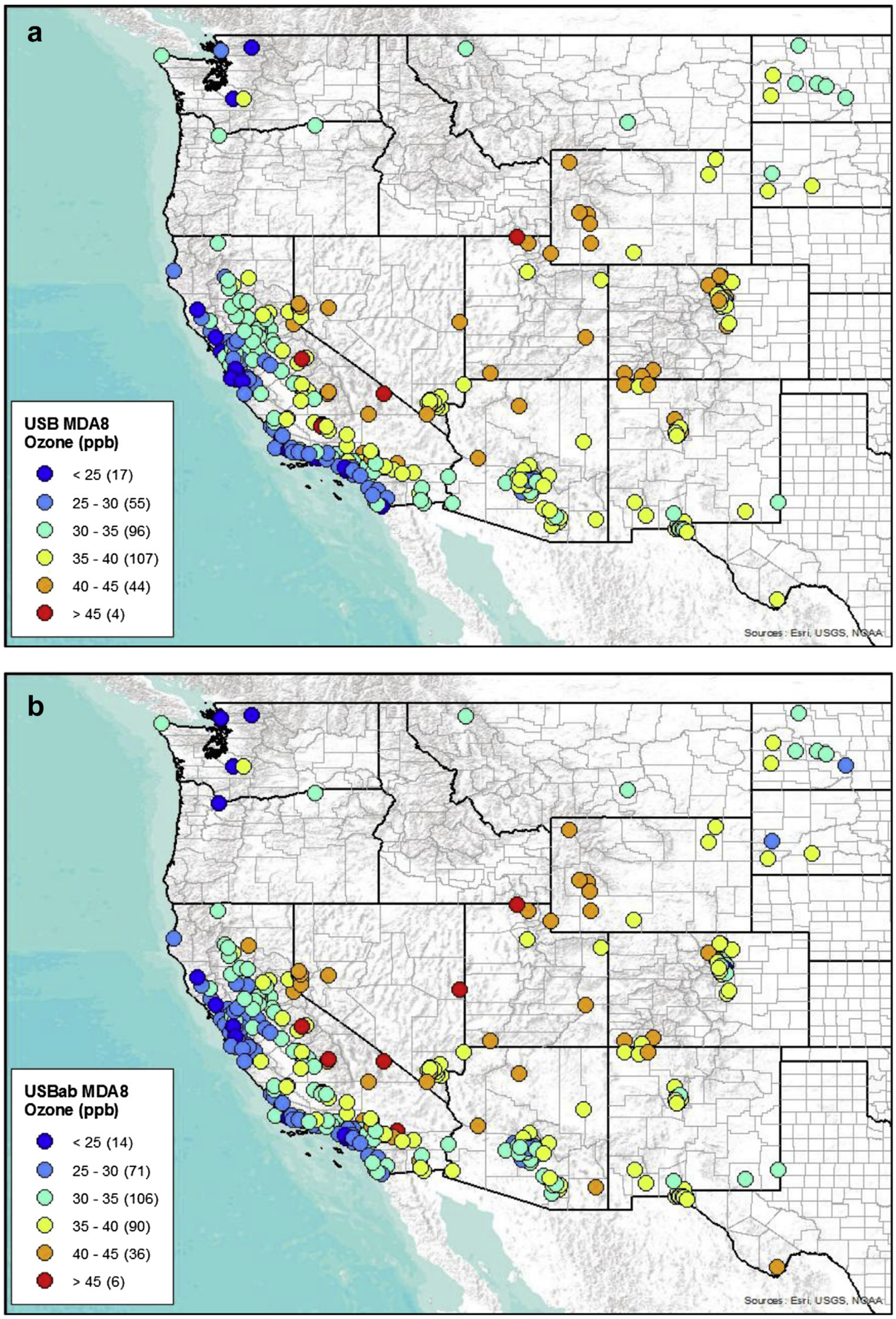

Fig. 6. April–October mean bias-adjusted USB MDA8 ozone (ppb) at monitoring locations across the western U.S., as estimated by a 2007 CMAQ zero out simulation. b. April–October mean bias-adjusted USB_{AB} MDA8 ozone (ppb) at monitoring locations across the western U.S., as estimated by a 2007 CAMx source apportionment simulation.

3.2. Model estimates of seasonal mean background ozone

Because of the potential differences in background estimates resulting from model performance differences across the two simulations, a bias adjustment was made to the raw background estimates to account for deviations between the base case modeled and observed ozone concentrations. The adjustment assumes that the proportion of model bias attributed to background is the same

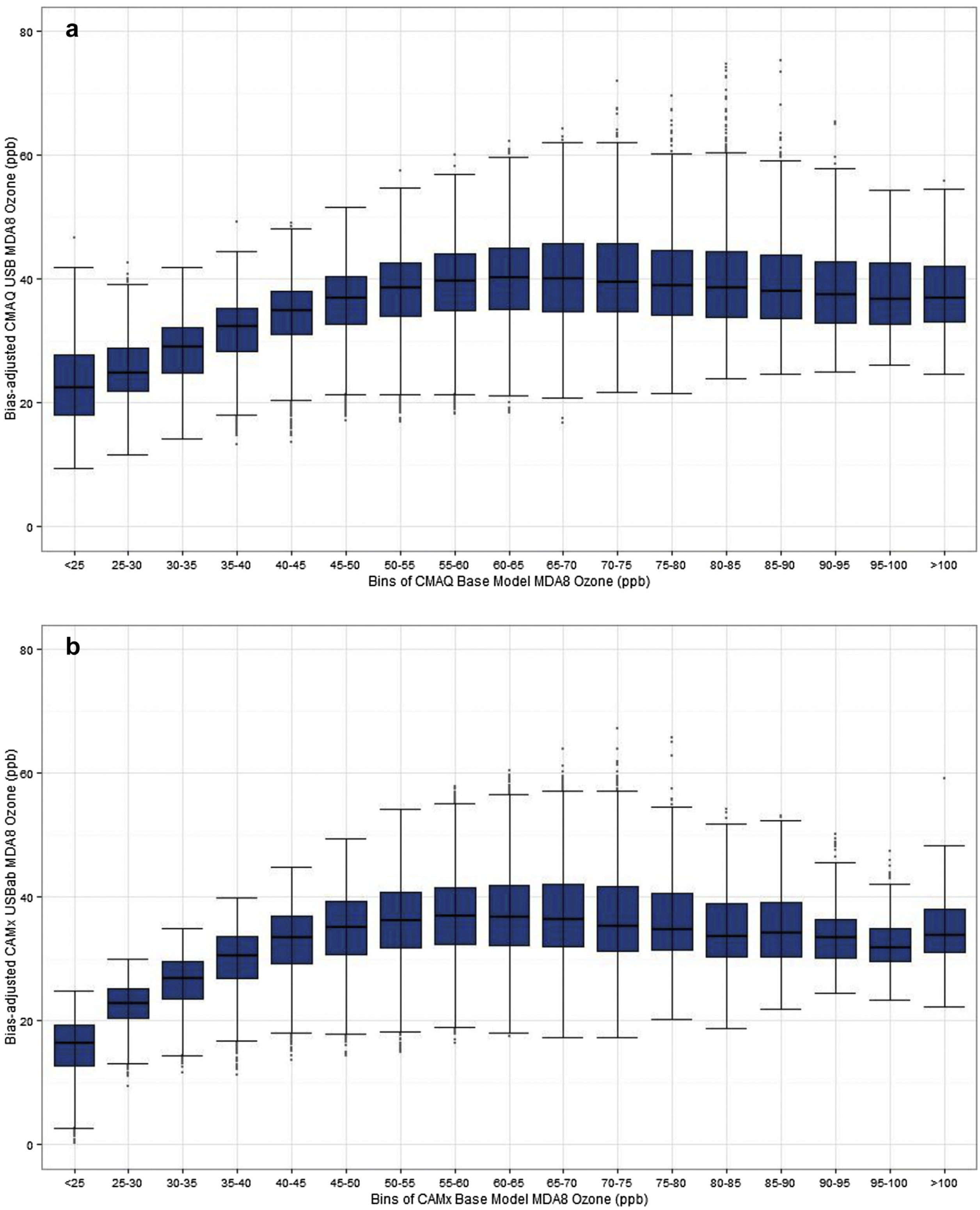

Fig. 7. CMAQ-estimated daily distributions of bias-adjusted USB MDA8 ozone (ppb) at monitoring locations across the western U.S. for the period April–October 2007, binned by base model ozone. b. CAMx-estimated daily distributions of bias-adjusted USB_{AB} MDA8 ozone (ppb) at monitoring locations across the western U.S. for the period April–October 2007, binned by base model ozone.

as the proportion of total ozone attributed to background. Daily model calculated USB/Base MDA8 ozone fractions are multiplied by daily MDA8 bias at each monitoring location and the product is then subtracted from to the original USB (or USB_{AB}) estimate. For example, for a day-site combination with a base MDA8 prediction of 50 ppb that underestimates the observed MDA8 by 10 ppb and a USB estimate of 30 ppb (i.e., 60% of the total is background), the bias-adjustment would add 6 ppb to the original estimate (i.e., 36 ppb). Fig. 5a, b shows the difference between the estimates of USB and USB_{AB}, before and after the simple bias adjustment is made. Prior to the application of the bias-adjustment, there was a clear tendency for the CMAQ zero-out modeling to estimate higher levels of USB MDA8 ozone, compared to source apportionment, over most sites in the western U.S. Constraining the USB and USB_{AB} estimates for model bias brings the estimates across the two methodologies much closer together, especially across the inter-mountain western U.S. and the San Joaquin Valley, where most sites have differences of less than 2.5 ppb when base model biases are accounted for. Over 75 percent of the sites shown in Fig. 5b feature USB-USB_{AB} differences smaller than $\pm$ 2.5 ppb. The similarity in the estimates of U.S. background are encouraging and bolster confidence in our overall characterization of background in the western U.S.

Fig. 6a, b shows the bias-adjusted model estimates for USB and USB_{AB} from the CMAQ and CAMx models, respectively. The spatial patterns between the two sets of estimates are similar with the largest U.S. background impacts occurring in the intermountain western U.S. The zero out modeling estimates that seasonal mean USB levels range from 40 to 45 ppb over many locations in the intermountain western U.S. (e.g., 48 sites exceed 40 ppb). Locations with USB concentrations greater than 40 ppb are confined to Colorado, Nevada, Utah, Wyoming, northern Arizona, eastern California, and parts of New Mexico. The source apportionment modeling suggests that seasonal mean USB_{AB} levels are similar across this region (e.g., 42 sites have seasonal mean USB_{AB} greater than 40 ppb). Both sets of modeling show the lowest background concentrations (25–35 ppb) occur along the Pacific coast.

3.3. Model estimates of background ozone on high ozone days

As a first-order understanding, it is valuable to be able to characterize seasonal mean levels of background ozone. However, it is well established that background levels can vary substantially from day-to-day. From a policy perspective, estimating the contribution of background ozone on higher ozone days (e.g., days that approach or exceed the current ozone NAAQS of 75 ppb) is a more meaningful construct. The 2007 modeling completed as part of this study is consistent with the finding from previous modeling analyses (Henderson et al., 2012) and shows that the highest modeled ozone site-days tend to have background ozone levels similar to mid-range ozone days. Fig. 7a displays the distribution of April–October bias-adjusted USB MDA8 levels from the CMAQ zero out run. The "box and whisker" plots shown in these figures display four key features of the distributions: a) the median concentration (black horizontal line) per bin, b) the inter-quartile range (blue colored box) (in web version) which represents the 25th-75th percentile range in values within the distribution, c) the "whiskers" (dark gray vertical lines with top and bottom whiskers) which represent the range of values within 1.5 times the inter-quartile range, and d) the "outliers" (gray points) which are any values outside the whiskers. Median values and the interquartile range of site-day USB MDA8 levels are essentially unchanged for base model ozone concentrations above 50 ppb, and decline slightly above about 80 ppb. Fig. 7b shows the same graph for bias-adjusted USB_{AB}. Keep in mind that each bin may contain differences in the regional representativeness of the data (e.g., highest bins are mostly locations in southern California). For the very lowest values, when ozone concentrations are less than 25 ppb, the estimates of background contribution are much lower in the CAMx source apportionment modeling compared to the CMAQ zero-out modeling. This is not unexpected as zeroing out emissions can remove the effects of local NO titration and other chemical reactions that destroy ozone. In the CAMx source apportionment approach, some background ozone is destroyed by these chemical reactions, which results in modeled USB_{AB} values that are lower than the zero out approach. The USB_{AB} distributions indicate that the highest median values of background tend to occur in association with mid-range ozone values (i.e., 50–70 ppb) and decline slightly above about 70–75 ppb.

Fig. 8a, b shows the equivalent plots as Fig. 7a, b, but use background fractions (background MDA8/base MDA8) as the dependent variable instead of the absolute background concentrations. These plots show the same effect; that is, the relative contribution of background sources and processes decreases as peak ozone increases. In the zero out modeling, the median fractions drop from approximately 80% USB for values between 45 and 50 ppb to approximately 55% USB for base MDA8 values between 70 and 75 ppb. Again, the source apportionment modeling estimates a slightly lower relative contribution from non-U.S. anthropogenic emissions. Based on that modeling approach, the median fractions drop from approximately 75% background for values between 45 and 50 ppb to only 50% background for base MDA8 values between 70 and 75 ppb. It should be noted that both modeling techniques suggest that there are some high ozone days where sources other than U.S. manmade emissions are contributing substantially to the total modeled ozone. These high cases are more common in the zero-out modeling than in the source apportionment modeling.

Fig. 9a, b displays the spatial estimates of USB and USB_{AB} fractions only on days in which the base MDA8 ozone at the site was above the 90th percentile value for that site-season (a metric which is not comparable to the form of the current ozone NAAQS, but which was chosen here as an indicator of a high ozone day). While there are some site-to-site differences, for the most part the two methodologies yield similar estimates of background influence on these highest modeled ozone days, as was also seen for the seasonal mean background. In rural portions of the western U.S., it is not uncommon for high ozone days (e.g., days in the worst 10 percent for a season) to be affected substantially (e.g., >70–80 percent) by U.S. background ozone. The sites that are proportionately most affected by background tend to be the most rural sites and not urban areas in the western U.S. like Denver, Phoenix, or Las Vegas where the high days are estimated to be 40–60 percent background.

4. Summary and discussion

For a variety of reasons, it is challenging to present a comprehensive summary of all the components and implications of background ozone. In many forums the term "background" is used generically and the lack of specificity can lead to confusion as to what sources are being considered. Additionally, it is well established that the impacts of background sources can vary greatly over space and time which makes it difficult to present a simple summary of background ozone levels. Further, background ozone can be generated by a variety of processes (e.g., fires, stratospheric intrusion, biogenic and geogenic sources), each of which can lead to differential patterns in space and time. Finally, background ozone is difficult to measure directly and thus, air quality modeling must be used to assess background levels. As with any other modeling exercise, these simulations will have uncertainties and potential biases and error. The results presented here might be affected by

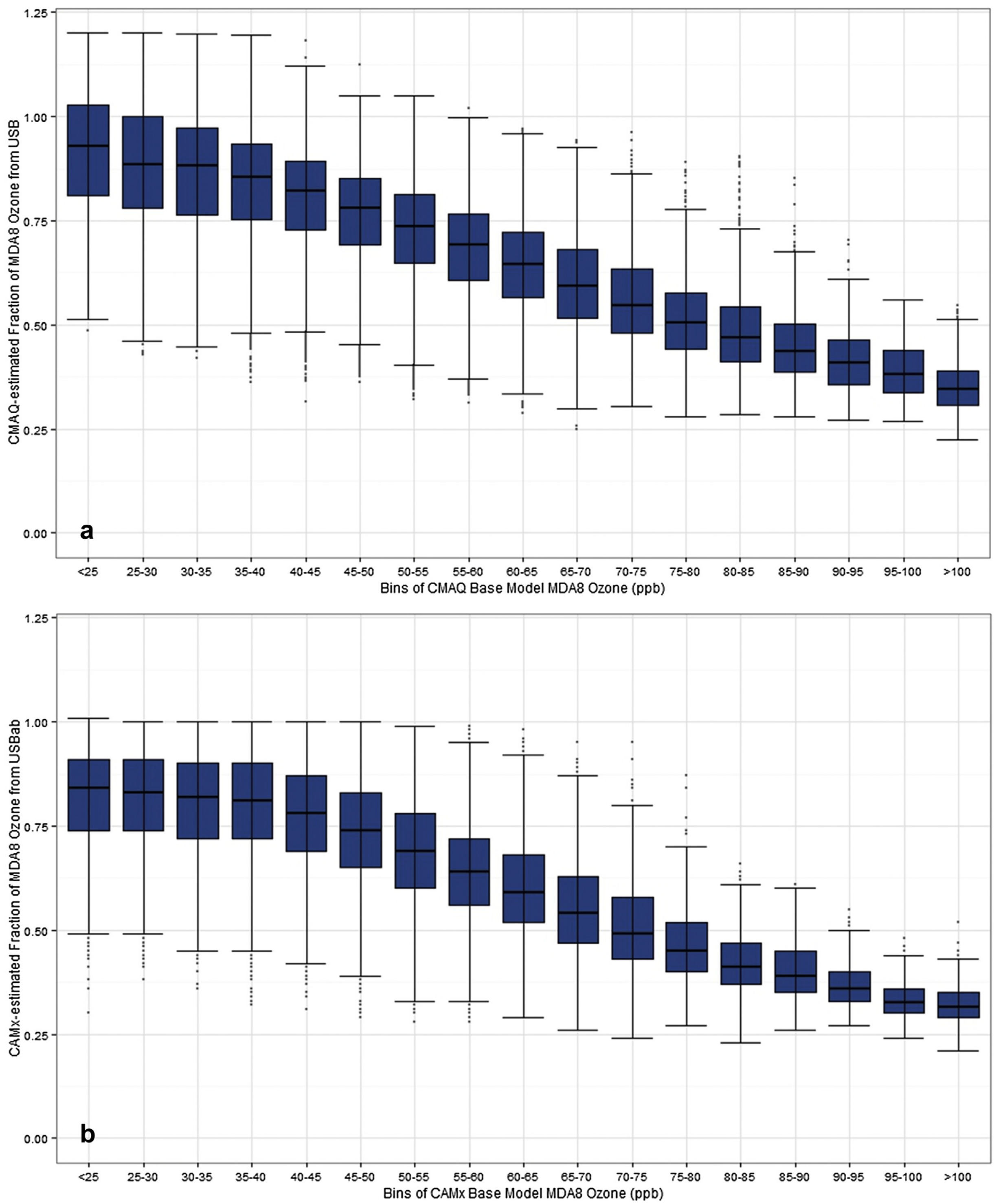

Fig. 8. CMAQ-estimated daily distributions of bias-adjusted USB ozone fractions at monitoring locations across the western U.S. for the period April–October 2007, binned by base model MDA8 ozone. b. CAMx-estimated daily distributions of bias-adjusted USB_{AB} ozone fractions at monitoring locations across the western U.S. for the period April–October 2007, binned by base model MDA8 ozone.

systematic error or bias in model inputs, including emissions and boundary conditions. Differences in the algorithms used to represent transport and dispersion in CMAQ and CAMx likely also contribute to differences in the model predicted background levels.

The results from this 2007 warm season modeling analysis with two distinct model methodologies contribute to an emerging conceptual model of the impact of background sources to ozone in the western U.S. From a seasonal mean perspective, it is not uncommon for modeled MDA8 USB ozone values at monitors in the intermountain western U.S. to approach 40–45 ppb and for USB to represent 60–80 percent of total ozone. Higher ozone days tend to have smaller fractional contributions from background and larger influences from domestic sources of ozone precursors. However, these analyses suggest that U.S. background can comprise a

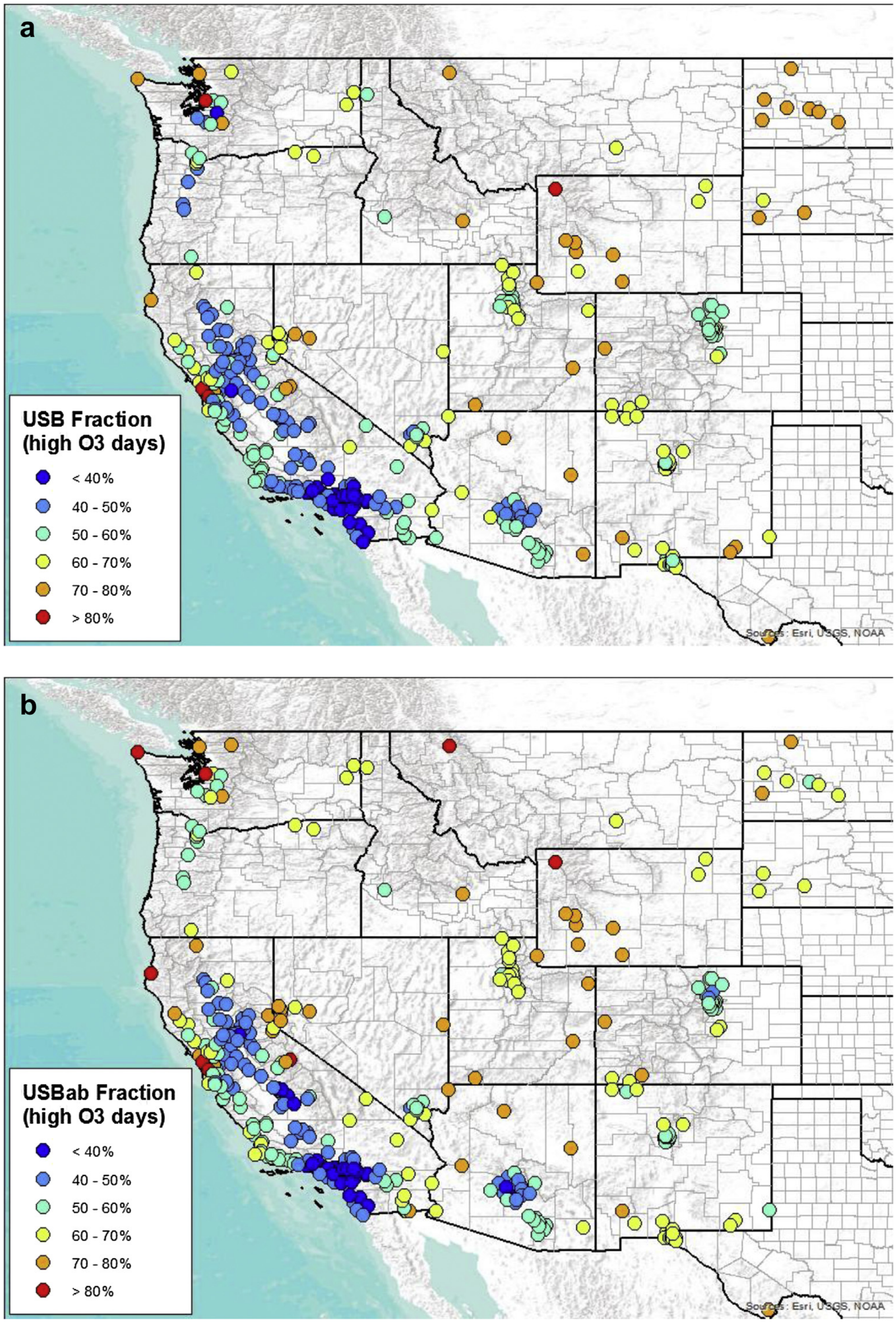

Fig. 9. CMAQ-estimated bias-adjusted USB ozone fractions at monitoring locations across the western U.S. for the period April–October 2007, on base model days with MDA8 ozone >90th percentile levels. b. CAMx-estimated bias-adjusted USB_{AB} ozone fractions at monitoring locations across the western U.S. for the period April–October 2007, on base model days with MDA8 ozone >90th percentile levels.

substantial portion (e.g., >75 percent) of the total daily ozone at certain rural, high-elevation sites in the western U.S. The emissions inventory used in the modeling was based on typical wildfire and EGU emissions data, and the use of actual emissions data might result in larger estimates of background ozone on days that are influenced by large, atypical wildfire events. Because the two distinct model approaches estimate similar background impacts over the rural portions of the western U.S., we believe greater confidence can be placed on the combined results. While the CAMx and CMAQ model simulations used here provided consistent estimates of rural USB ozone levels in the western U.S., it was noted that the CAMx source apportionment approach predicts lower background contributions in urban areas, as expected, because anthropogenic emissions react with and destroy some fraction of the ozone in the CAMx tracer species used to track the background ozone contribution. The zero-out modeling provides another useful estimate of the influence of background ozone because it quantifies the contribution of background in the absence of US anthropogenic emissions.

Additional research is needed to more thoroughly evaluate the model performance in the rural and remote western US. There were only a limited number of rural ozone monitoring sites available for model evaluation in 2007, however, new ozone monitors began operation in remote areas in the inter-mountain west in 2010 and 2011 (Wheeler et al., 2011). Accurate representation of exchange between the free troposphere and boundary layers is an important uncertainty in modeling background ozone at high elevation sites. Emery et al. (2011) found that excessive vertical transport from the free troposphere to the surface caused an earlier version of the CAMx model to overestimate surface ozone mixing ratios, and an updated vertical advection algorithm was developed to improve model performance. However, few ozone vertical profile measurements are available at high elevation western sites, so it is difficult to evaluate how well models simulate this important process. The DISCOVER-AQ and FRAPPE field studies were carried out in Colorado in July–August 2014, and these studies include extensive aircraft and ground based LIDAR vertical profiles of ozone. Eventual modeling studies for other year (e.g., 2014) that include these new data will allow more accurate assessments of background ozone at high elevation sites in the western U.S.

Disclaimer

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy.

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