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Air pollutant source characterization using the revised regional haze tracking metric and a photochemical grid model and implications for regional haze planning

Patricia Brewer[a], **Gail Tonnesen**[b], **Ralph Morris**[c], **Tom Moore**[d], **Uarporn Nopmongcol**[c], **Debra Miller**[e]

[a]Air Resources Division, National Park Service, Lakewood, CO, USA;

[b]Environmental Protection Agency, Region 8, Air Program, Denver, CO, USA;

[c]Ramboll, Novato, CA, USA;

[d]Western States Air Resources Council and Western Regional Air Partnership, Fort Collins, CO, USA;

[e]Natural Resources Division, Intermountain Region, National Park Service, Lakewood, CO, USA

Abstract

The 2017 revisions to the Regional Haze Rule clarify that visibility progress at Class I national parks and wilderness areas should be tracked on days with the highest anthropogenic contributions to haze (impairment). We compare the natural and anthropogenic contributions to haze in the western United States in 2011 estimated using the Environmental Protection Agency (EPA) recommended method and using model projections from the Comprehensive Air Quality Model with Extensions (CAMx) and the Particulate Source Apportionment Tool (PSAT). We do so because these two methods will be used by states to demonstrate visibility progress by 2028. If the two methods assume different natural and anthropogenic contributions, the projected benefits of reducing U.S. anthropogenic emissions will differ. The EPA method assumes that episodic elevated carbonaceous aerosols greater than an annual 95th percentile threshold are natural events. For western U.S. IMPROVE monitoring sites reviewed in this paper, CAMx-PSAT confirms these episodes are impacted by carbon from wildfire or prescribed fire events. The EPA method assumes that most of the ammonium sulfate is anthropogenic in origin. At most western sites CAMx-PSAT apportions more of the ammonium sulfate on the most impaired days to global boundary conditions and anthropogenic Canadian, Mexican, and offshore shipping emissions than to U.S. anthropogenic sources. For ammonium nitrate and coarse mass, CAMx-PSAT apportions greater contributions to U.S. anthropogenic sources than the EPA method assigns to total anthropogenic contributions. We conclude that for western IMPROVE sites, the EPA method is effective in selecting days that are likely to be impacted by anthropogenic emissions and that CAMx-PSAT is

CONTACT Patricia Brewer patricia_f_brewer@nps.gov Air Resources Division, National Park Service, 7333 W. Jefferson, Lakewood, CO 80227, USA.

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an effective approach to estimate U.S. source contributions. Improved inventories, particularly international and natural emissions, and further evaluation of global and regional model performance and PSAT attribution methods are recommended to increase confidence in modeled source characterization.

Introduction

The 1999 Regional Haze Rule (RHR; U.S. EPA 1999) implemented the 1977 Clean Air Act Amendments' mandate to protect visibility in Federal Class I national parks and wilderness areas. The 1999 Rule instructed states to submit plans every 10 years that demonstrate reasonable progress toward the goal of achieving natural visibility conditions on most impaired days by the year 2064. Most impaired days were defined as days with highest total haze. For many Class I areas in the western United States, episodes of elevated carbonaceous aerosols or crustal material, attributable to wildfires or dust storms, respectively (Hand et al. 2013, 2016), dominate aerosol concentration on haziest days and make it more difficult for states to demonstrate visibility progress due to reductions in U.S. anthropogenic emissions.

In January 2017, the Environmental Protection Agency (EPA) revised the Regional Haze Rule (U.S. EPA 2017), including the metric used to track visibility progress. Instead of tracking the 20% haziest days in each year, states are now to track visibility progress on the 20% of days with the highest anthropogenic impairment. In the 2016 proposed Guidance for the second regional haze implementation period (U.S. EPA 2016a) and the accompanying Technical Support Document (U.S. EPA 2016b), the EPA recommended using operationally defined estimates of natural and anthropogenic pollutant contributions (EPA method) based on the measured chemical aerosol species concentrations and calculated aerosol light extinction from the Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring network. The revised definition of most impaired days is intended to limit the influence of episodic wildfires and dust storms on the visibility tracking metric.

States are to define a uniform rate of visibility progress (URP) for most impaired days that is calculated as the straight line glidepath between visibility for the 2000–2004 baseline period and estimated natural visibility conditions for the target year 2064, as illustrated in Figure 1. Average natural contributions on the most impaired days in the years 2000 to 2014 are used to represent the natural conditions endpoint in 2064. The western states will use a photochemical grid model to demonstrate visibility progress by 2028 compared to the URP. If the modeled estimates of natural and anthropogenic pollutant contributions differ from EPA's estimates, the two approaches could project different rates of visibility progress by 2028. States can propose methods to adjust the 2064 endpoint to account for contributions from international anthropogenic emissions and/or prescribed fires. A photochemical grid model is one approach being considered by the western states to apportion contributions from U.S. anthropogenic, U.S. natural, and international emissions to the IMPROVE observations and evaluate natural condition assumptions in the modeled years.

The objective of our analysis is to compare the EPA method to assign natural and anthropogenic contributions to results of the Comprehensive Air Quality Model with Extensions (CAMx; Ramboll 2018) with the Particulate Source Apportionment Tool

(PSAT). In this paper we (1) describe the methods used in the EPA and CAMx-PSAT approaches as applied for 2011 IMPROVE aerosol data for western Class I areas, (2) compare the natural and anthropogenic contributions assigned by the two methods, (3) consider the influence of boundary conditions on model results, and (4) discuss the implications for demonstrating the visibility benefits of reducing U.S. anthropogenic emissions for western Class I areas in the second regional haze planning period.

Methods

EPA's recommended method to track visibility progress

The EPA method relies on the measured chemical aerosol species concentrations and calculated aerosol light extinction from the IMPROVE monitoring network (Hand et al. 2011). Light extinction is calculated for each IMPROVE sample day (24-hr aerosol samples collected every third day) based on measured aerosol composition and mass using parameters for aerosol extinction efficiency that vary as a function of relative humidity, and clear air Rayleigh scattering in the atmosphere (Pitchford et al. 2007). Aerosol light extinction is expressed in inverse megameters (Mm^{-1}.) The RHR visibility tracking metric is a haze index expressed in terms of deciview (dv), calculated as the logarithm of cumulative light extinction due to all aerosol and gas species (Pitchford and Malm 1994) as

$$\mathrm{dv} = 10 \ln\left(\mathrm{bext}/10\ \mathrm{Mm}^{-1}\right) \quad (1)$$

where the denominator in the log term is a default constant value for clear air scattering.

Terms and definitions relevant to the revised visibility tracking metric are summarized in Table 1. EPA's revised visibility tracking metric focuses on the days with the highest fraction of anthropogenic impairment. The EPA method recommends operational definitions to assign IMPROVE aerosol extinction to natural episodic extreme events and daily routine natural contributions (USEPA, 2016a). Site-specific thresholds define episodic extreme events for carbonaceous species (organic matter estimated from carbon, OMC, plus elemental carbon, EC) and crustal material (fine soil plus coarse mass), as indicators of wildfires and dust storms, respectively. The recommended threshold is the minimum of the annual 95th percentile daily aerosol extinction values during the 15-year period 2000–2014. Daily species extinction values greater than the 95th percentile threshold are assigned as episodic natural in origin. Smaller, routine natural contributions from biogenic or geogenic emissions are assumed to be a constant fraction of the IMPROVE aerosol extinction on each day, with the daily fraction calculated as the ratio of a previously estimated annual average natural extinction ("Natural Conditions II," NCII; IMPROVE 2007) divided by the annual average calculated extinction for each species. For example, the NCII estimated annual average natural ammonium nitrate extinction for Glacier NP is 0.95 Mm^{-1}, and the 2011 calculated annual average ammonium nitrate extinction at Glacier NP is 1.47 Mm^{-1}; thus, 65% of the daily ammonium nitrate extinction at Glacier NP is assumed to be routine natural in 2011. This same calculation format applies for ammonium sulfate. For carbon and dust species, the fraction of routine natural is calculated based on the annual average nonepisodic (total minus episodic) aerosol extinction. All sea salt extinction and Raleigh scattering are assigned as natural. The remainder of total extinction not assigned to natural contributions is

assumed to be anthropogenic in origin. Daily anthropogenic impairment is calculated as the difference between total extinction and natural extinction as

$$dv_{anthropogenic} = 10 \ln (bext_{total}/bext_{natural}) \quad (2)$$

Daily impairment values are ranked from highest to lowest impairment to select the 20% most impaired days in each year. The URP glidepath for the most impaired days uses the 5-year average deciview for the 2000–2004 baseline period as the starting point for the straight line slope to estimated natural conditions in 2064 (U.S. EPA 2016a). The 2064 endpoint of the glidepath is estimated as the 15-year (2000 to 2014) average of annual average natural conditions on the 20% most impaired days in each of the 15 years. The reader is referred to EPA draft guidance (U.S. EPA 2016a), Technical Support Document (USEPA 2016b), and Gantt et al. (2018) for further details. Uncertainties in the EPA method could affect the days in a calendar year that are selected as most impaired and the estimates of natural conditions on most impaired days that contribute to the 2064 endpoint for the URP glidepath.

CAMx-PSAT source apportionment in the 2011 Western Air Quality Study

Visibility on the most impaired days is influenced by emissions from diverse sources and geographic areas (Supplement Table 1). Source attribution tools can estimate contributions to haze from U.S. anthropogenic, international, and natural emissions. The CAMx-PSAT model can track, within the limits and accuracy of the model, specific source categories and boundary transport and provide evidence for emission contributions to haze. The CAMx-PSAT source apportionment technique has been evaluated against sensitivity methods (e.g., Dunker et al. 2002; Koo et al. 2009). Additional details on model formulation are available in the CAMx user's guide (Ramboll 2018).

We took advantage of an existing modeling exercise for the 2011 Western Air Quality Study that was completed in 2016 (prior to EPA's revised RHR guidance.) The CAMx (version 6.10) photochemical grid model was applied for the 2011 annual period with spin-up days in the end of December 2010 (Adelman et al. 2016). Boundary conditions were defined using the Model for Ozone and Related Chemical Tracers (MOZART) global chemistry model (Emmons et al. 2015). Boundary conditions from the MOZART model may include some contributions from U.S. anthropogenic emissions through flow reversals or recirculation around the globe. U.S. emissions were based on the 2011 National Emissions Inventory version 2 (NEI; U.S. EPA 2015) with updates to several source categories including oil and gas sources (Parikh, Grant, and Bar-Ilan 2015) and fire emissions (Mavko, Moore, and Morris 2016). Global inventories include 2005 Canadian and Mexican emissions and 2006 Asian and European emissions, as described in Emmons et al. (2015).

Particulate matter source apportionment modeling for the 2011 annual period was conducted using the CAMx-PSAT tool. CAMx-PSAT uses reactive tracers that operate in parallel to the host model and uses the host model scientific algorithms (e.g., transport, chemistry, and deposition) to calculate the contributions of user-defined source regions and/or source categories to each particulate matter (PM) species. The CAMx-PSAT source apportionment tool traces back PM species to their primary precursor. Thus, sulfate (SO_4) is traced back to SO_2 emissions, nitrate (NO_3) to nitrogen oxide (NO_x) emissions, ammonium (NH_4) to

ammonia (NH_3) emissions, and primary PM species (OMC, EC, fine crustal and coarse mass) to their respective emissions. Volatile organic compounds (VOC) are mapped to secondary organic aerosols (SOA) that are operationally defined as natural or anthropogenic (e.g., SOA due to isoprene and terpene VOC emissions were assigned to natural sources, and SOA due to aromatic VOC emissions were assigned to anthropogenic sources). In this analysis we assumed (consistent with IMPROVE) that SO_4 and NO_3 were fully neutralized by NH_4, and the CAMx-PSAT results were not used to track the NH_4 particulate concentrations back to the NH_3 emissions.

For visibility at Class I areas, the 2011 Western Air Quality Study (WAQS) applied CAMx-PSAT on the 36-km Continental U.S. (CONUS) domain for the numbered source regions in Figure 2 (Intermountain West Data Warehouse 2016). Modeled PM aerosol contributions were separated into anthropogenic and natural emission categories and by western state, eastern United States, Mexico, and Canada inside the 36-km CONUS domain and the remainder of international transport through the boundary conditions outside the 36-km CONUS domain. The 2011 WAQS CAMx-PSAT modeling results were used to apportion the daily measured IMPROVE species as follows (see Supplement Table 1 for additional description):

- U.S. anthropogenic emissions, including agricultural burning but not prescribed fires.
- U.S. background (USB):
  - Natural emissions in the CONUS domain.
    - Biogenic, lightning NO_X, windblown dust, sea salt.
    - Wildfires, prescribed fires.
  - International transport.
    - Anthropogenic emissions in CONUS domain attributed to Mexico, Canada, and marine shipping.
    - MOZART CONUS boundary conditions (natural and anthropogenic fractions were not separated).

A comprehensive model performance evaluation for the 2011 WAQS is reported in Adelman et al. (2016). Figure 3 illustrates model performance at 36 km grid resolution for speciated PM for the average of the EPA-defined 20% most impaired days at the six IMPROVE monitors for the Class I areas identified in Figure 2. These sites were selected to represent a broad range of geographic areas and PM species contributions. At these sites, performance was best for SO_4, EC, and fine soil, with some over predictions of OMC, under prediction of coarse mass, and variable performance for NO_3. The CAMx-PSAT source apportionment has larger uncertainties for those species with poorer model performance. OMC model performance is more uncertain because fire emissions are highly uncertain and are a major component of primary OMC in the western United States. Secondary organic aerosol (SOA) is a major component of routine OMC concentrations, and biogenic emissions (e.g., terpenes and isoprene) are a major component of SOA. The chemistry of nitrate formation is complex

depending on temperature and availability of cation buffering compounds (e.g., ammonia and minerals in dust) to form particulate nitrate. The CAMx 2011 base case was also evaluated for gaseous species, with ozone performance generally good, better at rural than urban sites, and underestimating the highest observed values. Nitrogen dioxide (NO_2) was generally overestimated, while SO_2, CO, and NH_3 were underestimated (Adelman et al. 2016).

CAMx produces output for all 365 days in a modeled year, while the IMPROVE network samples up to 122 days per year (1:3 day sampling frequency). For this analysis, we used the IMPROVE days defined by the EPA method as the 20% most impaired days in the 2011 (U.S. EPA 2016b). To account for differences between the modeled and measured aerosol values, the CAMx-PSAT source apportionment results were used to calculate the percentage of each aerosol species attributed to the tagged emission categories and boundary conditions. These percentages were then applied to the daily light extinction calculated from IMPROVE measurements for each PM component in the IMPROVE reconstructed light extinction equation. For example:

$$\begin{aligned}&\text{IMPROVE ammonium sulfate\_US anthropogenic}\\&= (\text{IMPROVE ammonium sulfate}) \times (\text{ammonium sulfate PSAT\_US anthropogenic}\\&/\text{ammonium sulfate CAMx total})\end{aligned} \quad (3)$$

We tracked CAMx-PSAT apportionment of U.S. anthropogenic and USB and compared these categories to the EPA estimates of natural and anthropogenic contributions. By using the same IMPROVE days as the EPA method for the 20% most impaired days and grouping the modeled source categories, we aligned the existing model outputs to the extent possible to the EPA method. The exception is that the MOZART global model used as boundary conditions for the 2011 WAQS CAMx model application did not separate boundary conditions (attributed to international emissions) into natural and anthropogenic components.

The CAMx-PSAT model results have uncertainty associated with emissions inventories, meteorology, atmospheric chemistry and transport, and in the global model simulation that is used to define boundary conditions entering the CONUS modeling domain. Emissions uncertainty is generally lowest for U.S. anthropogenic emissions and higher for U.S. natural and international emissions (Adelman et al. 2016). To test the sensitivity of CAMx simulations to boundary conditions, we compare USB estimates from three CAMx simulations with zero U.S. anthropogenic emissions and boundary conditions from three different global chemistry models: (a) MOZART as used in most of the analyses presented in this paper; (b) GEOS-Chem (Bey et al. 2001); and (c) AM3 (Donner et al. 2011). We also compare USB estimates from CAMx-PSAT in this analysis to previous model simulations.

Results

Source characterization comparing EPA method to CAMx-PSAT

Our results focus on the average aerosol contributions to light extinction for the 20% most impaired IMPROVE days in 2011 to illustrate the differences between the EPA method and the CAMx-PSAT source apportionment results. The six example Class I areas for which

aerosol light extinction is illustrated in Figure 4 are the same sites for which model performance evaluation is illustrated in Figure 3 and represent a range of geographic areas from the Pacific Northwest and California to the arid southwestern United States (Figure 2). For the 2011 most impaired IMPROVE days, of the six example sites, Olympic National Park (NP) on the western peninsula of Washington has the highest ammonium sulfate contributions to light extinction (consistent with local industry, marine shipping, and international transport), Joshua Tree NP in southern California has the highest contributions from ammonium nitrate (consistent with local transportation sources) and coarse mass (consistent with windblown dust), and Glacier NP in the Northern Rocky Mountains on the Montana border with Canada has the highest contributions from carbon (suggesting wildfire or prescribed fire contributions on the most impaired days.) The most impaired days in 2011 at Yellowstone NP in the Northern Rocky Mountains in Wyoming, Mesa Verde NP in southwestern Colorado, and Grand Canyon NP in northern Arizona are dominated by ammonium sulfate and ammonium nitrate.

In Figure 4 the sum of CAMx-PSAT U.S. anthropogenic and USB is equal to the sum of the EPA anthropogenic and natural extinction. Because natural and anthropogenic contributions to the boundary conditions were not tracked separately in the global model, a fully consistent comparison is not feasible. Future regulatory modeling will need to track separately the international anthropogenic and natural contributions from global transport models to be consistent with the EPA method. Nevertheless, insights can be gained by comparing the currently available CAMx-PSAT source categories to the EPA method. Results for individual aerosol species are discussed next.

Organic matter carbon

EPA’s method begins by defining the minimum annual (for the 15 years 2000–2014) 95% threshold for light extinction due to carbon, the sum of OMC plus EC, at each IMPROVE site to identify days with large, episodic wildfire contributions to natural haze. The EPA method further assumes that the routine natural contribution to OMC is a constant fraction of the nonepisodic OMC on any given day.

For five of the six sites in Figure 4, the EPA method estimates that more of the OMC on the average of the most impaired days is due to natural than to anthropogenic sources. For all six sites, CAMx-PSAT attributes more of the OMC on most impaired days to USB than to U.S. anthropogenic. Daily EPA estimates of episodic natural and routine natural contributions to OMC extinction are further illustrated for the period June to December 2011 in Figure 5a for Yellowstone NP and Figure 6a for Glacier NP. Daily CAMx-PSAT apportionment of OMC at Yellowstone and Glacier NPs for the same June to December 2011 period are illustrated in Figures 5b and 6b. In these graphs CAMx-PSAT apportionment of nonfire natural (biogenic), U.S. wildfire, U.S. prescribed fire, Canadian and Mexican wildfire and prescribed fire within the CONUS domain (non-U.S. fire), and boundary plus initial conditions contributions to OMC are labeled separately, while combined anthropogenic U.S., Canadian, Mexican, and marine shipping contributions within the 36-km domain are labeled as anthropogenic.

On 10 days at Yellowstone NP and 8 days (including one most impaired day on December 14, 2011) at Glacier NP, OMC extinction exceeds EPA's episodic thresholds of 10.1 and 22.2 Mm^{-1}, respectively. CAMx-PSAT consistently predicts large wildfire contributions to OMC on each of the days identified using the EPA method as episodic natural carbon. For Yellowstone NP, the daily CAMx-PSAT attribution of wildfire, prescribed fire, nonfire natural, and anthropogenic contributions to OMC agrees well with the EPA estimates of episodic natural, routine natural, and anthropogenic contributions. However, for Glacier NP, there are several days for which CAMx-PSAT predicts large fire contributions to OMC (Figure 6b), but the EPA method does not assign these days as episodic wildfire events (Figure 6a.) For Glacier NP, the EPA method assigns more of non-episodic OMC to anthropogenic contributions than to routine natural contributions. In contrast, CAMx-PSAT attributes most of the elevated OMC in September 2011 to U.S. wildfire and in October and November 2011 to U.S. prescribed fire and non-U.S. (e.g., Canadian) fires (Figure 6b.) The WRAP Fire Emissions Tracking System (2018) confirms wildfire and prescribed fire activity in Montana, Idaho, and Canada during these periods of high measured and modeled OMC (Supplement Figure 1).

The EPA and CAMx-PSAT methods are in general agreement for natural and wildfire contributions to OMC at Yellowstone NP and across a geographic range of 12 western Class I areas (see Supplement Figures 2–5). The exception is Glacier NP, where CAMx-PSAT results suggest that EPA's episodic extreme event threshold does not adequately capture the contributions of wildland fire smoke events to aerosol extinction. Moreover, for the days that the EPA method does not flag as episodic events, a large fraction of OMC is assigned as anthropogenic, while CAMx-PSAT attributes OMC to wildfires and prescribed fires. By applying a constant fraction of daily nonepisodic carbon to natural contributions, the EPA method misses the seasonal variation identified by CAMx-PSAT for natural non-fire (biogenic) emissions and wildland fire smoke in 2011 at Glacier NP (Figure 6b). The 95% episodic threshold at Glacier (22.2 Mm^{-1}) is higher than at most other western sites (U.S. EPA 2016b.) If a lower episodic threshold were used for Glacier NP, more OMC would be assigned as episodic natural, non-episodic OMC values would be lower, and the fraction of routine natural to anthropogenic OMC would be more consistent with the CAMx-PSAT apportionment. Sequoia NP is another site (Supplement Figure 2b and 2e) where a relatively high episodic threshold leads to a larger non-episodic OMC fraction and higher anthropogenic contributions to daily OMC than apportioned by CAMx-PSAT.

Elemental carbon

For four of the six sites in Figure 4, the EPA method and CAMx-PSAT both assign more of the EC on the most impaired days to anthropogenic or U.S. anthropogenic contributions, respectively, than to natural or USB contributions. However, for Glacier NP and Yellowstone NP, CAMx-PSAT attributes more of the EC to wildfires, while the EPA method assigns more of the EC to anthropogenic sources. For the same reasons as discussed in the preceding section for OMC, the EPA episodic threshold for carbon appears to underestimate fire contributions to EC at Glacier NP.

Crustal materials

For the six example sites in Figure 4, crustal material (fine soil and coarse mass) is a relatively small fraction of total aerosol extinction on the most impaired days. The EPA method appears to be effective in identifying high dust episodes. While the EPA method assigns most of the observed crustal material to natural sources, CAMx-PSAT attributes a greater fraction to U.S. anthropogenic sources (see example of daily CAMx-PSAT attribution for Mesa Verde NP in Supplement Figure 6.) This may be a result of uncertainty in windblown dust emissions and a tendency for the model to be biased low for the highest measured coarse mass and fine soil concentrations. When CAMx underestimates large natural sources of coarse mass associated with high wind events, CAMx will be biased low compared to observations and will also overestimate the anthropogenic fraction of coarse mass.

Ammonium sulfate

For all six example sites in Figure 4, and regionally across the western states (Table 2), on most impaired days the EPA method assigns the majority of the ammonium sulfate to anthropogenic contributions (Gantt et al. 2018; U.S. EPA 2016b), while CAMx-PSAT apportions more of the ammonium sulfate to international emissions (boundary conditions from the global model or combined Canadian, Mexican, and offshore shipping anthropogenic emissions within the CONUS domain) than to U.S. anthropogenic emissions. The exception in Table 2 is Theodore Roosevelt NP, where U.S. anthropogenic contributions to ammonium sulfate are greater than international. In Table 2 CAMx-PSAT assignment of U.S. anthropogenic contributions varies from a high of 10.25 Mm^{-1} at Theodore Roosevelt NP, North Dakota (consistent with local coal-fired power plants and oil and gas operations), to a low of 0.45 Mm^{-1} at Jarbridge Wilderness, Nevada, where there are few local sources.

The EPA estimates of natural contributions to ammonium sulfate are higher than the CAMx-PSAT apportionment of natural contributions within the CONUS domain. The EPA and CAMx-PSAT results are not necessarily inconsistent because boundary conditions from the MOZART global model are not separated into anthropogenic and natural sources in this analysis. For the sites in Table 2, CAMx-PSAT attributes an average of 11% of total CONUS ammonium sulfate to natural sources. If natural sources were an equivalent fraction of boundary conditions in this analysis, CAMx-PSAT attribution of ammonium sulfate would be in better agreement with the EPA estimates of anthropogenic and natural contributions.

Supplement Figures 7a–7d further illustrate that CAMx-PSAT apportions more of 2011 daily ammonium sulfate aerosol light extinction at Olympic, Glacier, Yellowstone, and Mesa Verde NPs to USB than to U.S. anthropogenic contributions. Glacier NP has the highest contributions to ammonium sulfate from wildfire and prescribed fire. Olympic NP has the highest contributions to ammonium sulfate from marine shipping and Canadian sources. Kotchenruther (2017) confirmed the contribution from marine shipping to ammonium sulfate at Olympic NP and other coastal sites using positive matrix factorization (PMF) receptor modeling.

Ammonium nitrate

For the 2011 most impaired days, the EPA method assigns most of the ammonium nitrate at Joshua Tree NP to anthropogenic sources, more of the ammonium nitrate at Glacier NP to natural sources, and more evenly divides the ammonium nitrate between natural and anthropogenic contributions at the other sites in Figure 4. CAMx-PSAT apportions most of the ammonium nitrate on most impaired days at Joshua Tree, Grand Canyon, Mesa Verde, and Yellowstone NPs to U.S. anthropogenic sources and very little to USB. The EPA estimates for anthropogenic ammonium nitrate are lower than the CAMx-PSAT apportionment to U.S. anthropogenic, and the differences between the two methods would be greater if evaluated using combined CAMx-PSAT U.S. plus international anthropogenic ammonium nitrate. The EPA estimated natural contributions to ammonium nitrate are higher than the CAMx-PSAT apportionment of ammonium nitrate to USB. The EPA method may overestimate the natural contribution to ammonium nitrate, particularly on winter days when biogenic emissions are low, by assigning the same daily fraction of measured ammonium nitrate to natural sources.

Influence of global model boundary conditions

USB estimates using CAMx with different global model inputs—The preceding results indicate that boundary conditions are important contributors to ammonium sulfate but not to ammonium nitrate. Because these results have important implications for source characterization at western Class I areas, we further evaluate the sensitivity of CAMx simulations to boundary conditions using CAMx simulations with zero U.S. anthropogenic emissions and boundary conditions from three global chemistry models: MOZART, GEOS-Chem, and AM3. (Note that the global model simulations include U.S. anthropogenic emissions, and boundary conditions may include some contributions from U.S. anthropogenic emissions through flow reversals or recirculation.) Model simulations using zero U.S. anthropogenic emissions can be used to estimate USB contributions and can also be compared to USB estimates from the CAMx-PSAT simulation. Results of the two approaches may differ because CAMx-PSAT simulates current emissions and atmospheric chemistry, while CAMx simulations with zero U.S. anthropogenic emissions alter atmospheric chemistry compared to current conditions.

Modeled USB for three CAMx simulations using each of the global models and zero U.S. anthropogenic emissions and the EPA natural assignments for aerosol species light extinction for 2011 most impaired days are reported in Table 3 for Mesa Verde NP, an interior site with comparatively low contributions from boundary conditions. For USB ammonium sulfate light extinction, the MOZART and GEOS-Chem global model estimates at Mesa Verde NP are similar (2.97 and 3.06 Mm^{-1}, respectively), but higher than those of AM3 (1.74 Mm^{-1}) and much higher than EPA's estimate of natural ammonium sulfate (0.96 Mm^{-1}.) The large variability in the USB ammonium sulfate estimates among the three global model simulations points to uncertainty in international and marine shipping emission inventories and the importance of additional global model performance evaluation for aerosol ammonium sulfate.

For USB ammonium nitrate on the 2011 most impaired days at Mesa Verde NP, MOZART estimates (0.08 Mm^{-1}) are lower than those of GEOS-Chem (0.16 Mm^{-1}) and AM3 (0.32 Mm^{-1}) and also lower than the EPA natural estimate of ammonium nitrate (0.89 Mm^{-1}). In this case, the model USB estimates are substantially lower than the EPA natural ammonium nitrate estimates, and excluding international anthropogenic ammonium nitrate would result in still larger differences between the EPA natural estimates and model estimates of natural ammonium nitrate. The EPA method may overestimate the natural contribution to ammonium nitrate, particularly on winter days, by assigning the same daily fraction of measured ammonium nitrate to natural sources. The variations among the global models again highlight the importance of model performance evaluation for the global model that is used as boundary conditions for future photochemical modeling.

For OMC and EC, the EPA natural and modeled USB using the three global model inputs have similar contributions (OMC, 2.11–2.47 Mm^{-1}, and EC, 0.26–0.34 Mm^{-1}) to aerosol light extinction. Higher fractions of coarse mass and fine soil are assigned as natural using the EPA method than the modeled USB using the three global models for CAMx boundary conditions.

USB estimates from CAMx-PSAT and CAMx simulation with zero U.S. anthropogenic emissions—We compare consistency between USB estimates at Mesa Verde NP using CAMx-PSAT source apportionment and a CAMx simulation using zero U.S. anthropogenic emissions. Both 2011 CAMx simulations use boundary conditions from the MOZART global model. At Mesa Verde NP the highest daily USB concentrations of ammonium sulfate are very similar between the CAMx simulation with zero U.S. anthropogenic emissions and the CAMx-PSAT simulation (Supplement Figure 8.)

In contrast, Figure 7 illustrates that for Mesa Verde NP on the days with the highest USB ammonium nitrate mass in the CAMx simulation with zero U.S. anthropogenic emissions, CAMx-PSAT has lower estimates of USB ammonium nitrate. There are several factors leading to different results of these two approaches for ammonium nitrate, but not ammonium sulfate. CAMx-PSAT apportions source contributions under 2011 emissions and atmospheric chemistry, while CAMx sensitivities using zero U.S. anthropogenic emissions significantly change atmospheric chemistry and aerosol species concentrations. Since sulfate is a stronger acid than nitrate, ammonium will preferentially neutralize sulfate over reacting with nitric acid (HNO_3) to form ammonium nitrate aerosol. In the CAMx-PSAT simulation less USB ammonium nitrate will be formed due to the presence of the U.S. anthropogenic SO_2 emissions that are not present in the CAMx zero-out sensitivity simulation. Another reason USB ammonium nitrate will be lower in the CAMx-PSAT than zero-out simulations is that CAMx and PSAT use the ISORROPIA aerosol thermodynamics module (Nenes et al. 1998a, 1998b; Fountoukis and Nenes 2007) that treats ammonium nitrate as being in instantaneous equilibrium with gaseous HNO_3 and NH_3. When transported USB ammonium nitrate interacts with HNO_3 produced from U.S. emissions of NO_x, the transported nitrate tracer is partially replaced with domestic nitrate tracer. CAMx-PSAT assigns most of the modeled ammonium nitrate to U.S. anthropogenic sources rather than USB. The CAMx simulation with zero U.S. anthropogenic emissions shows greater mass assigned to USB because there is no U.S. anthropogenic sulfate or HNO_3 to interact with the transported

ammonium nitrate. To the extent that ammonium nitrate is globally transported, the CAMx-PSAT results will estimate lower international transport of ammonium nitrate than a zero-out method. The implication is that USB contributions to ammonium nitrate are likely bounded by the two methods.

Comparison 2011 CAMx-PSAT USB to USB estimates from previous modeling —Park et al. (2004) reported for 2001 GEOS-Chem global modeling that approximately 28% of the 2011 spatial average ammonium sulfate mass in the western United States (1.52 $\mu g/m^3$) was due to USB (0.43 $\mu g/m^3$), with 72% (1.09 $\mu g/m^3$) due to U.S. anthropogenic contributions. Of the USB contributions, 0.11, 0.13, and 0.15 $\mu g/m^3$ were attributed to natural, Asian, and combined Canadian and Mexico emissions, respectively. In comparison, CAMx predicts 2011 modeled annual average ammonium sulfate mass as 0.59 to 1.25 $\mu g/m^3$ across the six example sites (Table 4), lower than the 2001 GEOS-Chem average. CAMx-PSAT attributes 17–31% of 2011 modeled annual average ammonium sulfate mass to U.S. anthropogenic contributions and 69–83% to USB. The differences between the two modeled estimates are a function of changes in U.S. and international emissions between 2001 and 2011 and differences in the model configurations.

For ammonium nitrate, the 2001 GEOS-Chem modeling (Park et al. 2004) estimated that U.S. anthropogenic emissions contributed 81% (1.24 $\mu g/m^3$) of the average ammonium nitrate (1.54 $\mu g/m^3$) in the western U.S. with USB contributing 18% (0.27 $\mu g/m^3$). Natural contributions were estimated as 2% of total ammonium nitrate (0.03 $\mu g/m^3$.) Table 4 reports that CAMx modeled 2011 annual average ammonium nitrate mass for the six example sites varies from 0.02 to 1.03 $\mu g/m^3$, much lower than the 2001 average GEOS-Chem results. CAMx-PSAT estimates that U.S. anthropogenic contributions vary from 50% at Glacier NP, to 65% at Olympic NP, and up to 93% at Mesa Verde and Joshua Tree NPs.

Van Donkelaar et al. (2008) used satellite, aircraft, and ground-based measurements and the GEOS-Chem model to interpret contributions from eastern Asia to sulfate aerosol over western British Columbia during the Intercontinental Chemical Transport Experiment Phase B (INTEX-B) campaign in April and May 2006. They attributed 0.3 $\mu g/m^3$ sulfate to transport from eastern Asia. In comparison, for April–May 2011 in this study, CAMx-PSAT apportioned an average of 0.29 $\mu g/m^3$ sulfate (0.40 $\mu g/m^3$ ammonium sulfate) at Olympic National Park on the northwestern U.S. coast to boundary conditions (trans-Pacific transport) and another 0.06 $\mu g/m^3$ sulfate (0.08 $\mu g/m^3$ ammonium sulfate) to Canadian anthropogenic emissions. Thus, the 2011 CAMx-PSAT estimate of trans-Pacific contribution to sulfate on the Pacific Coast is similar the 2006 estimates in Van Donkelaar et al. (2008).

Discussion

EPA method for the revised visibility tracking metric

The EPA method is effective in removing the influence of the highest episodes of carbon and dust (attributed to wildfires and dust storms) on the days that are tracked for visibility progress. The method is an improvement over the haziest days metric used to demonstrate progress in the first RHR implementation period (2000–2018) because it focuses on days when visibility is more likely to be responsive to reductions in anthropogenic emissions.

Uncertainties remain in the estimates of episodic natural, routine natural, and anthropogenic contributions to daily aerosol extinction and the extent that natural and uncontrollable international emissions contribute to total haze on the most impaired days. These uncertainties can influence which days are tracked over time as most impaired days and the uniform rate of progress (URP) glidepath 2064 endpoint that is based on average natural conditions on most impaired days from 2000 to 2014.

CAMx-PSAT source apportionment approach

This analysis demonstrates that CAMx-PSAT is an effective tool to evaluate the EPA method. Because regional photochemical grid models will be used to project the visibility benefits of states' emission reductions and modeled visibility improvements will be compared to the URP calculated based on EPA's assignment of most impaired days, it is important to understand the extent of agreement between these two distinct approaches.

Model accuracy

The CAMx photochemical model and CAMx-PSAT source apportionment tool provide very precise quantification of U.S. anthropogenic impairment and specific source contributions to aerosol extinction, but due to uncertainties in emissions and atmospheric transport, the model may not be accurate. Even when using the model in a relative sense to apportion the IMPROVE observations, if the model errs in assigning species concentrations to specific source types, the relative source apportionment for aerosol species could also be incorrect. Thus, the performance of the model is of paramount importance. In the 2011 Western Air Quality Study, CAMx-PSAT simulations were run at 36 km grid resolution. In future regulatory modeling for the western United States, CAMx-PSAT performance may be improved by running simulations at 12 km resolution to improve representation of topographic features and tracking of emissions transport.

This 2011 CAMx-PSAT model analysis predicts that international emissions, including Canadian, Mexican, marine shipping, and Asian transport, are major sources of ammonium sulfate at western Class I areas. Kotchenruther (2017) confirmed contributions from marine shipping to ammonium sulfate at Class I areas on the Pacific coast and demonstrated reductions in measured ammonium sulfate in response to emissions reductions from marine shipping required under the North American Emissions Control Area treaty. Future CAMx-PSAT modeling also should be able to identify reductions in marine shipping contributions to ammonium sulfate at Pacific coastal sites. Future global modeling will need to update international SO_2 emissions, particularly from China, to reflect the observed decreases in SO_2 emissions since 2010 (e.g. Li et al. 2017; van der A et al. 2017). Updated Canadian and Mexican emissions are also needed. Accuracy of modeled international transport should be evaluated for aerosol species using satellite and surface measurements.

To be consistent with EPA's proposed tracking metric, future regulatory modeling will need to separately track anthropogenic and natural contributions to international transport. Currently, model simulations that zero out anthropogenic emissions are used to define natural boundary conditions. The CAMx-PSAT algorithm currently does not track separately

natural and anthropogenic contributions to boundary conditions. This is an area of future model development for CAMx-PSAT.

Comparison of EPA method and CAMx-PSAT

Carbon—The EPA method and the CAMx-PSAT apportionment generally are in good agreement in assigning elevated episodes of OMC and EC to fire events for a range of western Class I areas. At Glacier NP, CAMx-PSAT attributed more of the non-episodic OMC to natural contributions and less to anthropogenic than did the EPA method. For sites where episodic contributions of fire are underestimated, assigning a constant fraction of daily non-episodic OMC to routine natural contributions underestimates natural contributions and overestimates anthropogenic contributions. This can lead to days with large natural or fire OMC contributions being selected as most impaired days. In future applications, CAMx-PSAT could be used to evaluate whether alternative episodic thresholds or allowing the fraction of non-episodic carbon to vary across the year would better account for contributions from wildland fire smoke and biogenic emissions, at least for the years for which model results are available. Alternatively, allocation of non-episodic carbon might be improved by further incorporating the hybrid receptor model approach demonstrated by Schichtel et al. (2017). Chemical tracers (e.g., levoglucosan), fire activity and emissions data (e.g., WRAP Fire Emissions Tracking System, http://wrapfets.org/), or satellite detection (e.g., National Oceanic and Atmospheric Administration's Hazard Mapping System, http://www.ospo.noaa.gov/Products/land/hms.html) could be used by individual states to refine site specific estimated source contributions, but are probably too labor intensive for regional application. Fire will continue to be an important contributor to haze in Class I areas, and efforts to improve fire inventories and activity data will improve model performance for OMC and EC.

Crustal materials—EPA's method to operationally define source contributions to fine soil and coarse mass is likely accurate within the confidence of existing emission inventories. CAMx-PSAT identified natural and wildland fire contributions to fine soil and coarse mass, but assigned more of the fine soil and coarse mass to U.S. anthropogenic and less to natural sources than the EPA method. CAMx tended to be biased low, likely due to large uncertainty in estimates of natural windblown dust emissions, and this would cause CAMx to overestimate the anthropogenic fraction of coarse mass. Improvements are needed in future windblown dust inventories, and CAMx model runs should focus on model performance for known dust events.

Ammonium sulfate—The EPA method assigned most of the ammonium sulfate to anthropogenic contributions, while CAMx-PSAT apportioned more of the ammonium sulfate to international emissions than to U.S. anthropogenic emissions. These results are not inconsistent because most of the international emissions are also anthropogenic. Future regulatory modeling will need to separately track the international transport contributions to anthropogenic and natural sources to be consistent with EPA's definition of anthropogenic impairment. Natural contributions to ammonium sulfate can be estimated in model simulations with zero anthropogenic emissions, and international anthropogenic contributions to sulfate can be estimated in model simulations with zero U.S. anthropogenic

emissions. While there is uncertainty in model estimates of natural and anthropogenic SO_2 emissions, satellite measurements of SO_2 may help to improve model performance for ammonium sulfate.

Ammonium nitrate—The EPA method assigned a greater fraction of ammonium nitrate to natural contributions than did the CAMx-PSAT results, which attributed most of daily ammonium nitrate to U.S. anthropogenic emissions. Also, the EPA method of assigning a constant fraction of ammonium nitrate to routine natural sources without considering seasonal constraints to biogenic processes may overestimate levels of natural ammonium nitrate on winter days. The CAMx sensitivity simulation with zero U.S. anthropogenic emissions attributed more ammonium nitrate to international emissions than did CAMx-PSAT. The latter result is due to the difference in a zero-out sensitivity method and a PSAT source apportionment method and is likely a function of the presence of 2011 U.S. anthropogenic emissions in the PSAT source apportionment run, including the sulfate that displaces the ammonium from the ammonium nitrate and the assumption of instantaneous equilibrium between the gas and aerosol phases. These differences could be reduced by using model simulations with zero U.S. anthropogenic emissions to estimate USB ammonium nitrate, or by adding an option to CAMx PSAT so that transported nitrate is not replaced by local nitrate. We conclude that it may be appropriate to apply both approaches to estimate the U.S. contribution, rather than using one approach or the other.

Implications for the uniform rate of progress glidepath

The Regional Haze Rule requires states to adopt a long term strategy of emissions reductions and to estimate the reasonable progress goal (RPG) at each Class I area in 2028, the end of the next planning period. States must also compare the RPG to the URP glidepath in 2028. If photochemical model simulations that are used to estimate the RPG in 2028 include large contributions from uncontrollable emission sources on the most impaired days, and if the URP glidepath is not adjusted to account for these uncontrollable sources, the modeled visibility progress (RPG) from reducing U.S. anthropogenic emissions may fail to achieve the URP. This was observed for the haziest days glidepath in the first round of regional haze plans for some western Class I areas in which the haziest days included large uncontrollable emissions from wildfires, windblown dust, or international transport. EPA's proposed visibility tracking metric is intended to exclude days dominated by wildfires and dust storms from the URP glidepath, but comparison of modeled RPGs to the most impaired days URP glidepath could still be problematic if there are differences in the EPA estimates and the modeled estimates of uncontrollable sources on the most impaired days.

The 2016 guidance allows the URP glidepath to be adjusted to account for international contributions to impairment if these contributions can be quantified. Source apportionment modeling can be used to estimate the contributions from international anthropogenic sources on the most impaired days in the base year and future-year model simulations to evaluate how these international contributions affect the URP glidepath for those years. However, because the CAMx-PSAT results presented here do not distinguish between the natural and anthropogenic contributions to international transport, they cannot be used to adjust the URP to account for international impairment. Nor can modeled natural haze be directly compared

to the EPA estimates of natural haze. Both the EPA and CAMx-PSAT estimates of natural haze and impairment contain uncertainties. As a result, it may not be possible to conclude which approach is more accurate. The key implication is that substantial differences between the EPA and CAMx-PSAT methods can result in differences in the rate of the visibility improvement that can be achieved in the photochemical model compared to the URP that is based on EPA's assumptions for natural contributions.

We have demonstrated that U.S. anthropogenic contributions to IMPROVE aerosol measurements can be estimated using model source apportionment techniques, although additional evaluation is needed for ammonium nitrate source attribution to better understand the differences in source contributions using source apportionment and zero-out sensitivity methods, such as examining total nitrate (nitrate plus nitric acid) and total ammonia (ammonia plus ammonium) contributions. We conclude that additional evaluation of international emissions inventories and coupled global and regional chemical transport models are needed before applying source apportionment models to adjust the URP glidepath to account for international contributions.

Because CAMx-PSAT simulates a smaller contribution of U.S. anthropogenic emissions to visibility impairment compared to the method used to calculate the URP, the rate of progress projected by CAMx for a future year may not show as much progress as the URP. As an alternative, the most impaired days could be defined using CAMx-PSAT to rank days by U.S. anthropogenic contribution. Another option would be to use the same USB emissions in the base and future year CAMx-PSAT modeling to define the rate of visibility progress specifically for changes in U.S. anthropogenic emissions only.

The IMPROVE monitoring data demonstrate that visibility has improved over much of the western U.S. over the past two decades (WRAP Technical Support System, http://views.cira.colostate.edu/tssv2). As U.S. anthropogenic emissions continue to decrease and visibility approaches natural conditions, the most impaired days tracked in RHR planning will include larger relative contributions from uncontrollable emissions (e.g., natural and non-U.S. anthropogenic sources). Refinements to assumptions for natural conditions, URP, and uncontrollable sources will continue to be important to state implementation planning.

Summary

EPA designed the revised regional haze tracking method to focus on days that are most likely to be responsive to changes in anthropogenic emissions. Using the 2011 Western Air Quality Study CAMx-PSAT simulation to evaluate the EPA estimates of natural and anthropogenic contributions, we conclude:

- For most western sites, CAMx-PSAT identified that carbon from wildfire or prescribed fire impacted the IMPROVE monitors on days that the EPA method assigned to episodic extreme fire events.
- Coarse mass and fine soil are not major contributions to light extinction on most impaired days and the EPA method appears to be effective in identifying dust episodes. CAMx-PSAT tended to underestimate coarse mass from natural dust

events, so comparison to EPA's estimates of natural episodic dust events is inconclusive.

- Ammonium sulfate and ammonium nitrate dominate on most impaired days at western Class I areas. The EPA method estimates that anthropogenic contributions to these aerosol species are greater than natural contributions on most impaired days.
- CAMx-PSAT attributes more of the ammonium sulfate at western Class I areas to international emissions than to U.S. anthropogenic emissions.
- CAMx-PSAT attributes more of the ammonium nitrate at western Class I areas to U.S. anthropogenic emissions than international, while the EPA methods attributes more of the ammonium nitrate to natural emissions.
- Variability among global model simulations indicate that improved international inventories and additional global model performance evaluation will be important for the next regional haze modeling efforts.
- Differences between the EPA method and the CAMx-PSAT estimates of natural and anthropogenic contributions to haze could lead to different assumptions for visibility improvement by 2028.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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About the authors

Patricia Brewer is a regulatory policy specialist with the Air Resource Division of the National Park Service in Denver, CO.

Gail Tonnesen is an environmental engineer at EPA Region 8 in Denver, CO.

Ralph Morris is a Principal at Ramboll in Novato, CA.

Tom Moore is the Air Quality Program Manager for the Western Regional Air Partnership (WRAP) in Fort Collins, CO, and works for the Western States Air Resources Council (WESTAR).

Uarporn Nopmongcol is a managing consultant at Ramboll, in Novato, CA.

Debra Miller is an air quality specialist with the Intermountain Region of the National Park Service in Denver, CO.

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Implications:

The western states intend to use the CAMx model to project visibility progress by 2028. Modeled visibility response to changes in U.S. anthropogenic emissions may be less than estimated using the EPA assumptions based on total U.S. and international anthropogenic contributions to visibility impairment. Additional model improvements are needed to better account for contributions to haze from natural and international emissions in current and future modeling years. These improvements will allow more direct comparison of model and EPA estimates of natural and anthropogenic contributions to haze and future visibility progress.

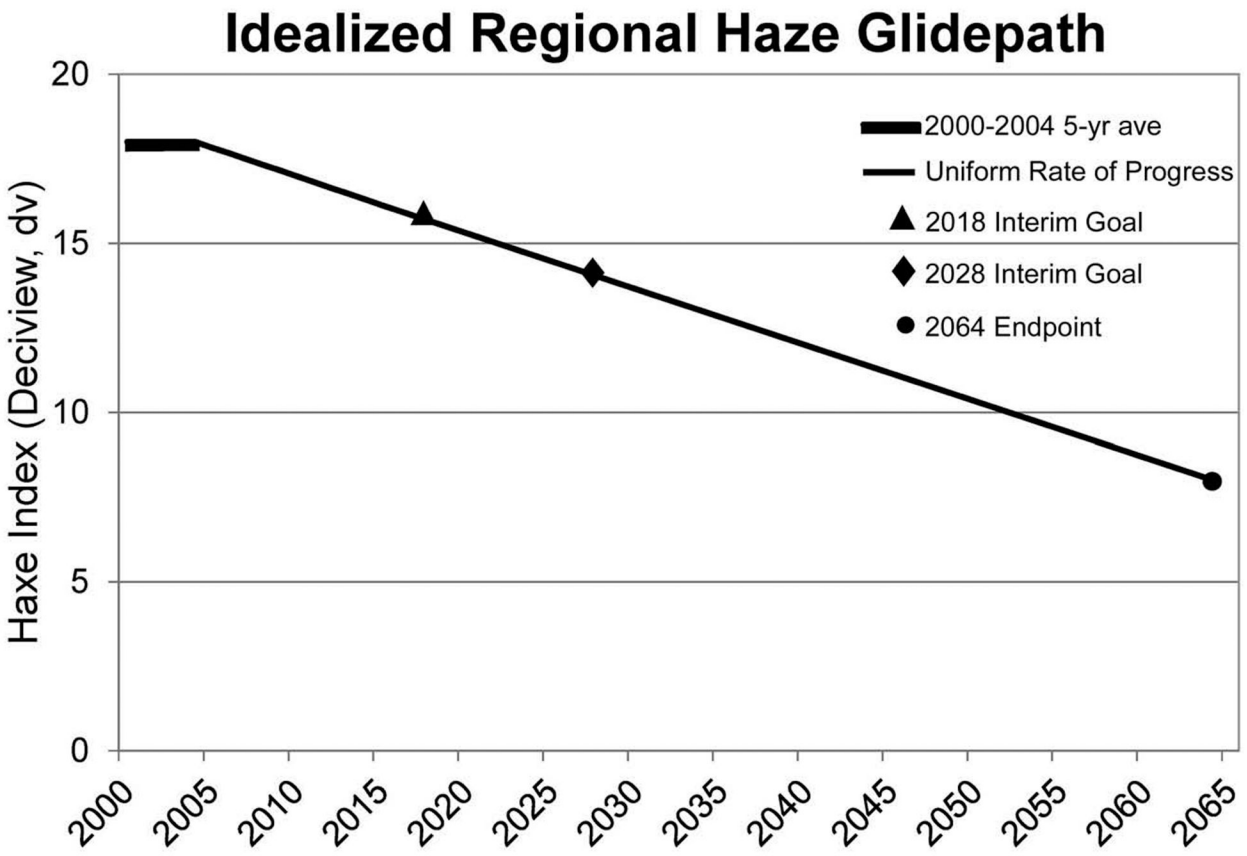

Figure 1.
Idealized regional haze rule glidepath.

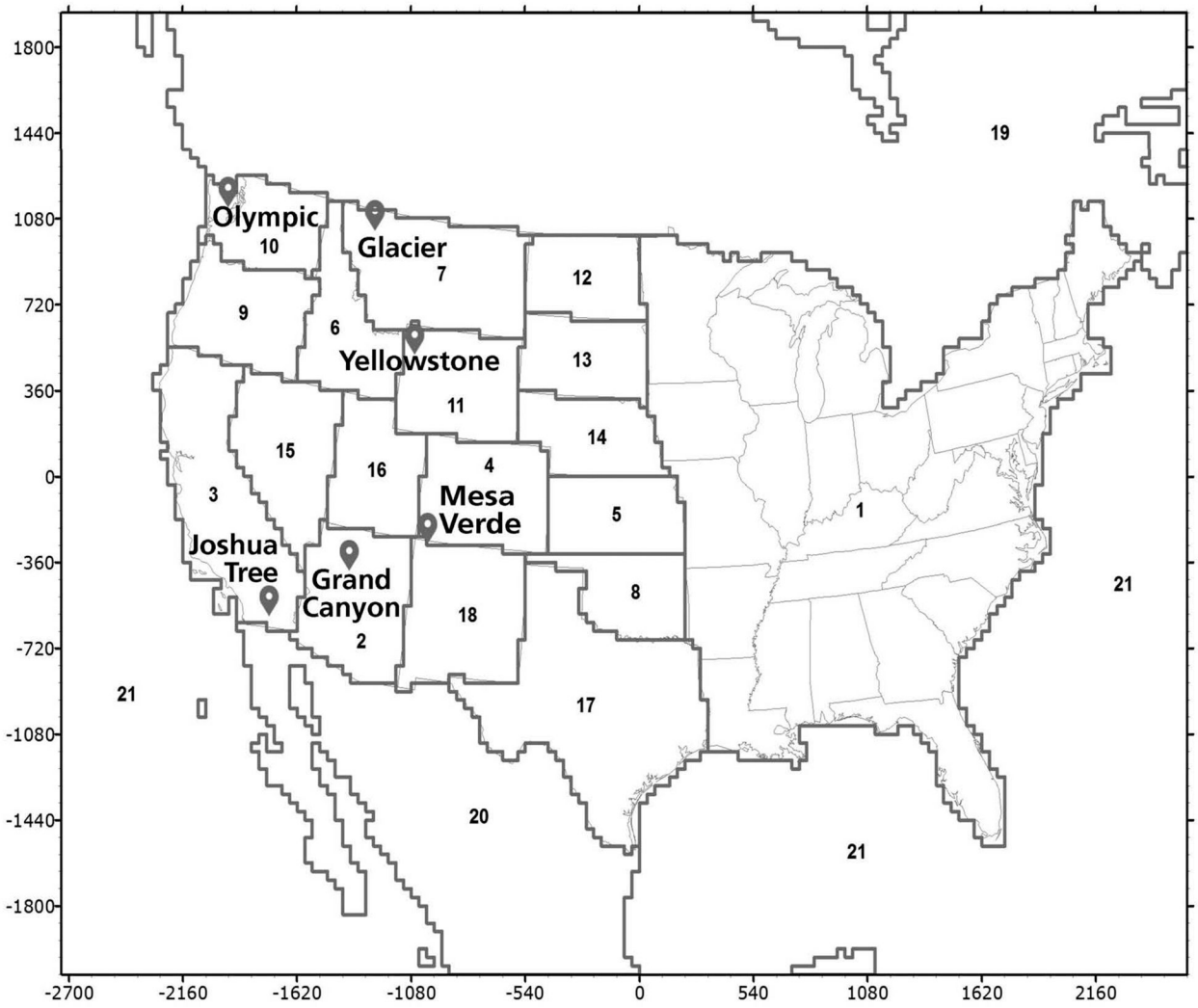

Figure 2.

Western Air Quality Study CAMx version 6.10 2011b Continental U.S. 36-km modeling domain and source regions defined for the Particle Source Apportionment Tool (PSAT) for visibility.

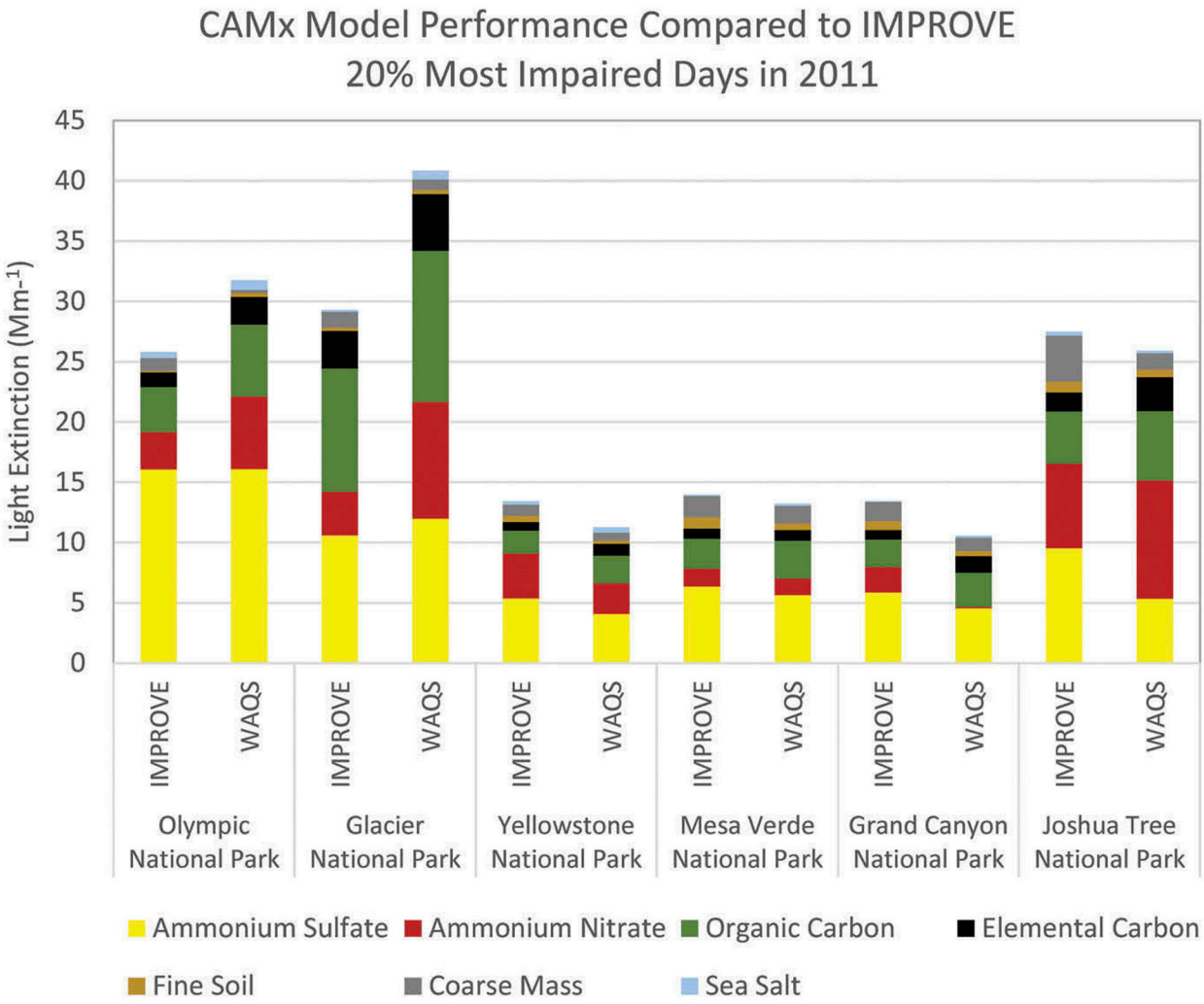

Figure 3.

CAMx model performance in the 2011 Western Air Quality Study (36 km domain) on the 20% most impaired days in 2011 (selected using EPA method) compared to IMPROVE measurements for Olympic, Glacier, Yellowstone, Mesa Verde, Grand Canyon, and Joshua Tree National Parks.

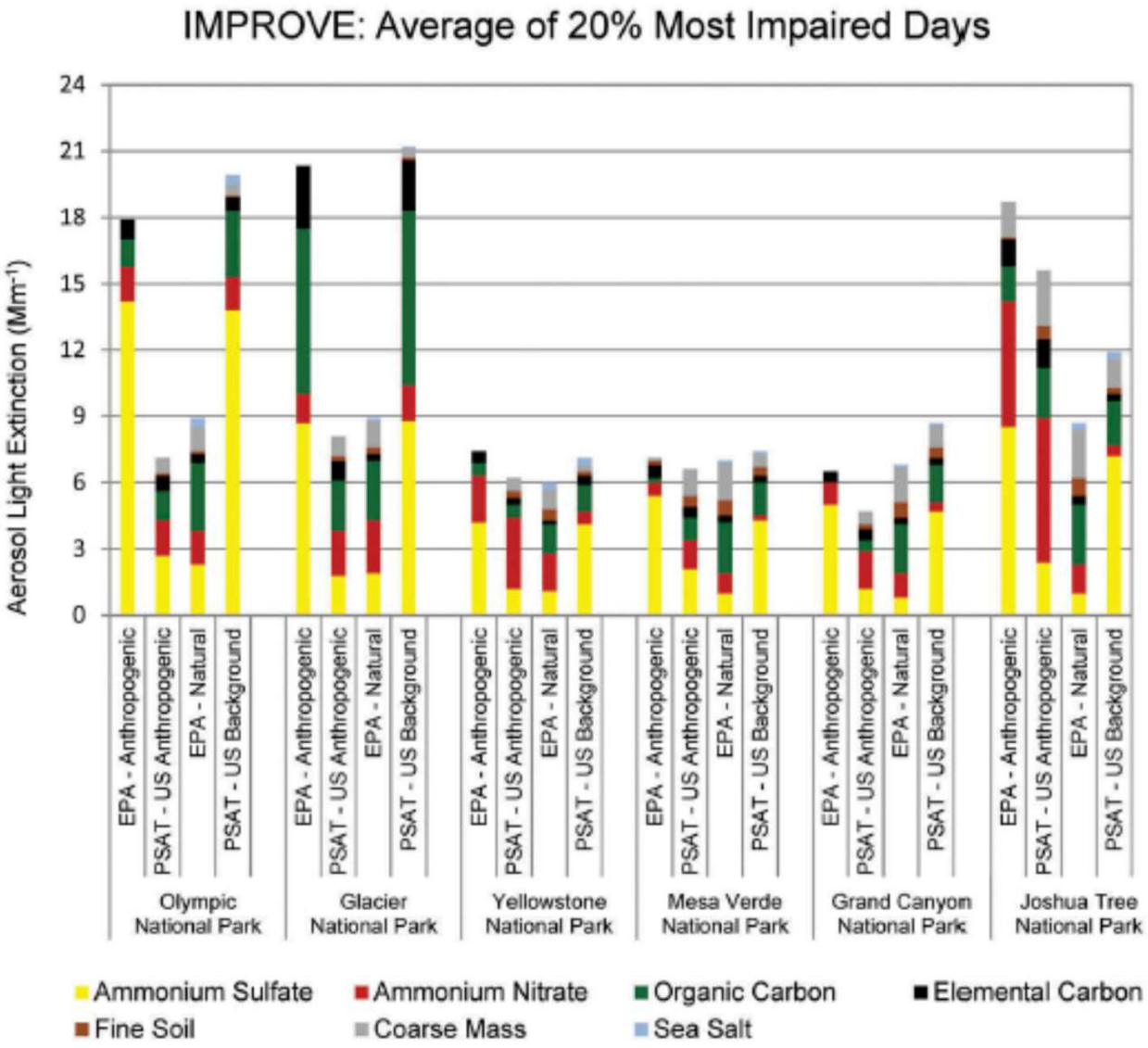

Figure 4.

2011 IMPROVE aerosol light extinction (Mm^{-1}) for average of 20% most impaired days: comparison of EPA anthropogenic and EPA natural contributions to PSAT U.S. anthropogenic and PSAT U.S. background (U.S. natural plus international natural and anthropogenic) contributions for Olympic, Glacier, Yellowstone, Mesa Verde, Grand Canyon, and Joshua Tree National Parks.

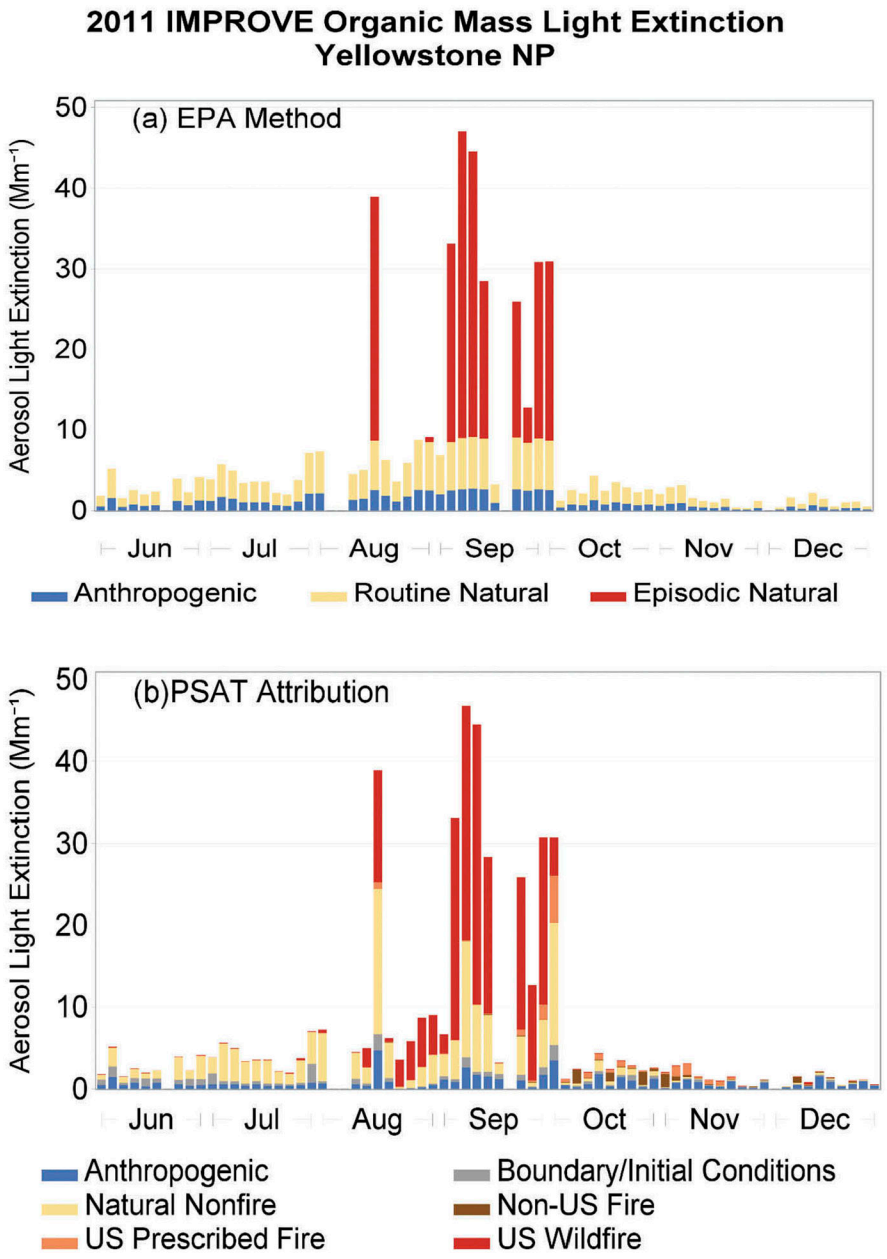

Figure 5.

For Yellowstone National Park, IMPROVE organic mass light extinction (Mm^{-1}) for August–December 2011, illustrating (a) the EPA method's assignment to episodic natural, routine natural, and anthropogenic contributions and (b) CAMx-PSAT source apportionment.

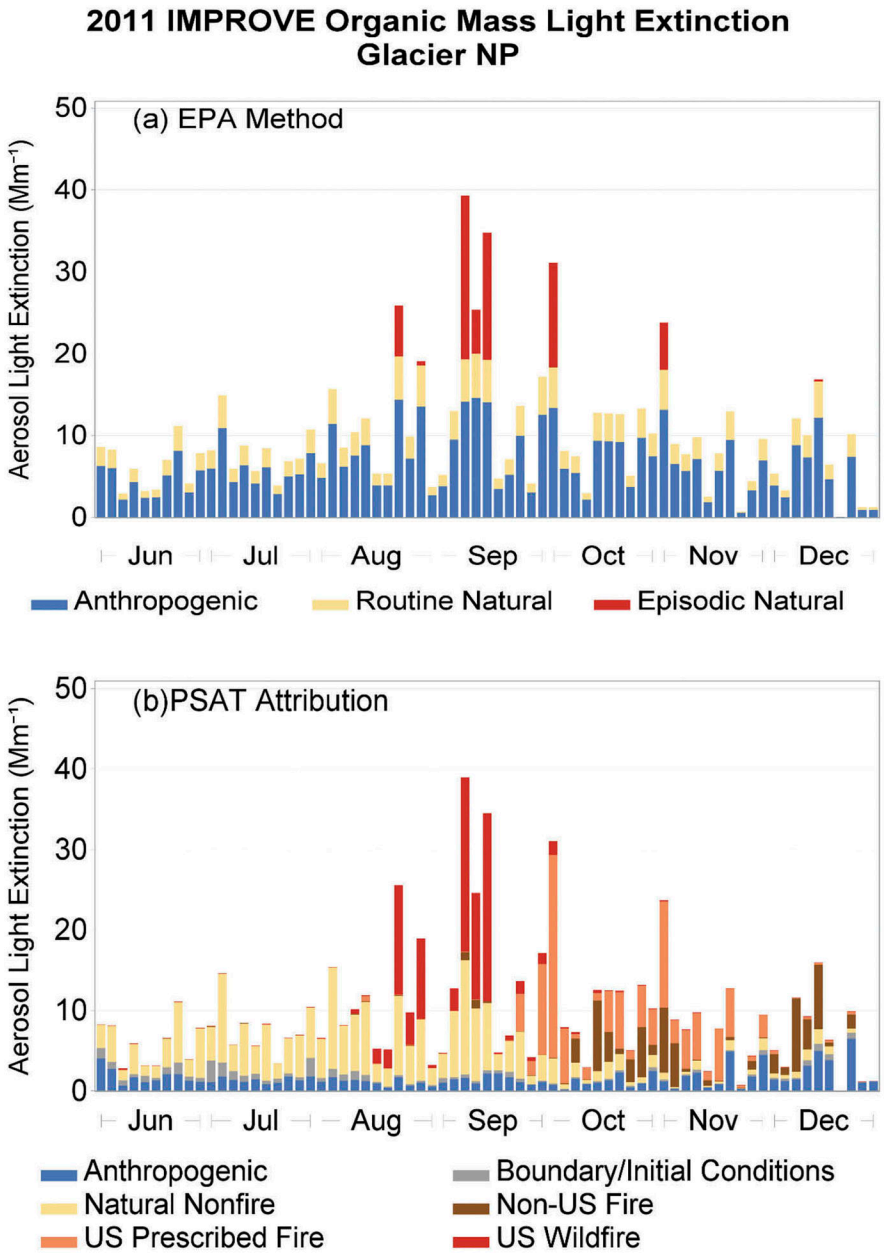

Figure 6.

For Glacier National Park, IMPROVE organic mass light extinction (Mm^{-1}) for August–December 2011, illustrating (a) the EPA method's assignment to episodic natural, routine natural, and anthropogenic contributions and (b) CAMx-PSAT source apportionment.

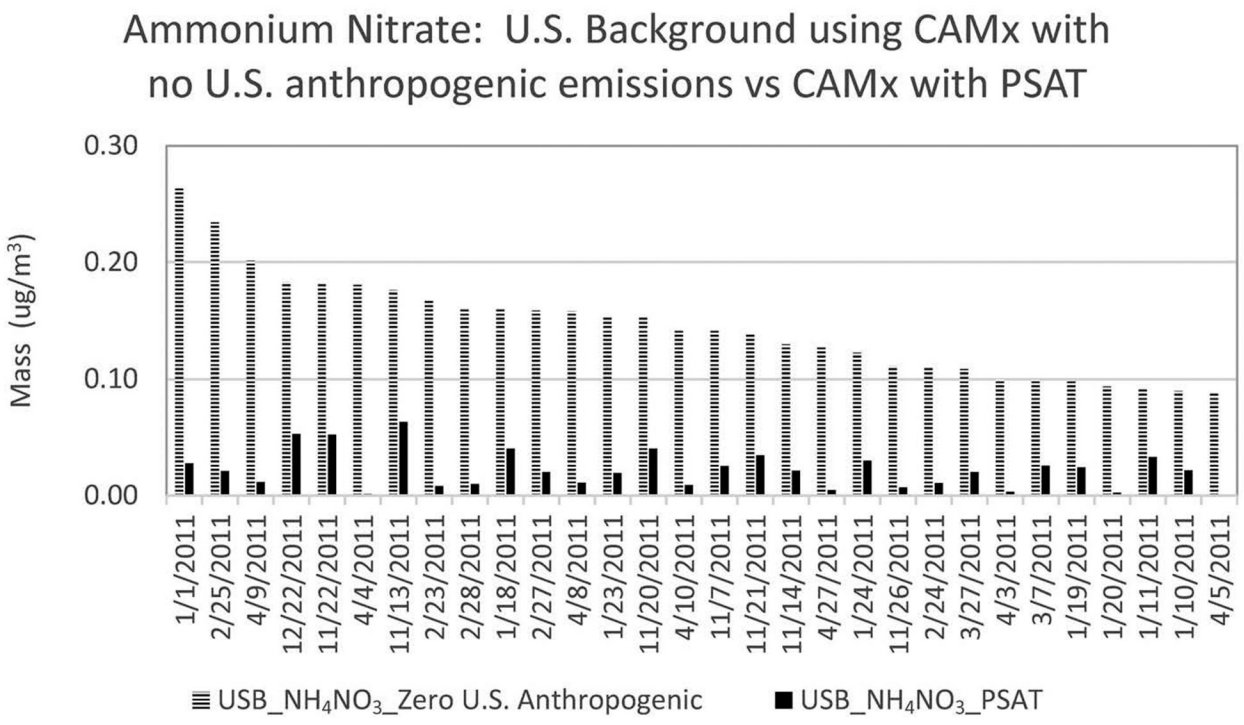

Figure 7.

2011 Ammonium nitrate mass (μg/m^3) at Mesa Verde National Park on days with highest U.S. background (USB) defined by CAMx simulation with no U.S. anthropogenic emissions compared to results of CAMx PSAT apportionment.

Table 1.

Terms and definitions related to the visibility tracking metric used in regional haze planning.

Term	Definition
Impairment or anthropogenic impairment	As defined in U.S. EPA 2017: “any humanly perceptible difference due to air pollution from anthropogenic sources between actual visibility and natural visibility on one or more days. Because natural visibility can only be estimated or inferred, visibility impairment also is estimated or inferred rather than directly measured.”
Most impaired days	The 20% of IMPROVE monitored days in a calendar year with the highest fraction of total haze attributed to anthropogenic sources.
U.S. anthropogenic	U.S. emissions defined in national, state, and local inventories as being emitted by anthropogenic activities
Natural haze	Fraction of total haze attributed to naturally occurring phenomena
Episodic natural	Aerosol light extinction representing episodic extreme natural events of carbon (organic matter carbon plus elemental carbon) or dust (coarse mass plus fine soil) and calculated as the minimum annual 95th percentile value in the period 2000 to 2014
Routine natural	Aerosol light extinction representing for each aerosol species the fraction of daily non-episodic aerosol light extinction assigned to natural emissions. The fraction is calculated as the average Natural Conditions II aerosol extinction divided by the total annual average non-episodic aerosol extinction.
U.S. background (USB)	Combined U.S. natural emissions and international natural plus anthropogenic emissions
Natural conditions	As defined in U.S. EPA 2017: “*Natural conditions* reflect naturally occurring phenomena that reduce visibility as measured in terms of light extinction, visual range, contrast, or coloration, and may refer to the conditions on a single day or a set of days. These phenomena include, but are not limited to, humidity, fire events, dust storms, volcanic activity, and biogenic emissions from soils and trees. These phenomena may be near or far from a Class 1 area and may be outside the United States.”

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Table 2.

2011 IMPROVE ammonium sulfate light extinction (Mm^{-1}) on most-impaired days, EPA estimates of anthropogenic and natural contributions, and CAMx-PSAT apportionment to U.S. anthropogenic, Canadian, Mexican, and shipping anthropogenic, boundary conditions, and North American natural contributions.

	2011 IMPROVE most impaired days: ammonium sulfate aerosol light extinction (Mm^{-1})						
		EPA method		PSAT apportionment			
Class I Areas	**IMPROVE**	**Anthropogenic**	**Natural**	**U.S. anthropogenic**	**Canadian, Mexican, shipping anthropogenic**	**Boundary conditions**	**North American Natural (biogenic + fire)**
Canyonlands NP, UT	5.45	4.61	0.84	1.99	0.45	2.88	0.13
Glacier NP, MT	10.59	8.72	1.87	1.78	2.94	4.74	1.13
Grand Canyon NP, AZ	5.88	5.04	0.84	1.17	1.16	3.36	0.19
Guadalupe Mountains NP, TX *	13.95	12.73	1.21	4.85	5.44	3.50	0.15
Jarbridge Wilderness, NV	5.63	4.54	1.10	0.45	0.33	4.57	0.31
Joshua Tree NP, CA	9.55	8.55	1.00	2.39	1.78	4.83	0.55
Olympic NP, WA	16.43	14.18	2.26	2.68	5.11	7.63	1.01
Mesa Verde NP, CO	6.37	5.40	0.96	2.08	1.06	3.11	0.11
Rocky Mountain NP, CO	6.76	5.65	1.10	2.60	0.64	3.37	0.15
Sequoia NP, CO	14.51	13.64	0.88	1.81	1.00	11.23	0.47
Theodore Roosevelt NP, ND	17.80	16.13	1.67	10.25	4.14	3.30	0.11
Yellowstone NP, WY	5.36	4.23	1.13	1.22	0.24	3.70	0.20

* The IMPROVE monitor at Guadalupe Mountains NP, TX, also represents Carlsbad Caverns NP, NM.

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Table 3.

Aerosol light extinction (Mm^{-1}) for average of 20% most impaired days in 2011 at Mesa Verde National Park: Comparison of EPA natural extinction to modeled U.S. background (U. S. natural plus international, USB) extinction using three global models, MOZART, GEOS-Chem, and AM3, for boundary conditions and zero U.S. emissions for the CAMx continental U.S. modeling domain.

Scenario	$AmmSO_4$	$AmmNO_3$	Organic carbon mass	Elemental carbon	Fine soil	Coarse mass	Sea salt	Total[b]
USB MOZART [a]	2.97	0.08	2.11	0.34	0.10	0.29	0.39	6.29
USB GEOS-Chem [a]	3.06	0.16	2.31	0.26	0.48	0.78	0.43	7.48
USB AM3 [a]	1.74	0.32	2.47	0.28	0.19	0.39	0.35	5.73
EPA natural	0.96	0.89	2.27	0.28	0.75	1.71	0.10	6.97

[a]Zero out U.S. anthropogenic in CAMx simulation.

[b]Excluding Rayleigh light scattering.

Table 4.

2011 CAMx-PSAT apportionment of U.S. anthropogenic and U.S. background (natural plus international) contributions to ammonium sulfate and ammonium nitrate concentrations.

	CAMX PSAT 2011 annual average concentration, μg/m^3					
Class I national parks	**Ammonium sulfate total**	**Ammonium sulfate: U.S. anthropogenic**	**Ammonium sulfate: U.S. background**	**Ammonium nitrate total**	**Ammonium nitrate: U.S. anthropogenic**	**Ammonium nitrate: U.S. background**
Olympic	1.254	0.226	1.028	1.027	0.668	0.360
Glacier	0.780	0.133	0.648	0.468	0.234	0.234
Yellowstone	0.584	0.128	0.456	0.232	0.179	0.053
Mesa Verde	0.789	0.245	0.545	0.128	0.119	0.009
Grand Canyon	0.749	0.165	0.585	0.024	0.016	0.008
Joshua Tree	0.873	0.175	0.698	0.657	0.611	0.046