Atmospheric Environment 45 (2011) 7341–7351

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Investigation into approaches to reduce excessive vertical transport over complex terrain in a regional photochemical grid model

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ARTICLE INFO

Article history:
Received 24 May 2011
Received in revised form
27 July 2011
Accepted 28 July 2011

Keywords:
CAMx
CMAQ
Ozone transport
Vertical advection

ABSTRACT

Past photochemical modeling of the western United States (US) using the Comprehensive Air quality Model with extensions (CAMx) and the Community Multiscale Air Quality (CMAQ) model has resulted in large springtime ozone over predictions in the complex high-elevation terrain of the western United States (US). Comparisons against rural measurement data have shown that both models over predicted ozone levels by 20 ppb or more. A systematic investigation using CAMx revealed that excessive vertical transport in mountainous terrain draws down upper tropospheric ozone introduced by the lateral boundary conditions (developed by a global chemistry model), which can routinely exceed 1 ppm near the tropopause. This is not an unreasonable concentration at such altitudes during the spring, and there is observational evidence that stratospheric ozone intrusions result in occasional large ground-level concentrations in the western US, but not at the frequency and intensity simulated by the CAMx and CMAQ models. Past versions of CAMx and CMAQ possess similar algorithms to diagnose vertical velocity, and similar first-order accurate vertical advection algorithms. These similarities, in conjunction with poor vertical resolution of the upper troposphere, have resulted in similar ozone performance issues. Numerous approaches were explored with CAMx in an attempt to externally reduce the rates of vertical transport over complex terrain. Ultimately, we formulated and tested a new vertical advection methodology that included improvements to how vertical velocities are determined and introduced a second-order accurate advection solver technique. Together these improvements proved to yield the most successful results in reducing upper tropospheric ozone transport to the surface. CAMx was then run to simulate ozone throughout the western US for a full year to evaluate the effects of the new vertical transport algorithm on model performance. Ozone performance improvements exceeded those achieved through the application of arbitrary reductions in the upper tropospheric/stratospheric lateral boundary conditions.

1. Introduction

Past regional photochemical modeling conducted with the Comprehensive Air-quality Model with extensions (CAMx; ENVIRON, 2008) and the Community Multiscale Air Quality model (CMAQ; Byun and Schere, 2006) has resulted in springtime ozone over predictions in the complex terrain of the western United States (US). Although these two models were run with different horizontal resolution and for different years (2005 and 2002, respectively) using diverse meteorology and emission inputs, they both generated the highest ozone concentrations over the high terrain of the Rocky Mountains in the spring, most notably April and May (Stoeckenius et al., 2009). Comparisons with rural observations showed that both models over predicted ozone levels in the western Colorado area by 20 ppb or more during these months. The ability to properly simulate the usually pristine environment of the western US is critically important given that ambient ozone levels, especially in the spring, are near background levels.

CAMx tests conducted by Stoeckenius et al. (2009) showed that the highest ground level ozone concentrations were caused by the vertical transport of upper tropospheric and stratospheric ozone carried into the domain from the lateral boundary conditions (BC). The CAMx and CMAQ simulations described above used identical 19-layer vertical grid structures, with boundary conditions derived from the same 2002 annual run of the GEOS-Chem global chemistry model (Bey et al., 2001). The CMAQ application was run for the year 2002, and employed BC extracted from 3-hourly GEOS-Chem concentration fields over that year. The CAMx application was run

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doi:10.1016/j.atmosenv.2011.07.052

for the year 2005, with BC extracted from 2002 monthly-averaged GEOS-Chem concentration fields. Fig. 1 shows the variation over the entirety of 2002 of the maximum ozone BC in the top four model layers: layer 16 (2500–3400 m), 17 (3400–5200 m), 18 (5100–8100 m) and 19 (8100–14,600 m). The maximum ozone BC occurred in the spring and exceeded 200 ppb, which is reasonable at these altitudes in the spring when the tropopause is low. Surface observations during the spring suggest that stratospheric ozone intrusion events can result in high ground-level ozone concentrations at both low and high-elevation monitoring sites in the western US (Lefohn et al., 2001). However, such high surface ozone values do not occur at the frequency and intensity as predicted by the models.

As "off-line" models, both CAMx and CMAQ are run separately from the numerical weather prediction models used to generate meteorological input fields. To maximize efficiency, off-line chemistry models are configured with coarser vertical resolution than the meteorological models, and ingest instantaneous meteorological fields once per hour. Furthermore, the chemistry models employ vastly different transport algorithms and solvers. All of these deviations contribute to mass transport inconsistencies in the off-line models (Byun and Ching, 1999). Furthermore, numerical artifacts from the meteorological model can be transferred to the air quality models and possibly even magnified. Coupled or "on-line" meteorology-chemical transport models such as WRF-Chem (Grell et al., 2005) may not have this problem, depending how the advection of tracers is integrated with the dynamics solver.

Both CAMx and CMAQ internally conserve mass and minimize the sources of mass inconsistency as they integrate forward in time. Much of this is accomplished by internally diagnosing vertical velocity profiles at each time step that balance the local atmospheric continuity equation for the specific grid configuration employed;

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \rho V,$$

where the tendency of atmospheric density ρ is known according to the input meteorological fields. In CAMx, the vertical velocity profile with height $w(z)$ is determined from vertical integration of the divergent continuity equation from the surface to the top of the model,

$$\rho w(z) = -\int_0^z \left(\frac{\partial \rho}{\partial t} + \nabla_H \cdot \rho V_H \right) \mathrm{d}z, \quad (1)$$

which assures a balance between the imposed density tendency and the resolved horizontal momentum divergence (as denoted by the subscript H) in each grid cell at each time step. When there is convergence in a particular vertical layer, a positive (upward) vertical velocity increment is applied; the opposite occurs with divergence. This vertical velocity is used for subsequent vertical transport calculations for all pollutants. A zero vertical velocity is specified at the ground, while a pollutant-specific zero-gradient mixing ratio condition is applied at the top of the model. To further guarantee mass consistency, the horizontal momentum divergence is determined by using the CAMx horizontal advection solver to calculate the atmospheric density flux. In simple tests in which a uniform pollutant field of unity mixing ratio is transported throughout a regional grid over several days using actual episodic meteorological inputs, this approach has been shown to provide nearly exact (to 10^{-3} to 10^{-4}%) consistency between the density and pollutant fields.

Analyses of vertical velocity profiles in the CAMx modeling suggested that the low- to mid-tropospheric vertical velocities were well-behaved but problems were associated with "noisy" vertical velocities in the uppermost and thickest layers (e.g., layer 19 is an aggregate of five meteorological model layers). Over the highest terrain, significant vertical transport in only a few upper layers was needed to bring stratospheric ozone down to the ground:

- The bottom of layer 19 is approximately 8 km above sea level (MSL), which is about the minimum altitude of the wintertime tropopause (the top of the model is at 15 km);
- The top of the highest terrain is approximately 3 km MSL, requiring only 5 km of vertical transport from the bottom of layer 19 to reach the ground;
- With deep afternoon mixing of 2–3 km above the highest terrain, vertical transport from the bottom of layer 19 would only need to extend downward by 2–3 km before mixing would efficiently transport ozone the remaining distance to the surface.

Whereas the CAMx springtime ozone over predictions reported by Stoeckenius et al. (2009) were mitigated by arbitrarily reducing the highest ozone BC in layer 19, this solution merely masked the underlying problem associated with excessive vertical transport. This paper documents an investigation with CAMx that examined several potential solutions to correct or mitigate vertical transport over complex terrain, and describes modeling results using the final adopted improvements. The US Environmental Protection Agency

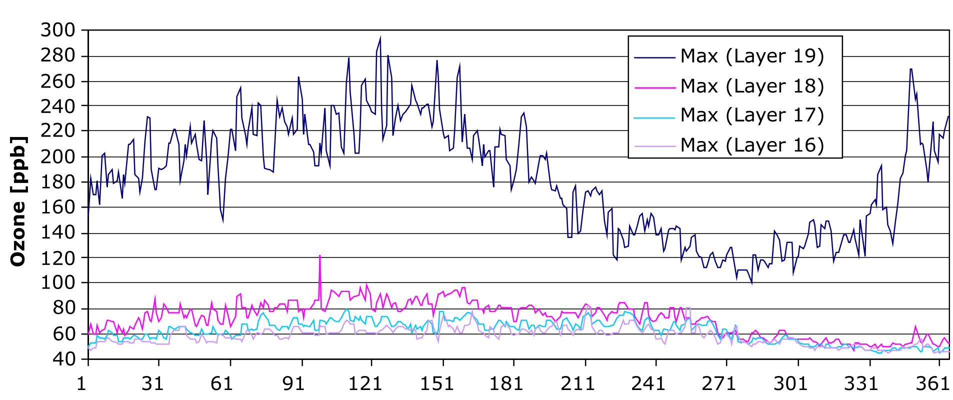

Fig. 1. Time series throughout 2002 of absolute maximum ozone anywhere within layers 16 through 19 of the CMAQ/CAMx lateral BC extracted from GEOS-Chem.

(EPA) has independently incorporated similar improvements for CMAQ (Young et al., 2009).

1.1. Approach

An initial set of experiments was designed to analyze the characteristics of vertical velocity profiles over the Intermountain West, and to evaluate, implement, and test various techniques external to CAMx that would reduce excessive vertical transport within the model. These tests included smoothing, divergence minimization, and mass flux filtering; although these techniques were not particularly effective, they ultimately revealed the need to improve the top boundary condition treatment within CAMx. An additional series of tests were run in which the vertical layer structure was expanded from 19 to 22 and 34 layers to better resolve the upper troposphere (34 layers exactly matched each of the meteorological model's layers). With just this change, large and widespread surface ozone reductions were found without any modifications to the input wind fields. CAMx lateral BC were subsequently updated using 3-hourly output from a new 2005 GEOS-Chem simulation, replacing the 2002 monthly averages. Finally, we developed and tested alternative vertical velocity and advection algorithms in CAMx, which proved to yield the most successful results in reducing ozone transport to the surface.

All tests described above were performed with CAMx version 4.51 (ENVIRON, 2008) over the month of April 2005 on a modeling grid covering the western US with 12 km grid spacing. The 19-layer vertical grid structure is shown in Table 1 for a grid column spanning a pressure altitude of 100 to 1000 hPa. The CAMx terrain-following height coordinate (z) is determined from the normalized pressure (or σ_p) coordinate used in the meteorological model, and is thus spatially variable as defined by the underlying resolved terrain. All CAMx test simulations were run for one inert tracer (ozone): precursor emissions, chemistry, and deposition were excluded so that the resulting ozone concentration fields were strictly the result of transport of initial and boundary conditions throughout the three-dimensional grid. A final best configuration of the model was used to run a full photochemical simulation of the entire 2005 calendar year. Monthly ozone performance against rural measurement data was then compared to the original results reported by Stoeckenius et al. (2009).

Table 1
Three CAMx vertical layer structures employed in the CAMx tests described in the text.

19 layers	22 layers	34 layers	Press (hPa)	Height (m)	Depth (m)
19	22	34	100	15,676	2004
		33	145	13,672	1585
	21	32	190	12,087	1322
		31	235	10,765	1139
	20	30	280	9626	1004
18		29	325	8622	900
	19	28	370	7721	817
		27	415	6904	750
	18	26	460	6154	693
17		25	505	5461	645
	17	24	550	4816	604
		23	595	4213	568
16	16	22	640	3645	536
		21	685	3109	508
15	15	20	730	2601	388
		19	766	2212	282
14	14	18	793	1931	274
		17	820	1657	178
13	13	16	838	1478	175
		15	856	1303	172
12	12	14	874	1131	169
11	11	13	892	961	167
10	10	12	910	795	82
		11	919	712	82
9	9	10	928	631	81
		9	937	550	80
8	8	8	946	469	80
7	7	7	955	390	79
6	6	6	964	310	79
5	5	5	973	232	78
4	4	4	982	154	39
3	3	3	986.5	115	39
2	2	2	991	77	38
1	1	1	995.5	38	38
0	0	0	1000	0	38

2. Inert model tests

2.1. Smoother–desmoother

A series of inert CAMx simulations over April 2005 was first conducted to identify an external processor (i.e., filter or smoother) that when applied to the input winds and/or density fields would lead to reductions in the intensity of deep vertical motions and circulations over elevated complex terrain. In the first case we applied a smoother–desmoother filter (Yang and Chen, 2008) to the CAMx wind and density fields using various smoothing factors and number of sweeps. Tests included applying the algorithm to various combinations of CAMx layers, smoothing just input horizontal wind fields, and smoothing both wind and density fields (pressure and temperature). The smoother–desmoother filter helped reduce the magnitude of vertical velocity in upper layers with relatively little impact on horizontal wind fields. However, even with the most aggressive application of the smoother–desmoother technique, it could not adequately reduce the vertical transport of upper-layer ozone to the surface. The largest reduction to the April-maximum surface ozone concentration was much less than 10 ppb, or less than 10% of the peak simulated surface ozone.

2.2. Divergence minimization

In the second case we adapted the divergence minimization scheme used in the CALMET diagnostic meteorological model (Scire et al., 2000) to the grid structure and variable staggering used in CAMx. CAMx sensitivity tests were run with various strengths of divergence minimization, different number of iterations, and various combinations of layers. Whereas divergence minimization led to rather large reductions in surface ozone, the procedure generated spurious and troublesome numerical artifacts at the top of the model, including strong gradients and directional reversals (horizontally and vertically) in the vertical velocity fields. These numerical artifacts, in combination with the simplistic time- and space-invariant CAMx top boundary condition treatment, resulted in a strong artificial dilution of ozone in the topmost layer. Further experimentation revealed that reductions in surface ozone were entirely caused by the dilution artifact at the top of the model, and not by modifications to the vertical velocity profiles induced by divergence minimization.

2.3. Improved top boundary conditions

Historically, CAMx top boundary conditions have been externally specified as a set of temporally and spatially invariant concentrations that are assumed to be representative of average conditions above the model top. This approach becomes increasingly problematic as the spatial and temporal modeling domains grow, and is entirely too simplistic for high concentration constituents such as ozone that rapidly evolve at stratospheric altitudes. To address these conceptual issues and the numerical problems described in Section 2.2, the CAMx top BC treatment was replaced by a "zero-gradient" technique. In this approach, the concentration of each chemical species above the top of each vertical grid column is internally set as the vertical advection step is solved, where the species mass mixing ratio above the top-most grid cell is set to the value in that cell.

The zero-gradient top BC treatment was tested in an inert CAMx run, and it resulted in an increase in the simulated ground-level ozone concentrations in complex terrain by roughly 10 ppb due to the removal of artificial dilution at the top of the model. This signal was consistent regardless of whether divergence minimization was applied or not. Since the zero-gradient top boundary condition is a more technically defensible approach, this modification was used for the remainder of all CAMx tests reported herein.

2.4. Improved vertical resolution

Additional tests were conducted in which the number of vertical layers was increased from 19 to all 34 layers available from the meteorological model (i.e., no layer aggregation or collapsing). No other external modifications were applied to the wind or density inputs. Improved vertical resolution significantly reduced surface ozone concentrations relative to the 19 layer run (Fig. 2, top and middle), but at the price of doubling computational requirements. An alternative 22 layer model configuration, which added more vertical resolution in the upper portion of the domain, produced comparable results to the full 34 layer model (Fig. 2, bottom) with only a modest (~10%) increase in computer time over the 19 layer model configuration. A comparison of the three different layer structures is provided in Table 1.

Daily peak ozone in the inert April 2005 simulation was reduced by more than 10 ppb over wide areas of the Intermountain West by simply increasing the number of layers. Likely reasons for this result include: (1) improved consistency with the meteorological model in representing the transport field; (2) reduced numerical error in diagnosed vertical velocities associated with higher resolution wind and density profiles; and (3) reduced impact from numerical diffusion generated by the vertical advection solver.

A new 2005 GEOS-Chem simulation was subsequently used to generate lateral BC for the April 2005 test database. The 2005 3-hourly ozone BC in the upper CAMx layers were significantly higher than the original monthly-averaged 2002 BC, reaching well over 1 ppm during the springtime of 2005, and resulting in enormous surface ozone concentrations in the inert 19 layer run (Fig. 3, top). Furthermore, the introduction of 2005-specific ozone BC resulted in substantial shifts in the spatial pattern of CAMx surface ozone concentrations, clearly demonstrating how inter-annual variability invalidates the assumption that monthly or seasonally averaged global background distributions from a particular year are sufficiently representative of other years. Although the 22 and 34 layer structures continued to reduce surface concentrations significantly in these tests (Fig. 3, middle and bottom), the over prediction problem was further exacerbated using the new 2005 BC. Obviously, additional solutions were necessary.

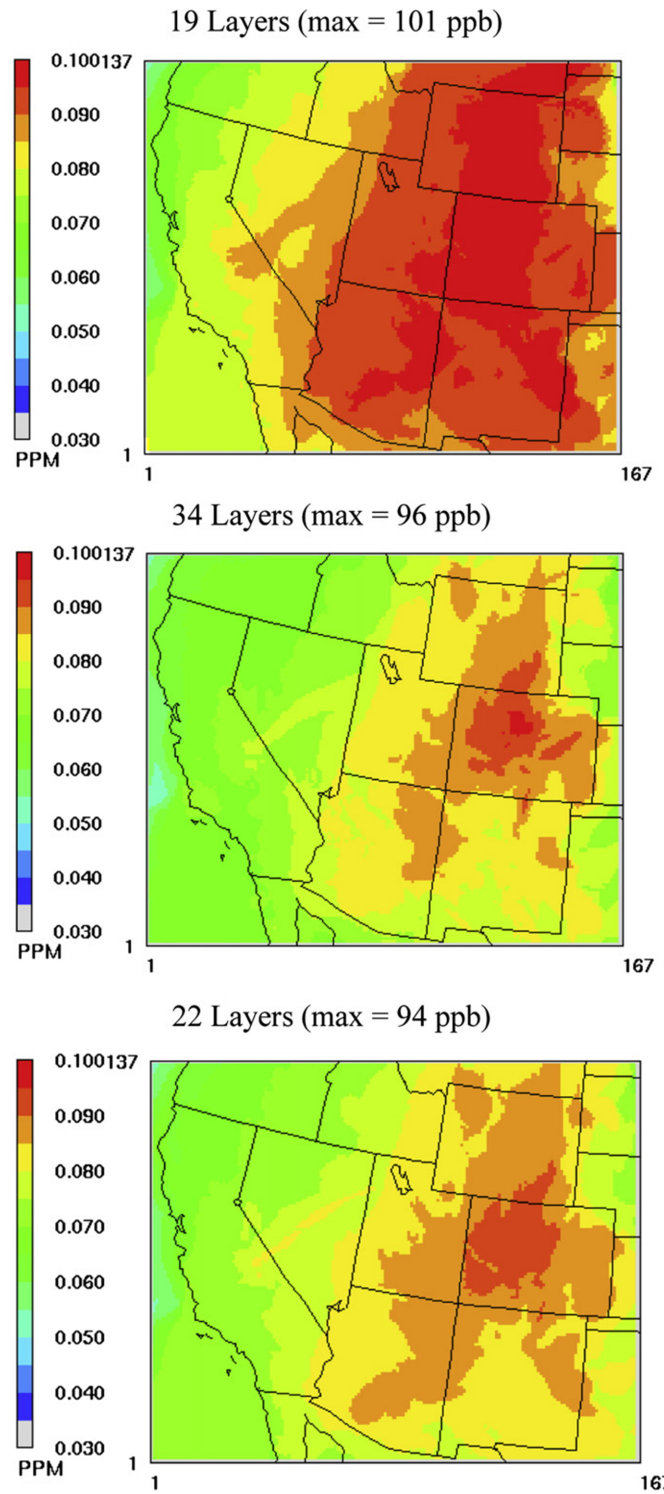

Fig. 2. April 2005 CAMx inert maximum ozone concentrations (ppm) using "zero-gradient" top BC and original 19 layer vertical structure (top), full 34 layer vertical structure (middle) and intermediate 22 layer vertical structure (bottom).

2.5. Mass filter

Subsequent to the updates adopted for the lateral and top BC, we identified a more promising approach to "filter" horizontal wind components according to mass/density perturbations arising from horizontal density flux divergence. Rotman et al. (2004) have developed a mass filter for the Lawrence Livermore National Laboratory IMPACT global chemistry model. Similarly to GEOS-Chem, IMPACT is run using global observation-based meteorological analyses. The global wind and density fields are not dynamically self-consistent and thus can include discrepancies among the

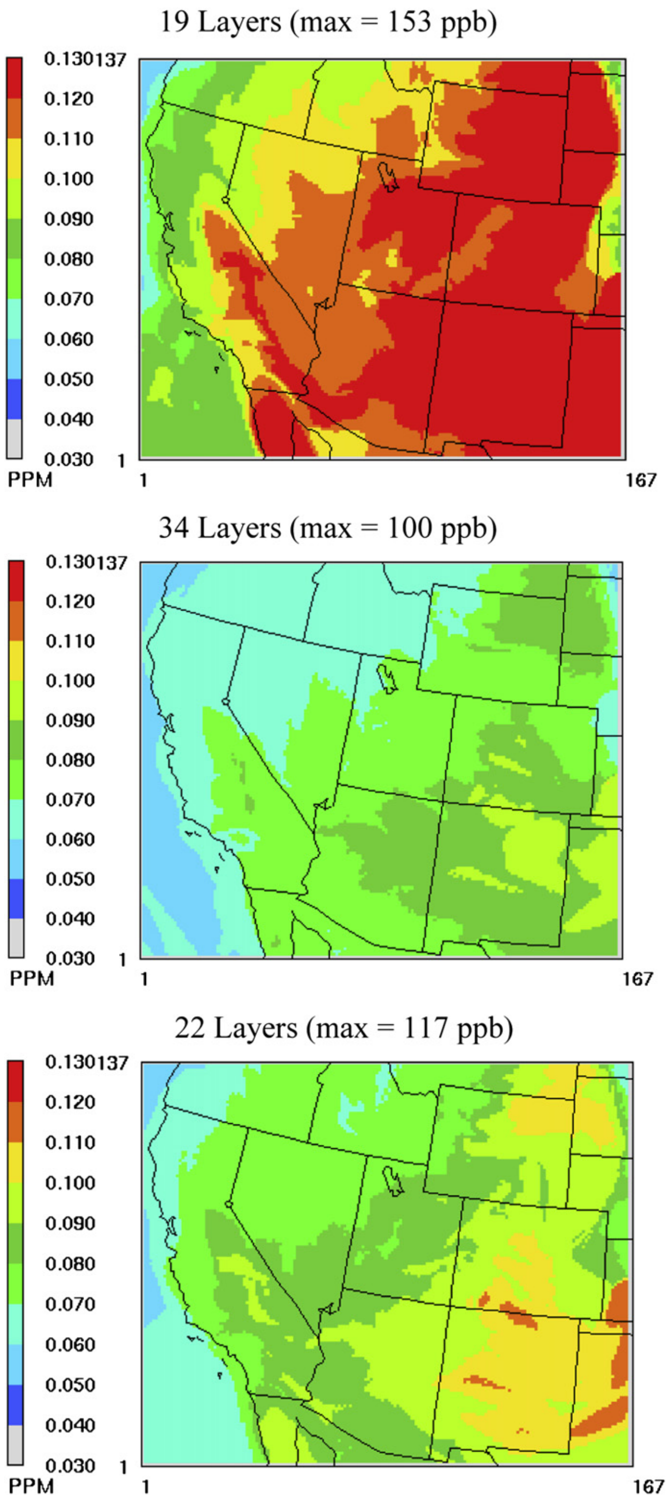

Fig. 3. April 2005 CAMx inert maximum ozone concentrations (ppm) using "zero-gradient" top BC, 2005 lateral BC, and original 19 layer vertical structure (top), full 34 layer vertical structure (middle) and intermediate 22 layer vertical structure (bottom).

meteorological variables that can produce spurious results, including excessive vertical velocities. The mass filter helps to eliminate these dynamic inconsistencies in the IMPACT model, so we expected that its application on the CAMx input meteorological fields could help reduce excessive vertical transport in these tests as well. This approach was more in line with the methodology used in CAMx to calculate vertical velocity, and did not include the inefficiencies and numerical problems inherent in the divergence minimization technique.

The mass filter was adapted to the grid structure and variable staggering used in CAMx, and applied to the input wind and density fields using varying levels of adjustments. CAMx was then run with the mass-filtered input fields using the 2005 lateral BC, zero-gradient top BC, and 19-layer configuration. Application of the most aggressive configuration of the mass filter reduced the April maximum surface ozone concentrations by 10–20 ppb (Fig. 4). However, this reduction was not sufficient to reduce the excessive surface ozone to anything close to observed concentrations in these inert tests.

2.6. Revised vertical transport algorithm

Given limited success in reducing excessive vertical transport in these inert CAMx tests by externally altering horizontal wind and density fields, we undertook a critical evaluation of the numerical approach employed within the CAMx vertical transport algorithm. Several issues were identified.

First, the vertical advection solver in this version of CAMx was a backward-Euler (in time) upstream-donor (in space) scheme. Backward-Euler time differencing is fully implicit, and coupled to the upstream space differencing, the scheme was absolutely stable. This allowed CAMx to solve vertical advection by inverting a tri-diagonal matrix once per time step without the need to subdivide

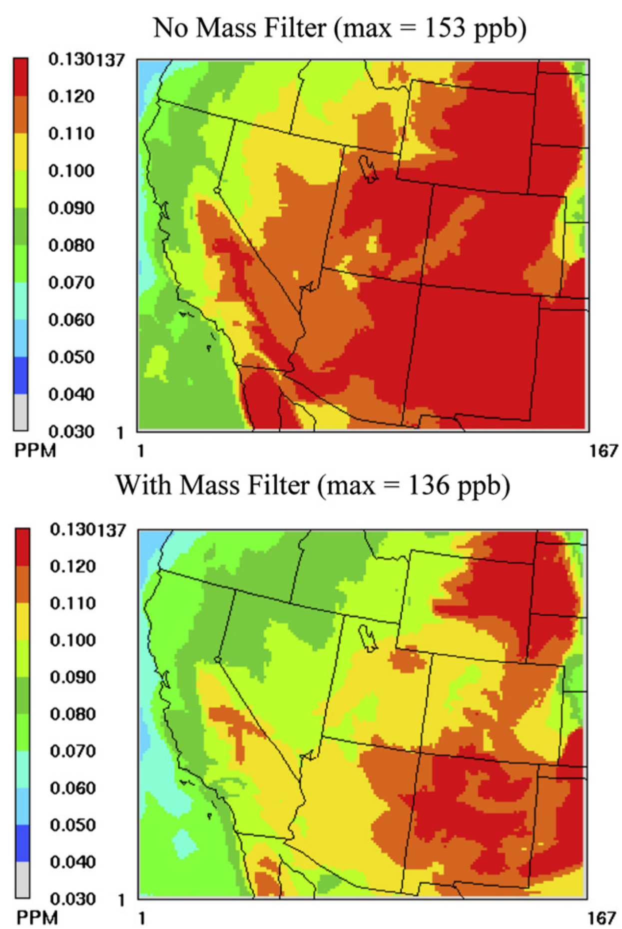

Fig. 4. April 2005 CAMx inert maximum ozone concentrations (ppm) using "zero-gradient" top BC, 2005 lateral BC, and original 19 layer vertical structure; original meteorology (top) and after application of the mass filter (bottom).

the step to maintain a stable solution (this was an explicit design consideration to minimize run time). However, the upstream-donor space differencing is only first-order accurate and is known to possess a high degree of "numerical diffusion", a process that artificially transfers mass through the grid purely as a numerical artifact of the solution technique. Numerical diffusion was indeed found to comprise a large fraction of the total vertical transport solution in CAMx. Furthermore, its contribution increased with fewer, coarser layers. This fact partially explains why increasing the number of layers to resolve the same spatial depth dramatically improved surface ozone results.

Second, the internal diagnostic vertical velocity algorithm was found to be asymmetric by over estimating downward motion and under estimating upward motion. Ideally, two adjacent grid columns with identical horizontal mass flux divergence profiles, but of opposite sign, would be expected to yield the same vertical velocity profiles, but of opposite sign. However, the finite difference calculation of vertical velocity at layer interface height k was developed to maximize consistency with the upstream-donor vertical advection algorithm, so dividing Equation (1) by the upwind layer density at $k \pm ½$,

$$w(k) = -\frac{1}{\rho_{k\pm 1/2}}\sum_{0}^{k}\left(\frac{\Delta\rho}{\Delta t} + \nabla_H \cdot \rho V_H\right)_k \Delta z_k,$$

leads to a stronger downward velocity profile (divide by smaller density above k) and weaker upward velocity profile (divide by larger density below k) for the two example grid columns. This problem is exacerbated when the vertical grid structure expands with altitude (as is usually the case in photochemical models), so the largest biases are generated in the uppermost and thickest layers.

Alternative numerical methods were considered within the constraints that the chosen approach must remain implicitly stable, non-iterative, mass conservative, and positive-definite. The vertical solver was modified to use a centered in space differencing algorithm to maintain the use of a single matrix inversion, and the vertical velocity diagnosis was modified to be consistent by interpolating density to the vertical wind level. The backward-Euler time differencing was not altered. Centered differencing is second-order accurate, which reduces numerical diffusion, and as an added benefit, naturally balances the diagnostic velocity profile. The drawback is that implicit centered differencing offers no guarantee of a positive-definite solution in cases with large vertical concentration gradients (i.e., gradients of several orders of magnitude between adjacent layers). When the new algorithm leads to a negative concentration in a particular grid cell, a hybrid upstream-donor condition is applied for that cell to place a flux limitation on the local solution.

The modified version of CAMx was run for the April 2005 inert ozone test case, including the 2005 lateral BC, zero-gradient top BC, and 19-layer configuration. Fig. 5 compares the daily maximum ozone concentrations for the April test run using the original and revised vertical transport algorithms. The original vertical transport algorithm resulted in maximum April surface ozone concentrations in excess of 100 ppb throughout the Rocky Mountain States and over the Sierra Nevada Mountains with a peak ozone value of 153 ppb (as seen in Figs. 3 and 4). The new vertical advection algorithm produced maximum April ozone concentrations in the 50–70 ppb range with a peak of 79 ppb, levels that are more comparable with the observed values. Additionally, the model was subjected to sensitivity tests to assure it remained mass consistent and mass conservative. CAMx results using the modified vertical advection algorithm for the 19, 22, and 34-layer configurations are

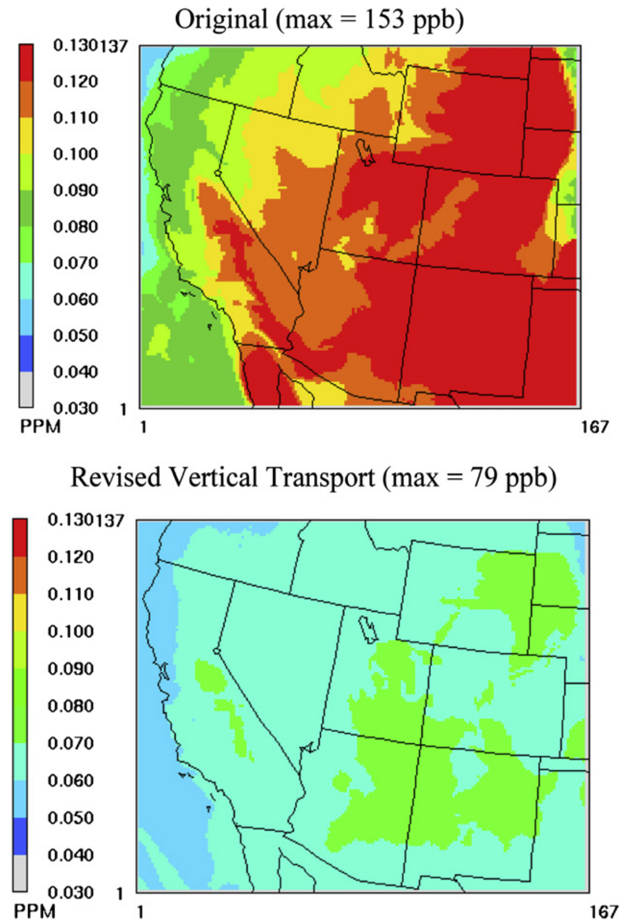

Fig. 5. April 2005 CAMx inert maximum ozone concentrations (ppm) using "zero-gradient" top BC, 2005 lateral BC, and original 19 layer vertical structure; original model (top) and revised vertical advection algorithm (bottom).

compared in Fig. 6. While some sensitivity to layer structure remains evident, the mitigation of numerical diffusion has reduced the impact of the additional layers.

3. 2005 Full chemistry evaluation

Based on the results from the inert CAMx tests described in Section 2, a final best configuration of the model was used to re-run the entire 2005 photochemical simulation described by Stoeckenius et al. (2009). In this case, CAMx was configured to run two grids simultaneously in a process referred to as "two-way nesting": the same 12 km grid used in the inert tests described earlier provided regional boundary conditions for a smaller domain at 4 km grid spacing that covered the Four Corners area of the southwest US (Utah, Colorado, Arizona and New Mexico). Ozone results were compared among three 12/4 km nested-grid CAMx runs, as described below:

Run A – original 2005 annual run reported by Stoeckenius et al. (2009):

- Original un-modified CAMx model;
- 19 vertical layers;
- Original monthly-averaged lateral BC developed from a 2002 GEOS-Chem simulation, with ozone in layer 19 set to the average ozone in layers 16–18;
- Original top BC (time/space invariant);

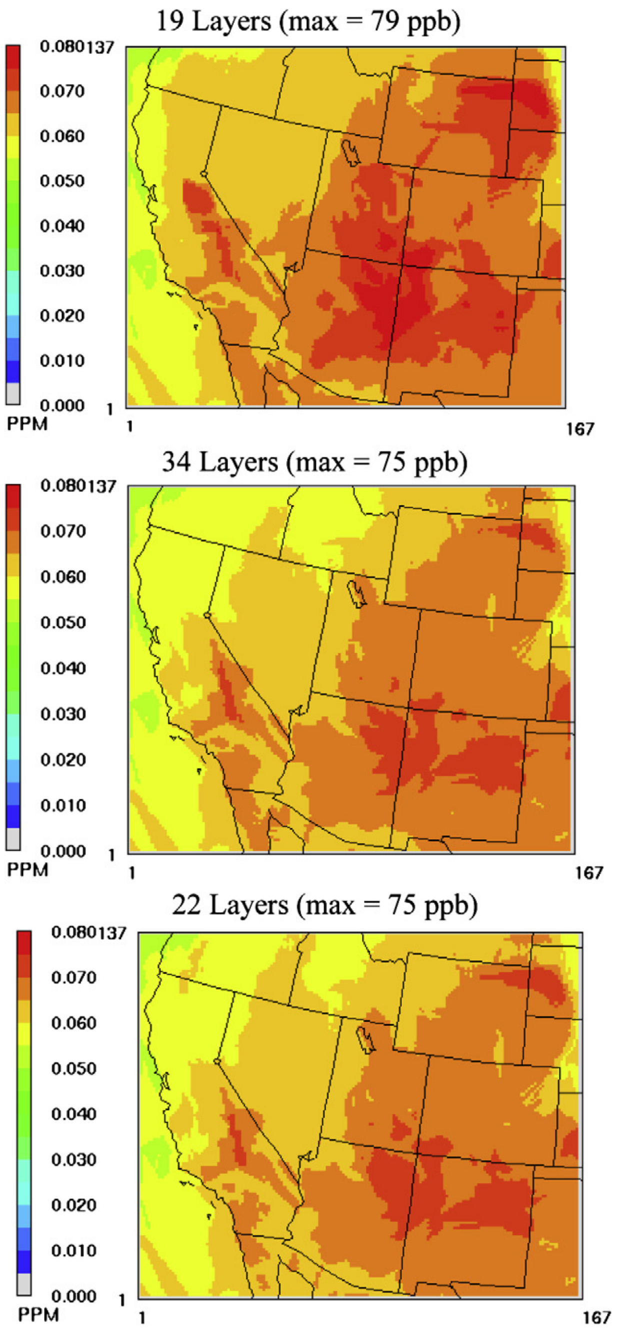

Fig. 6. April 2005 CAMx inert maximum ozone concentrations (ppm) using "zero-gradient" top BC, 2005 lateral BC, and revised vertical advection algorithm; original 19 layer vertical structure (top), 34 layer structure (middle), and 22 layer structure (bottom).

Run B – new 2005 annual run with improved inputs:

- Original un-modified CAMx model;
- 22 vertical layers;
- New 3-hourly lateral BC developed from a 2005 GEOS-Chem simulation (no arbitrary modifications);
- Original top BC (time/space invariant constant);

Run C – new 2005 annual run with the updated model:

- CAMx with new vertical advection algorithm;
- Internal zero-gradient top BC;
- Otherwise identical to Run B.

Table 2
AQS ozone monitoring sites in the Four Corners area (4 km grid) used to assess CAMx model performance.

Site ID	Site name	County	State
08-067-7001	Ignacio	La Plata	CO
08-067-7003	Bondad	La Plata	CO
08-083-0101	Mesa Verde	Montezuma	CO
35-045-0009	Bloomfield	San Juan	NM
35-045-1005	Farmington	San Juan	NM
N/A	Shamrock	La Plata	CO

The CAMx model performance evaluation included analyses of ozone predictions from each of the three CAMx runs listed above against available measurements at ground-level monitors. The approach followed the same methodology and used the same measurement data as described by Stoeckenius et al. (2009). The ozone evaluation was conducted using observational data from sites located in the 12 km and 4 km grids to generate monthly statistics for the entire simulation year. There were no upper air ozone measurement data for the 2005 period in the Four Corners Area. Routine surface measurements of ozone were taken from the US Environmental Protection Agency's (EPA) Air Quality Subsystem (AQS) and Clean Air Status and Trends Network (CASTNET) databases. Tables 2 and 3 list the sites from each measurement network used in this evaluation. Except for the "Shamrock" site, the AQS system includes mostly urban-oriented sites, and in the Four Corners domain these sites are located mostly near large towns in northeast New Mexico and southwest Colorado. CASTNET sites are located in rural areas, and as such were used to evaluate predictions at high elevations (>2 km) of the Rocky Mountains in the regional 12 km grid.

Figs. 7 and 8 display simulated maximum daily peak 8-hour surface ozone throughout the 12 km CAMx modeling grid for the months of April and July 2005, respectively. The introduction of updated 2005 lateral BC (Run B) resulted in much higher April surface ozone over elevated terrain (particularly the Rockies and Sierra Nevada) relative to the original run (Run A). This result was expected because the new 2005 BC was much higher than the original 2002 BC, especially in the upper troposphere. Even with the better layer structure, the vertical velocity algorithm was too diffusive and transported upper-level ozone to the surface rather efficiently. July ozone patterns also showed an impact over the Rockies and especially in urban areas. We expect the July signal was more the result of introducing the 22-layer structure rather than differences in lateral BC since the ozone aloft among the two runs were not as different in July as they were in April.

With the introduction of the new vertical advection algorithm and zero-gradient top BC (Run C), April surface ozone was reduced significantly throughout the Intermountain West. Note that April ozone patterns were similar to Run A in the Four Corners area and generally lower than Run A in other areas. Recall that in Run A, upper tropospheric ozone was artificially reduced from the

Table 3
High elevation CASTNET ozone monitoring sites in the Rocky Mountains (12 km grid) used to assess CAMx model performance.

Site ID	Site name	Elevation (m)	State
YEL408	Yellowstone	2400	WY
PND165	Pinedale	2388	WY
CNT169	Centennial	3178	WY
GTH161	Gothic	2926	CO
ROM206	Rocky Mountain	2743	CO
MEV405	Mesa Verde	2165	CO

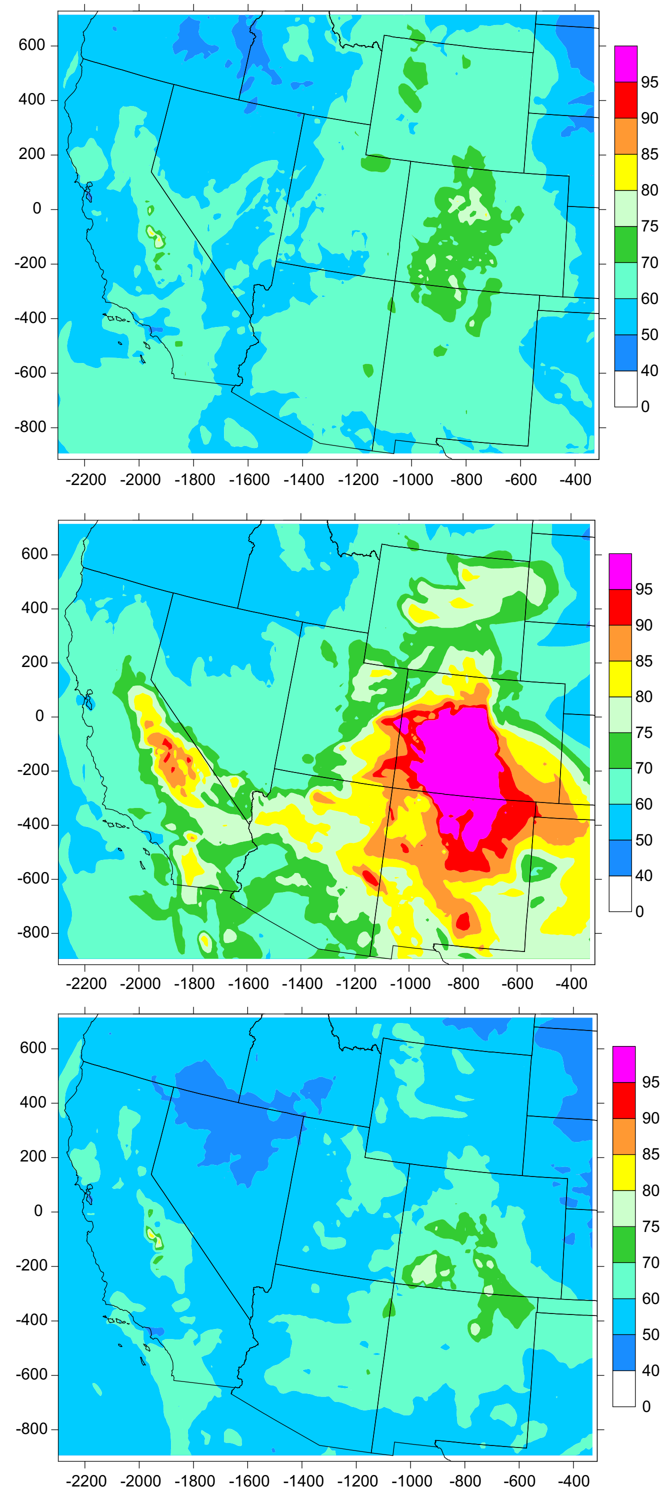

Fig. 7. April 2005 maximum daily peak 8-h ozone in the CAMx 12-km modeling grid; Run A (top), Run B (middle), and Run C (bottom).

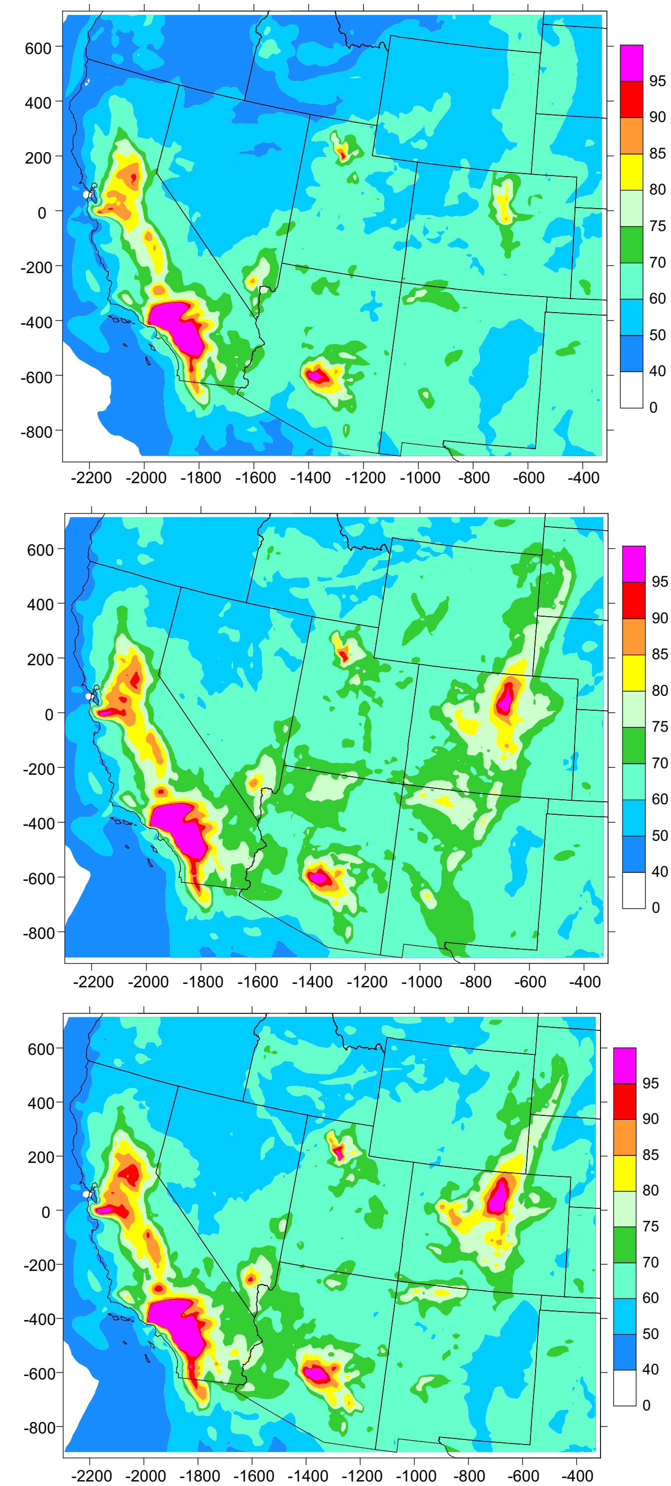

Fig. 8. July 2005 maximum daily peak 8-hour ozone in the CAMx 12-km modeling grid; Run A (top), Run B (middle), and Run C (bottom).

topmost layers while in Runs B and C ozone concentrations were maintained at the values directly extracted from the 2005 GEOS-Chem output. The improved vertical advection and top BC algorithms reduced the excessive April surface ozone generated in Run B to levels near or below the Run A levels without the need for arbitrary manipulation of the lateral BC. July ozone results were similar between Runs B and C, again supporting our expectation that differences in July were caused mainly by the 22-layer structure, while the improved vertical velocity algorithm had a much smaller effect. The fact that Run C maintained slightly higher ozone in urban areas (even higher than Run B) further suggests that a reduction in numerical diffusion likely allowed ozone to build up in the boundary layer where it was formed from fresh emissions.

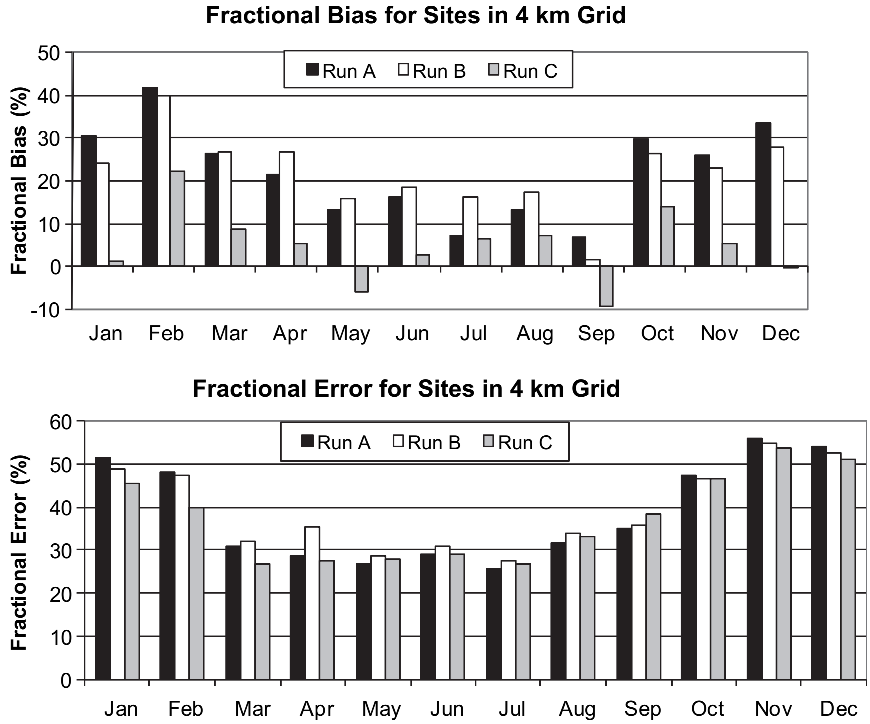

Fig. 9. Monthly ozone fractional bias (top) and error (bottom) for CAMx Runs A, B, and C based on all ozone measurements within the 4-km modeling domain.

Fig. 9 displays monthly means of fractional bias and error statistics for hourly ozone at all monitoring sites within the 4 km domain. Fractional metrics were chosen over normalized bias and error to evaluate performance since fractional statistics better address cases where observations approach zero concentrations (Seigneur et al., 2000). Results from all three CAMx simulations are shown. The performance in Run B was similar to the original Run A, yet the former indicated a tendency for slightly larger ozone over

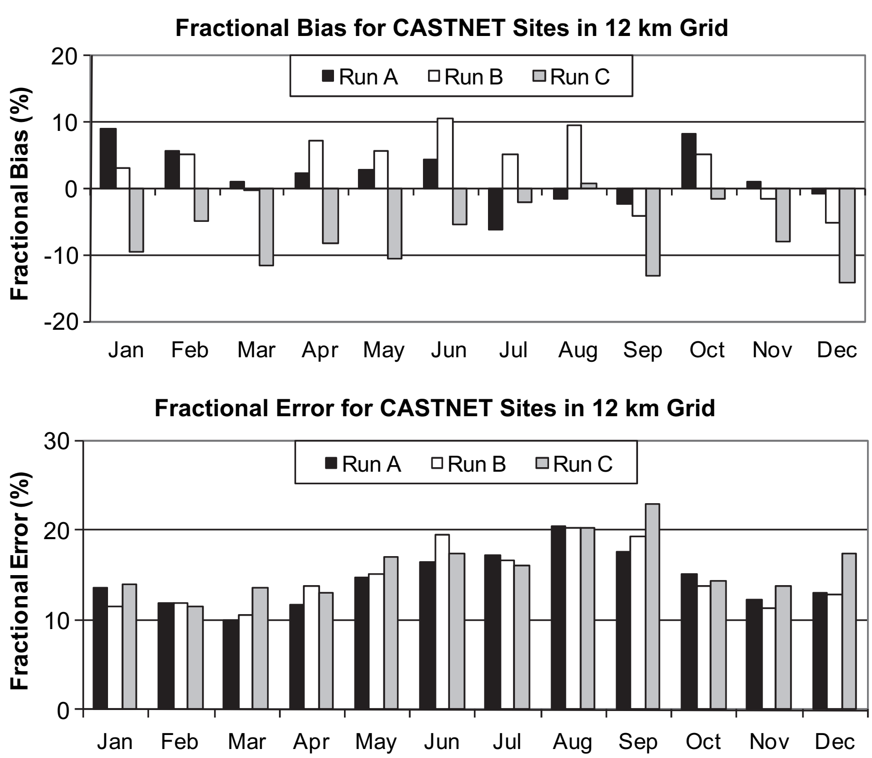

Fig. 10. Monthly ozone fractional bias (top) and error (bottom) for CAMx Runs A, B, and C based on selected high-elevation CASTNET ozone measurements within the 12-km modeling domain.

predictions in the spring and summer months. The performance in Run C showed a marked improvement in bias in all months, but only minor improvements to overall error (a surrogate for correlation). This means that while the model performed much better at simulating mean ozone on daily or longer time scales, hourly variability was not significantly improved. This signal agreed with hourly ozone time series plots (not shown), in which the model exhibited an inability to replicate the full range of the diurnal ozone profile. In these tests, the reduction in systematic bias was a more important signal because the improvements to BC and the vertical advection algorithm were expected to impact longer-term regional ozone patterns, as opposed to small-scale fluctuations brought about by local influences.

Fig. 10 displays similar monthly mean fractional bias and error statistics for hourly ozone at high elevation Rocky Mountain CASTNET sites within the 12 km domain. In this case, impacts to both bias and error were stronger. Run C exhibited a large and consistent downward shift toward under predictions, averaging between −5 and −10%. This shift also impacted fractional error, meaning that under predictions were sufficient in frequency and large enough in magnitude to increase observation-prediction variability (or scatter) on a wider range of time and space scales relative to results on the 4 km grid. On the other hand, the intermediate run (Run B) with only changes to lateral BC and layer structure exhibited an upward shift in bias during spring and summer months.

4. Conclusion

The CAMx and CMAQ photochemical models have over predicted springtime surface ozone concentrations in the complex terrain of the western US by 20 ppb or more. Previous investigations into the cause revealed that excessive vertical transport over mountainous terrain was drawing down stratospheric ozone concentrations at the top of the model that are introduced by the lateral boundary conditions. Both “off-line” models have possessed a similar internal diagnosis of vertical velocity in order to conserve mass and to maintain mass consistency with the input density and wind fields. In contrast, “on-line” models may not have this problem as long as chemical tracers are advected using the same dynamics solver. Furthermore, both CAMx and CMAQ have possessed similar first-order accurate vertical advection algorithms. These similarities, in conjunction with the historical use of coarse vertical resolution of the upper troposphere, resulted in similar ozone performance issues for both models. In the past, these over predictions have been alleviated by simply reducing the high ozone boundary conditions in the upper model layers.

This work investigated several potential solutions to correct or mitigate the vertical transport of ozone over complex terrain within the CAMx photochemical model. We first investigated several methods to reduce excessive vertical transport within CAMx that were designed to externally modify input horizontal wind fields: specifically, wind field smoothing, divergence minimization, and mass flux filtering. Additionally, improved vertical resolution, improvements to the BC inputs, and alternative vertical transport algorithm were examined. Tests were conducted with CAMx treating ozone as a single inert tracer to evaluate impacts on three-dimensional transport.

The two wind filters examined in this work helped reduce the magnitude of vertical velocity in upper layers with little impact on horizontal wind fields, but they could not adequately reduce the vertical transport of stratospheric ozone to the surface. Divergence minimization exhibited unexpected numerical features that that were both troubling and difficult to understand. These first tests revealed that the largest surface ozone impacts were caused by artificial dilution in the topmost layer associated with the simplistic and arbitrary top boundary conditions. By employing a more consistent, dynamically determined “zero-gradient” top boundary condition, the gains obtained by filters and divergence minimization on surface ozone patterns were nearly entirely removed.

Tests in which the vertical layer structure was expanded from 19 to 34 layers (i.e., the same number of layers used in the meteorological model) resulted in large and widespread surface ozone reductions without any modifications to the input wind fields. While this result is a strong testimonial for maximizing vertical resolution in areas of complex terrain, it leads to a significant impact on model computational speed. An intermediate 22-layer grid structure led to improvements similar to the 34-layer structure while maintaining acceptable speed performance. Finally, we improved the characterization of lateral BC and developed higher-order accurate vertical velocity and advection solver algorithms in CAMx. The vertical transport updates proved to yield the most successful results in reducing stratospheric ozone transport to the surface.

A final best configuration of CAMx was run for the entire 2005 calendar year to simulate ozone photochemistry over the western U.S. This configuration utilized the modified CAMx vertical advection algorithms, the intermediate 22-layer grid structure, and the updated lateral BC. Ozone performance in the Four Corners area of the western US showed that the updated model resulted in better performance than the original run in which arbitrary modifications to the lateral boundary conditions were necessary to achieve acceptable results.

The revised CAMx vertical transport algorithm has been made publicly available in version 5.20 (ENVIRON, 2010). The US EPA has independently incorporated similar changes into CMAQ (Young et al., 2009) and has released the model as version 4.7.1. CAMx and CMAQ modeling analyses for ozone and particulate matter are ongoing throughout the Rocky Mountain region of the western US. These applications will continue to employ updated emission estimates and to simulate various projection scenarios to assess the future direction of air quality in this pristine environment.

Acknowledgements

This work was funded by the American Petroleum Institute.

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