Development and evaluation of the Screening Trajectory Ozone Prediction System (STOPS, version 1.0)

Beata H. Czader[1], Peter Percell[1], Daewon Byun[1+], Soontae Kim[1,2], and Yunsoo Choi[1]

[1]Department of Earth and Atmospheric Sciences, University of Houston, Houston, USA

[2]currently at Ajou University, Suwon, S. Korea

[+]deceased

Correspondence to: B. H. Czader (bczader@uh.edu)

Abstract

A hybrid Lagrangian-Eulerian based modeling tool has been developed using the Eulerian framework of the Community Multiscale Air Quality (CMAQ) model. It is a moving nest that utilizes saved original CMAQ simulation results to provide boundary conditions, initial conditions, as well as emissions and meteorological parameters necessary for a simulation. Given that these file are available, this tool can run independently from the CMAQ whole domain simulation and it is designed to simulate source – receptor relationship upon changes in emissions. In this tool, the original CMAQ's horizontal domain is reduced to a small sub-domain that follows a trajectory defined by the mean mixed-layer wind. It has the same vertical structure and physical and chemical interactions as CMAQ except advection calculation. The advantage of this tool compared to other Lagrangian models is its capability of utilizing realistic boundary conditions that change with space and time as well as detailed chemistry treatment. The correctness of the algorithms and the overall performance was evaluated against CMAQ simulation results. Its performance depends on the atmospheric conditions occurring during the simulation period with the comparisons being most similar to CMAQ results under uniform wind conditions. The mean bias for surface ozone mixing ratios varies between -0.03 ppbV and -0.78 ppbV and the slope is between 0.99 and 1.01 for different analyzed cases. For complicated meteorological condition, such as wind circulation, the simulated mixing ratios deviate from CMAQ

values as a result of Lagrangian approach of using mean wind for its movement, but are still close, with the mean bias for ozone varying between 0.07 ppbV and -4.29 ppbV and slope varying between 0.95 and 1.06 for different analyzed cases. For historical reasons this hybrid Lagrangian – Eulerian based tool is named the Screening Trajectory Ozone Prediction System (STOPS) but its use is not limited to ozone prediction as similarly to CMAQ it can simulate concentrations of many species, including particulate matter and some toxic compounds, such as formaldehyde and 1,3-butadiene.

1. Introduction

Air pollution modeling is used to predict concentrations of pollutants and to understand physical and chemical processes involved as well as to develop necessary control strategies to improve air quality. Air pollution can be numerically simulated by several techniques that, based on the frame of references, are generally divided into two categories: Eulerian and Lagrangian.

In the Eulerian approach, the observer adopts a fixed frame of reference, usually the surface of the earth, with the modeling domain divided into many grid cells. This enables easy representation of the pollutant production and transformation processes. Most Eulerian models account for atmospheric dynamics (horizontal and vertical advection and diffusion), emissions sources, and chemical production and destruction. They are often used to forecast air quality. A widely used Eulerian type model is the Community Multiscale Air Quality (CMAQ) model developed by the U.S. Environmental Protection Agency (Byun and Schere, 2006).

Lagrangian (or Trajectory) models are based on species conservation equations describing atmospheric diffusion and chemical reactions stated in terms of moving coordinates. The observer adopts moving coordinates that follow sets of hypothetical air parcels. The air parcels move along with the prevailing winds (being advected), there is

no mass exchange between parcels and surroundings except emissions of pollutants that are accounted for when the air parcels pass over source regions Lagrangian models have much shorter run times and are therefore more computationally efficient than their chemical transport counterparts. These models have been successfully applied to simulate dispersion of several pollutants over length scales of the order of a few tens of kilometers or lesser. Often they do not account for chemical transformations as the chemistry is modeled as first order decay of pollutants; in such cases they are unable to adequately predict the atmospheric concentrations of species with short lifetimes, such as fast reacting ozone-forming VOCs and air toxics, an example of which is 1,3-butadiene.

An ideal air pollution model would combine the computational efficiency of a dispersion model with the chemistry details of a chemical transport model. In other words, it would be a hybrid system merging a chemical transport model with a Lagrangian movement. This paper presents the development, validation and an example of application of a hybrid modeling approach that utilizes Lagrangian advection scheme in an Eulerian modeling framework. This hybrid Eulerian-Lagrangian based modeling tool was designed to re-simulate only a part of a CMAQ modeling domain that is of interest. This makes it a computationally efficient tool to study a source-receptor relationship, such as the effect of emission events on the ozone concentration. In addition, it can quickly perform the analysis of physical and chemical process, so called process analysis, which is very time consuming to perform using the full-domain Eulerian air quality grid model. Compared to Lagrangian column models our approach has advantages of using detailed chemistry and dynamic boundary conditions. To assure the correctness of the algorithm's implementation, the results were thoroughly evaluated and compared with the CMAQ simulation results.

Currently many institutions perform air quality forecasting. When implemented into the real-time air quality forecasting this Eulerian-Lagrangian based hybrid tool can be used for a time efficient re-simulation utilizing the same inputs as already prepared for the

forecasting. As emission source can be directly added to this tool it can simulate effects of additional (non-routine) emission releases that are not included in the standard inventory, for example 'upset' emissions from industrial facilities or wild fire emissions. Other application could be a simulation of plumes form chemical industry upon hurricane damage or upon a release of chemical or biological agents. It can also be utilized to provide detailed process analysis information (a contribution of physical and chemical process to a simulated mixing ratio) for a moving window domain to capture chemical evolution of plumes. Performing process analysis is also very time consuming and it is not used in the air quality forecasting applications.

A hybrid modeling approach was previously used to simulate concentrations of benzene in Houston (Stein et al., 2007). It consisted of CMAQ, the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model, and the AMS/EPA Regulatory Model (AERMOD), where CMAQ was used to provide background concentrations. Although it successfully predicted benzene concentrations it has limitations in AERMOD being a steady-state plume dispersion model, which does not consider chemistry, and therefore, it is not suitable for simulations of more reactive species or secondary (not emitted) species. Lagrangian approaches were also developed for the purpose of detailed analysis of chemical interactions inside a plume. For example, Kimura at al., 2008 implemented algorithms inside grid model that allow tracking plume inside the grid model (Lagrangian approach) and to provide details of chemical transformations inside a plume. However, this tool does not operate independently from the host model, making re-simulation time consuming. Henderson et al. (2011) reported a pseudo-Lagrangian post-processing tool, which can be used outside the grid model to analyze its outputs in order to identify plumes and perform process analysis of the plume. In contrast, our tool can be run independently from the whole domain simulations of grid model and is designed to simulated effect upon emissions changes.

2. Development of a hybrid Eulerian-Lagrangian based modeling approach

A hybrid Eulerian-Lagrangian based modeling tool is derived from the CMAQ model in which the original CMAQ's horizontal domain is reduced to a small sub-domain that can move along a specific trajectory. Although not rigorously correct, as there is in- and out-flow through the domain boundaries that is in contrast to Lagrangian ideas, it was "inspired by Lagrangian methods" while taking advantage of the existing simulation machinery in CMAQ. Initially developed for ozone pollution applications was named the Screening Trajectory Ozone Prediction System (STOPS). Although it is not limited to ozone prediction, but similarly to CMAQ, it can simulate concentrations of many species, including particulate matter and some toxic compounds, such as formaldehyde and 1,3-butadiene, for historical reason we continue to use the name STOPS.
CMAQ domain is divided into grid cells with certain number of rows and columns in a horizontal direction and layers in a vertical direction. STOPS can be considered as a sub-domain of CMAQ, which is also divided into a grid cells in horizontal and vertical direction but opposite to CMAQ, STOPS domain moves with the mean wind as presented in Fig. 1. For each grid cell in a domain CMAQ calculates horizontal and vertical advection, horizontal and vertical diffusion, dry and wet deposition, chemical reactions in gas, aquas and particle phase, as well as photochemical processes and chemistry in clouds. The vertical layer structure and the physical and chemical processes in STOPS are the same as in the full domain CMAQ model, except that advection fluxes are obtained utilizing difference between a cell horizontal wind velocity and averaged velocity of STOPS. At its starting position STOPS grid is aligned with CMAQ grid, but as it moves with wind its grid may not necessarily align with CMAQ grids (see Fig. 1). The initial location of the STOPS domain can be defined by choosing position of the domain middle cell in terms of latitude and longitude coordinates or in terms of the column and row number corresponding to the CMAQ full domain. STOPS uses initial condition and the dynamic boundary conditions from saved original CMAQ simulation results as well as emission and meteorological parameters as

prepared for CMAQ. Because of that STOPS movement is limited by CMAQ domain boundaries.

Usually input and output files have hourly values. The calculation of science processes in CMAQ as well as in STOPS is based on so call synchronization time step, which is in a range of seconds to minutes and determined by the model to satisfy the Courant condition safe advection time step. Both, CMAQ and STOPS perform temporal interpolation of hourly values (initial conditions, boundary conditions, emissions, and meteorological parameters) to obtain a value at a smaller calculation time steps. In STOPS, in addition to temporal interpolation, we also added spatial interpolation. It was needed for cases when the STOPS grid cells do not align with the grid cells of a CMAQ files.

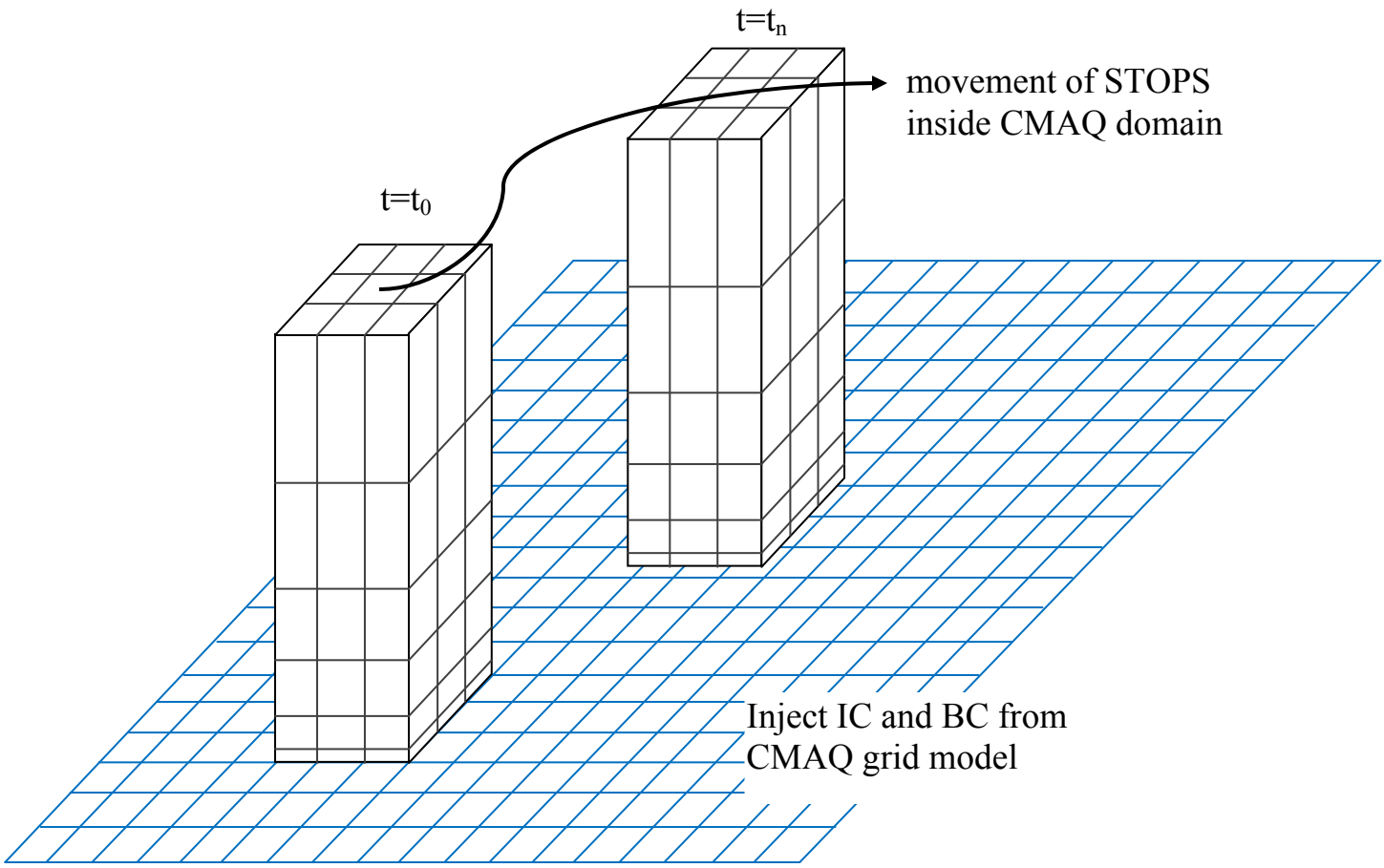


Figure1. The conceptual model of STOPS and its movement.

The trajectory for STOPS movement is calculated based on the mean wind in the middle column (thereafter mwind) that is averaged from surface layer up to the Planetary Boundary Layer (PBL) height and weighted by differences in pressure in each layer. The u and v components of wind (m/s) were calculated according to the following equations:

$$\tilde{u} = \frac{1}{\sum_{L=1}^{PBL} \Delta\sigma_F(L)} \sum_{L=1}^{PBL} u_L \cdot \Delta\sigma_F(L) \qquad (1a)$$

$$\tilde{v} = \frac{1}{\sum_{L=1}^{PBL} \Delta\sigma_F(L)} \sum_{L=1}^{PBL} v_L \cdot \Delta\sigma_F(L) \qquad (1b)$$

Where σF = 1-σ and σ (unitless) is a scaled atmospheric pressure in a sigma coordinate system defined as follow:

$$\sigma = \frac{(p - p_t)}{(p_s - p_t)} \quad (2)$$

where p is a pressure at the current level, p_t is a model top pressure, p_s is a surface pressure.

The trajectory can be also determined based on the averaged value from all cells inside STOPS domain (hereafter awind) as opposed to the middle column value.

The implementation of STOPS required modifications of the CMAQ source code which included the following:

- A Fortran-90 module, STOPS_MODLUE, was created to hold the additional data structure related to STOPS and subroutines associated with a coordinate conversion, position and velocity along the trajectory.
- The SUBHFILE subroutine was modified. This subroutine determines the spatial relationship between the CMAQ grid and grids of input data, e.g., inputs with emission or meteorological data may have different horizontal domains that the CMAQ domain. SUBHFILE subroutine was enhanced to support a moving horizontal sub-domain, whose grid points do not necessarily coincide with grid points of the input data, and may have different locations at every synchronization time step.
- The boundary subroutine, RDBCON, was modified to support a boundary thickness of 3 cells and to get boundary values for changing locations directly from the CMAQ full-grid concentration file.
- The netCDF output file, CONC, saves only STOPS grid concentrations. In addition, an ASCII output file is generated that holds trajectory information, this is latitude and longitude of the middle point of the STOPS domain for each output time step, along with the corresponding column and row numbers of a full CMAQ domain.

- For source-receptor applications the STOPS code was modified in a way that additional emissions can be directly injected into STOPS without a need of reprocessing an emission inventory. A name of the emitted compound(s) (in terms of model species), a location of emission release, starting and ending times, and the amount need to be specified by the user in the STOPS run script.
- Given that STOPS is based on the CMAQ source code and uses the same input files its results shall closely approximate those obtained with the 3-D CMAQ model. For the purpose of comparing STOPS results against CMAQ results the post processing program was developed and incorporated into the STOPS build and run scripts. With this, additional file, HCONC, is generated from the STOPS simulations. It holds CMAQ concentrations from grid cells that correspond to the current location of STOPS.

The advantage of STOPS compared to other Lagrangian models is the capability of utilizing realistic boundary conditions that change with space and time. Because of that, STOPS takes into account flow in and out of a domain, allowing for an exchange of mass between a moving domain and surroundings. This allows for simulations of conditions when a wind shear occurs for which the usual Lagrangian models are usually not suitable. On the other hand, in the case of significant deviations in a wind speed and direction some mass may be blown out of the STOPS simulation domain.

3. Verification of STOPS performance

CMAQ has been found to be a reliable modeling tool, whose performance has been evaluated in many studies [Smyth *et al*., 2006; Eder and Yu, 2006; Arnold and Dennis, 2006; Byun *et al*., 2007; Appel et al., 2012]. As a moving nest, which uses the same inputs as CMAQ and utilizes CMAQ's simulations results as dynamic boundary conditions and initial conditions, the STOPS performance is expected to be close to the

results of the original CMAQ model; therefore, the code implementation was verified by comparing its simulation results with those obtained using CMAQ.

The following statistical parameters were calculated for performance evaluation:

Number of dataset
$$N = NCOL * NROW * NTSTEP \quad (4)$$

Mean of host concentration
$$\overline{H} = \frac{1}{N}\sum_{i=1}^{N} H_i \quad (5)$$

Mean of STOPS concentration
$$\overline{S} = \frac{1}{N}\sum_{i=1}^{N} S_i \quad (6)$$

Mean Bias
$$MB = \frac{1}{N}\sum_{i=1}^{N} (H_i - S_i) \quad (7)$$

Mean Absolute Error
$$MAE = \frac{1}{N}\sum_{i=1}^{N} | H_i - S_i | \quad (8)$$

Root Mean Square Error
$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N} (H_i - S_i)^2 \right]^{\frac{1}{2}} \quad (9)$$

where H_i and S_i corresponds to instantaneous mixing ratios obtained with CMAQ and STOPS, respectively. NCOL and NROW are numbers of STOPS columns and rows, respectively, and NTSTEP is number of output time steps.

Daily ozone maximum values from CMAQ and STOPS simulations where also calculated and are indicated as *HMAX* and *SMAX*, respectively.

For cases when STOPS grids do not align with CMAQ grids the CMAQ values from several grid celsl corresponding to a STOPS cell are interpolated by means of weighted averaging of CMAQ values.

We performed verification for three cases: (1) a case when the STOPS domain does not move, which was performed to test an effect of boundary condition on STOPS results;

(2) cases with STOPS moving along different trajectories performed to test STPOPS performance for different atmospheric conditions as well as an effect of different ways of trajectory calculation on STOPS results; (3) cases with different STOPS domain sizes to test an effect of domain size on the STOPS results.

3.1 Effect of boundary conditions

First, the correctness of the STOPS code implementation was verified by performing STOPS simulations in the stationary mode, this is when it is not moving. In this configuration STOPS domain is like a CMAQ sub-domain in which the grid cells are aligned with CMAQ grid cells; thus, STOPS calculated values can be directly compared with CMAQ values from corresponding grid cells. With this setup, STOPS does not perform spatial interpolations of either initial or boundary values. The simulations were performed for three domains, differing in size and starting positions as presented in Fig. 2: “Houston” domain, “urban” domain that sits in the urban area and “industrial” domain that is over the industrial region. The size of a domain is defined by a number of padding cells around the middle cell. The location of the middle column in each STOPS domain relatively to the CMAQ (host) grid, number of padding cells in each direction around STOPS middle column, and a number of total STOPS columns and rows are presented in Table1. A 1x1 STOPS domain is possible, but is more likely to quickly lose the effect from a perturbation in the domain, like modified emissions. Thus, it is not likely to be used in practice and we did not perform tests on that domain.

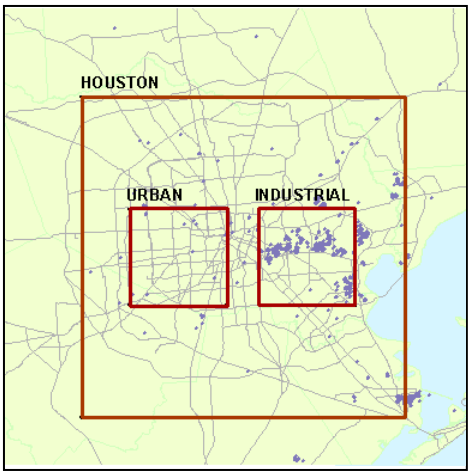


Figure.2. Starting locations of STOPS domains. Points indicate location of emission point sources in Houston.

Table1. Specifications of STOPS domains

Name	Column and row of middle STOPS cell in a host grid	Number of padding cells in each direction	Number of rows of STOPS domain	Number of columns of STOPS domain
Houston	25, 30	10	21	21
Urban	21, 30	2	5	5
Industrial	29, 30	2	5	5

Usually CMAQ boundary conditions as well as other input files are provided at hourly intervals and CMAQ calculated mixing ratios are also saved at hourly intervals. However, calculations are performed at much smaller time intervals that could be on the order of minutes; therefore, the boundary values are interpolated from two corresponding hourly values to match a specific computation time step. This is also a case for STOPS. For the comparison of STOPS results with CMAQ values we used CMAQ concentrations from the grid cells corresponding to cells in STOPS domain. These gird cells in CMAQ are not at the domain boundaries but inside domain; therefore, in these grid cells advection is calculated based on values from adjacent cells at every synchronization time step. In STOPS these cells are at the domain boundary

and hourly boundary values are interpolated for advection calculation. Because of that, we expect some differences between STOPS and CMAQ calculated mixing ratios. To justify them, CMAQ and STOPS simulations were performed for different output time steps, which were set to 1 hour, 5 minutes, and 1 minute. This allows for obtaining boundary conditions at small time steps, which is close to the synchronization time step and forcing CMAQ and STOPS to use the same values for advection calculation.

Three sample days out of the TexAQS 2000 episode were chosen for simulations: August 25, 28, and 30. For all cases the STOPS simulation started at 12 UTC and lasted 12 hours. Surface ozone values from CMAQ and STOPS were compared at each cell and each simulations output time step. The summary of statistical parameters calculated by CMAQ and STOPS in a stationary mode is presented in Table 2. Differences between the concentrations obtained from these two models are attributed to different values at the domain boundaries. Decreasing the hourly output time step to make it closer to the synchronization time step lessens the effect of different boundary conditions as STOPS values became closer to CMAQ values. At 1 minute output time step differences between ozone concentrations are less than 1 ppbV. Figure 3 shows comparison of STOPS and CMAQ values from simulation with 1 hour output time step (left) and 1 minute time step (right) with less scattering from 1 minute output time step, confirming that shortening the output time step makes STOPS results closer to CMAQ.

Table 2. Summary of statistical parameters for STOPS and CMAQ predicted ozone mixing ratios, when STOPS was used in the stationary mode. "hou" indicates results from the Houston domain; "ind" –from the industrial domain; "urb" –from urban domain.

NAME	N	HMAX	SMAX	MB	MAE	RMSE
stat_hou_1h.0825	5733	162.1	162.9	-0.1894	0.3822	0.6820
stat_hou_1h.0828	5733	115.6	115.8	-0.1160	0.1979	0.3229
stat_hou_1h.0830	5733	158.7	158.7	-0.3089	0.3870	0.5920
stat_hou_5m.0825	63945	166.4	167.1	-0.1183	0.2067	0.3946
stat_hou_5m.0828	63945	116.0	115.7	0.0369	0.1213	0.2075
stat_hou_5m.0830	63945	160.3	160.5	0.0167	0.1297	0.2295
stat_hou_1m.0825	317961	166.0	166.0	0.0140	0.0456	0.0906
stat_hou_1m.0828	317961	115.1	115.1	-0.0117	0.0365	0.0744
stat_hou_1m.0830	317961	158.9	158.9	-0.0138	0.0308	0.0715
stat_ind_1h.0825	325	108.7	113.9	-0.8562	1.0007	1.4691
stat_ind_1h.0828	325	88.5	88.0	-0.7096	0.8004	1.1424
stat_ind_1h.0830	325	145.1	147.8	-1.8936	1.9774	2.6690
stat_ind_5m.0825	3625	111.6	112.8	-0.5794	0.6502	0.9494
stat_ind_5m.0828	3625	88.6	87.7	-0.2883	0.4229	0.6003
stat_ind_5m.0830	3625	148.2	148.4	-0.4536	0.5636	0.7370
stat_ind_1m.0825	18025	112.0	112.6	-0.1275	0.2107	0.3356
stat_ind_1m.0828	18025	86.6	86.6	-0.0724	0.1045	0.1426
stat_ind_1m.0830	18025	146.6	146.7	-0.0974	0.1342	0.2249
stat_urb_1h.0825	325	162.1	161.4	-0.9287	1.3587	2.1596
stat_urb_1h.0828	325	69.2	70.7	-0.5708	0.6402	0.9812
stat_urb_1h.0830	325	145.9	148.0	-1.5667	1.5673	1.9527
stat_urb_5m.0825	3625	165.9	167.1	-0.5115	0.6070	0.9891
stat_urb_5m.0828	3625	70.5	71.0	-0.2271	0.3825	0.6278
stat_urb_5m.0830	3625	145.9	146.8	-0.3074	0.3411	0.4611
stat_urb_1m.0825	18025	165.4	165.8	0.0214	0.2073	0.3132
stat_urb_1m.0828	18025	69.9	69.7	-0.0300	0.0875	0.1292
stat_urb_1m.0830	18025	144.3	144.7	-0.1970	0.2114	0.3607

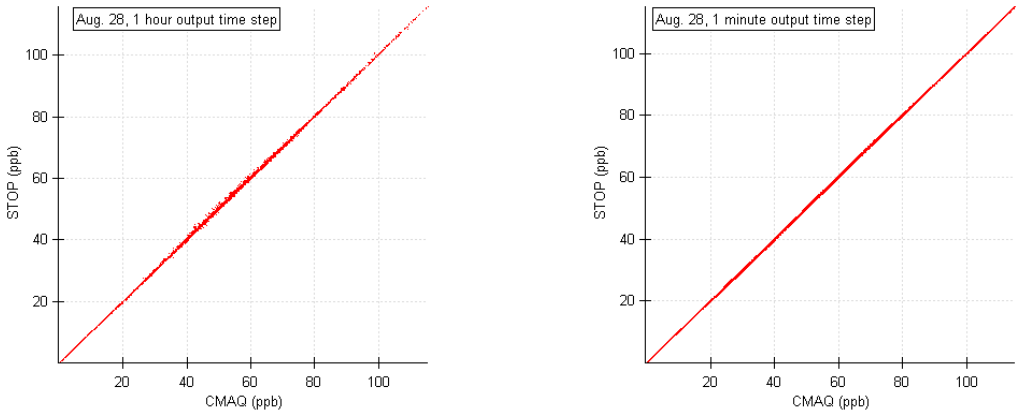


Figure 3. Comparison of CMAQ and static STOPS simulation results for Aug. 28 for 1 hour (left) and 1 minute (right) output time step. Both graphs correspond to simulation from the Houston domain.

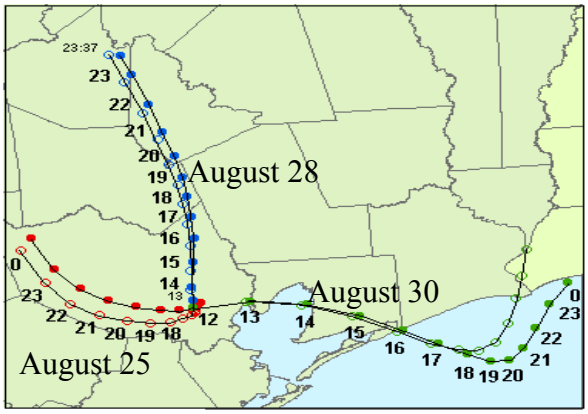
3.2 Uncertainties related to movement of STOPS

The next step in the STOPS verification was to analyze uncertainties related to the movement of a STOPS domain. A direct comparison between CMAQ and STOPS result was complicated due to the fact that when STOPS travels with wind its grid cells do not necessarily align with CMAQ grid cells. For the purpose of comparing STOPS values with CMAQ ones we utilized two approaches which were performed after STOPS finished its calculations. In the first approach we aligned the STOPS grid cells with the closest CMAQ grid cells (shifted the STOPS domain) and took the corresponding values for a comparison. In the second approach we performed spatial interpolation by calculating weighted average from several CMAQ grid cells that overlap with the STOPS grid cell. The performance evaluation was tested for these two approaches.

There are two options in STOPS that can be used for a trajectory calculation. A trajectory can be determined either based on the wind in the middle column of the STOPS domain as described by Eq. 1 (mwind) or based on the averaged value from the whole STOPS domain (awind). Two smaller sub-domains shown in Figure.2, which are urban and industrial, were selected for STOPS simulations in the moving mode with the two options for trajectory calculation being tested.

The days for which comparison was carried out were characterized by different meteorological conditions. August 25, 2000 was the day with complicated, circular wind patterns; on August 28th, 2000 strong, but uniform southerly winds were observed, and on Aug. 30 change of winds from south-easterly to south-westerly was observed in the early afternoon hours. STOPS trajectories for these three days, with the starting position at the location of industrial sub-domain, are presented in Figure 4. Trajectories determined based on the winds in the STOPS middle column are indicated by filled circles, and those determined based on the average winds in the whole STOPS domain with open circles. All trajectories start at 12 UTC and end the next day at 0 UTC, except trajectories on Aug. 28 that ended at 23 UTC due to subdomain reaching the boundaries of CMAQ domain earlier as an effect of strong winds on that day. On August 28 and 30 there are little differences in trajectories determined by the two different methods. However, as can be seen from Figure 4b, there are differences in trajectories for Aug. 25, especially during the first couple of hours of simulations. Both trajectories move south between hour 12 and 13 UTC. After that, the trajectory determined by the winds in the middle column moves east until 15 UTC and then west, making a circular pattern; at 17 UTC it comes back to the close proximity of the starting position. On the contrary, the trajectory determined by the winds averaged in the whole STOPS domain initially move south for couple of hours and then continuously moves west.

a)



b)

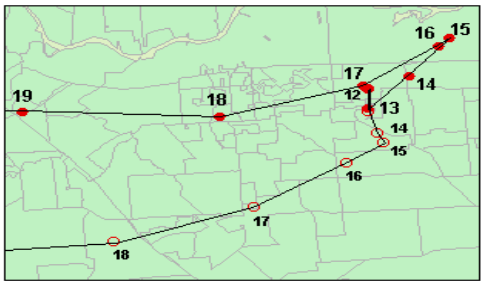


Figure 4. a) STOPS trajectories starting from the industrial sub-domain. Trajectories determined based on the winds in the STOPS middle column are indicated by filled circles, and those determined based on the average winds in the whole STOPS domain with open circles. Trajectories for Aug. 25 are indicated with red dots, those for Aug. 28 with blue dots, and for Aug. 30 with green dots. Numbers next to dots show UTC time b) details of the trajectory on Aug. 25.

In order to quantify the differences between numerous options available in STOPS several simulations were performed with changing the options one at a time. The analysis was performed for the cases when trajectory was determined based on the winds in the middle column (mwind) and the averaged winds in the whole STOPS domain (awind). The simulation results when the STOPS domain was shifted for the purpose of aligning its grids with CMAQ grid are indicated with 'sh'. The naming convention used to describe each case of interest is presented in the following example: 'awind_urb_1h.0825_sh' means that the trajectory was estimated based on the averaged winds in the whole STOPS domain, the trajectory starting position was urban sub-domain, the model output time step was set to 1 hour, the simulation was performed for Aug. 25, and the STOPS domain was shifted to be aligned with the host domain grid for the comparison purpose. The case 'awind_urb_1h.0825' means the same as above except that CMAQ concentrations were spatially interpolated to be compared with STOPS mixing ratios. Results of the statistical analysis of CMAQ and STOPS predictions of ozone concentrations when STOPS was used in the moving mode are presented in Table 3 for cases when simulations were initialized in the urban sub-domain and in Table 4 for starting positions in the industrial sub-domain.

Figure 5 shows scatter plots comparing CMAQ and STOPS concentrations of ozone for Aug. 25, 28, and 30 for the STOPS starting position at the urban sub-domain (left graphs) and industrial sub-domain (right graphs). Triangles correspond to values calculated with STOPS when its trajectory was determined based on the winds in the middle column (mwind), crosses to the trajectory obtained from the average winds in the whole STOPS domain (awind). Plotted are ozone mixing ratios from all cells in the

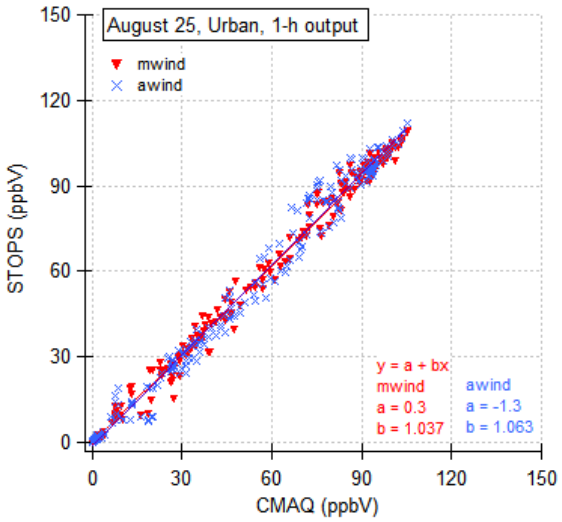
first model layer, at every output time step. Very good performance was found on Aug. 28 with the averaged mean absolute error of 1.3 ppbV and 1.5 ppbV for the urban and industrial domains, subsequently. Better agreement between CMAQ-STOPS concentration pairs was found when the STOPS trajectory was calculated based on the winds in the middle column. Shifting the STOPS domain to align it with the CMAQ grid resulted in better agreement than the case when CMAQ values had to be interpolated.

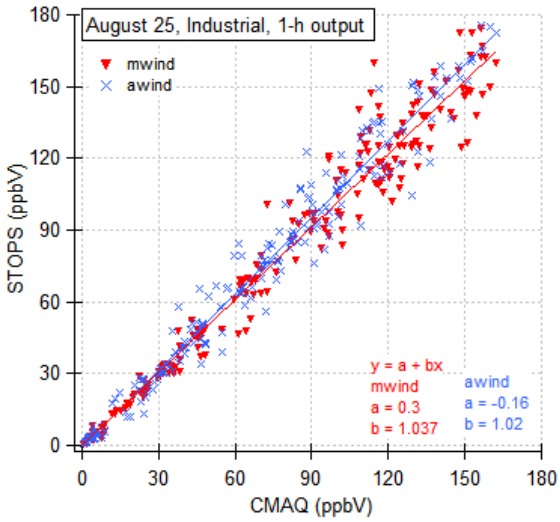
Table 3. A summary of statistical parameters for STOPS-CMAQ concentrations, when STOPS was used in the moving mode, with the starting position at the urban sub-domain.

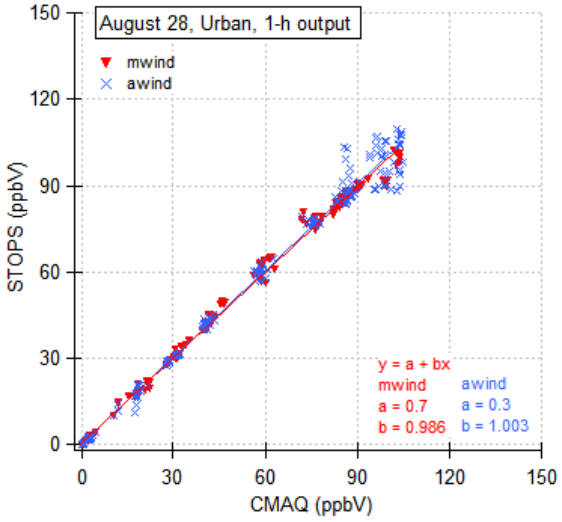
NAME	N	HMAX	SMAX	MB	MAE	RMSE
awind_urb_1h.0825	217	105.1	111.8	-1.7055	3.7246	5.4175
awind_urb_1h.0828	185	104.8	109.5	-0.5229	2.4865	4.1357
awind_urb_1h.0830	217	132.1	120.7	-0.6365	4.6031	7.0249
awind_urb_5m.0825	2329	107.9	108.1	-0.5235	2.9698	4.1889
awind_urb_5m.0828	1929	105.3	108.6	-0.062	2.2454	3.9979
awind_urb_5m.0830	2329	131.4	127.4	-0.9365	3.9527	5.9425
awind_urb_1m.0825	11545	107.8	107.3	-0.4557	3.1165	4.394
awind_urb_1m.0828	9449	103.2	109.2	-0.0297	2.2157	3.9464
awind_urb_1m.0830	11545	131.0	126.4	-0.8205	3.8026	5.743
mwind_urb_1h.0825	217	105.4	109.1	-1.5074	2.6628	3.8337
mwind_urb_1h.0828	169	104.0	102.4	-0.0594	1.4279	2.2759
mwind_urb_1h.0830	217	137.8	135.9	-0.5092	3.2716	5.2829
mwind_urb_5m.0825	2329	107.7	107.2	-0.663	2.4906	3.493
mwind_urb_5m.0828	1833	104.2	102.6	0.5222	1.8313	2.7969
mwind_urb_5m.0830	2329	137.6	137.5	-0.5207	3.8601	5.7908
mwind_urb_1m.0825	11545	107.8	106.5	-0.7221	2.6495	3.7622
mwind_urb_1m.0828	9129	103.0	101.4	0.6286	1.6039	2.4716
mwind_urb_1m.0830	11545	137.7	135.7	-0.0888	4.1309	6.0413
awind_urb_1h_sh.0825	325	108.2	111.8	-0.4767	1.521	2.3025
awind_urb_1h_sh.0828	275	105.0	109.5	-0.5584	1.5322	2.1738
awind_urb_1h_sh.0830	325	132.1	128.1	-0.1203	2.0124	3.16
awind_urb_5m_sh.0825	3625	110.0	108.1	-0.1248	1.4191	2.1658
awind_urb_5m_sh.0828	3000	105.5	109.4	0.0152	1.3118	2.1861
awind_urb_5m_sh.0830	3625	134.5	134.1	-0.4659	2.126	3.1923
awind_urb_1m_sh.0825	18025	110.7	107.3	0.0743	1.3337	1.9913
awind_urb_1m_sh.0828	14750	103.6	109.2	-0.0619	1.3074	2.2298
awind_urb_1m_sh.0830	18025	134.1	133.5	-0.1377	1.9516	2.9423
mwind_urb_1h_sh.0825	325	108.2	109.1	-0.1204	1.7139	2.5346
mwind_urb_1h_sh.0828	250	104.0	109.8	-0.3751	1.4664	2.7279
mwind_urb_1h_sh.0830	325	137.8	139.7	-0.1818	2.4477	3.7688
mwind_urb_5m_sh.0825	3625	108.9	107.2	-0.0929	1.4659	2.1744
mwind_urb_5m_sh.0828	2850	104.4	111.2	0.0849	1.1706	2.0956
mwind_urb_5m_sh.0830	3625	138.5	140.2	-0.5113	2.5097	3.7741
mwind_urb_1m_sh.0825	18025	109.2	106.5	-0.1237	1.3359	1.9914
mwind_urb_1m_sh.0828	14250	103.0	111.2	0.1064	1.2086	2.0841
mwind_urb_1m_sh.0830	18025	138.4	138.5	-0.4413	2.4165	3.5173

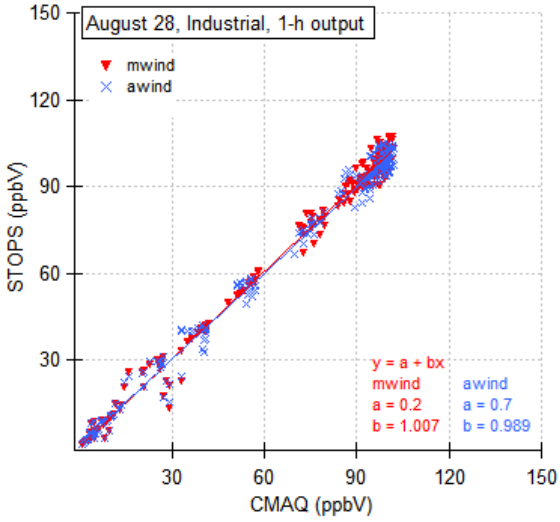
Table 4. A summary of statistical parameters for STOPS-CMAQ concentrations, when STOPS was used in the moving mode, with the starting position at the industrial sub-domain.

NAME	N	HMAX	SMAX	MB	MAE	RMSE
awind_ind_1h.0825	217	162.1	175.6	-3.7049	6.667	9.7334
awind_ind_1h.0828	201	102.0	104.5	-0.0743	2.7724	3.6884
awind_ind_1h.0830	217	141.4	140.1	0.5727	2.2085	3.4874
awind_ind_5m.0825	2329	166.2	179.9	-4.2896	6.9033	10.246
awind_ind_5m.0828	2281	102.0	105.4	-0.0317	2.8724	3.7569
awind_ind_5m.0830	2329	141.7	140.5	0.7063	2.4671	3.9274
awind_ind_1m.0825	11545	166.0	178.6	-4.0882	7.0306	10.1471
awind_ind_1m.0828	11373	101.5	106.2	-0.2101	2.9622	3.8751
awind_ind_1m.0830	11545	140.4	139.7	0.6337	2.3704	3.7275
mwind_ind_1h.0825	217	162.1	174.0	-1.2557	6.3057	9.6064
mwind_ind_1h.0828	201	101.6	107.3	-0.6898	2.3871	3.4938
mwind_ind_1h.0830	217	138.0	136.8	0.125	1.4439	1.9605
mwind_ind_5m.0825	2329	166.4	178.7	-1.0198	6.3622	9.4587
mwind_ind_5m.0828	2217	101.7	105.6	-0.2336	2.3862	3.3116
mwind_ind_5m.0830	2329	141.8	137.4	0.9498	2.0799	2.8743
mwind_ind_1m.0825	11545	166.0	177.7	-0.6788	6.2981	9.3914
mwind_ind_1m.0828	11017	101.1	105.7	-0.3779	2.2792	3.2517
mwind_ind_1m.0830	11545	140.0	136.6	0.743	1.9787	2.6921
awind_ind_1h_sh.0825	325	162.1	175.6	-2.7155	4.1153	6.5406
awind_ind_1h_sh.0828	300	102.6	104.5	-0.0949	1.5528	2.2241
awind_ind_1h_sh.0830	325	141.5	141.3	-0.0785	1.6427	2.3778
awind_ind_5m_sh.0825	3625	166.4	179.9	-1.0475	3.9286	6.2411
awind_ind_5m_sh.0828	3550	102.4	105.4	-0.0618	1.4688	2.0437
awind_ind_5m_sh.0830	3625	142.4	142.2	-0.1354	1.6548	2.502
awind_ind_1m_sh.0825	18025	166.0	178.6	-1.0034	4.0013	6.2608
awind_ind_1m_sh.0828	17750	101.9	106.2	-0.3176	1.4425	2.0392
awind_ind_1m_sh.0830	18025	141.0	141.1	-0.1505	1.6257	2.3916
mwind_ind_1h_sh.0825	325	162.1	174.0	-2.4646	3.9385	6.1064
mwind_ind_1h_sh.0828	300	101.9	107.3	-0.782	1.5209	2.1193
mwind_ind_1h_sh.0830	325	141.1	141.3	-0.224	1.3034	1.6851
mwind_ind_5m_sh.0825	3625	166.4	178.7	-1.0628	4.012	6.134
mwind_ind_5m_sh.0828	3450	101.7	105.6	-0.3803	1.3697	1.8761
mwind_ind_5m_sh.0830	3625	142.4	143.1	-0.1763	1.4963	2.0331
mwind_ind_1m_sh.0825	18025	166.0	177.7	-0.8412	3.9665	6.0567
mwind_ind_1m_sh.0828	17200	101.2	105.7	-0.6202	1.4004	1.9443
mwind_ind_1m_sh.0830	18025	140.8	141.6	-0.355	1.4364	1.9099


August 25, Urban, 1-h output
mwind
awind
y = a + bx
mwind
a = 0.3
b = 1.037
awind
a = -1.3
b = 1.063
STOPS (ppbV)
CMAQ (ppbV)
0
30
60
90
120
150


August 25, Industrial, 1-h output
mwind
awind
y = a + bx
mwind
a = 0.3
b = 1.037
awind
a = -0.16
b = 1.02
STOPS (ppbV)
CMAQ (ppbV)
0
30
60
90
120
150
180


August 28, Urban, 1-h output
mwind
awind
y = a + bx
mwind
a = 0.7
b = 0.986
awind
a = 0.3
b = 1.003
STOPS (ppbV)
CMAQ (ppbV)
0
30
60
90
120
150


August 28, Industrial, 1-h output
mwind
awind
y = a + bx
mwind
a = 0.2
b = 1.007
awind
a = 0.7
b = 0.989
STOPS (ppbV)
CMAQ (ppbV)
30
60
90
120
150

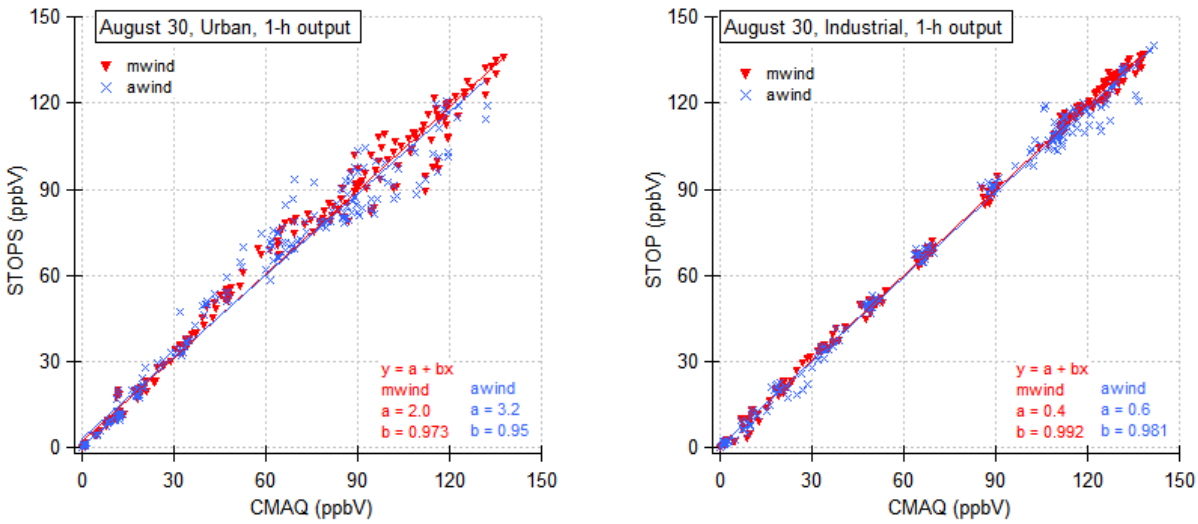


Figure 5. Comparison of ozone concentrations obtained with STOPS and CMAQ for Aug. 25, 28, and 30 for the STOPS starting position at the urban sub-domain (left figures) and the industrial sub-domain (right figures). Triangles correspond to the trajectory determined from winds in the middle column (mwind), crosses to the trajectory from average winds in the whole STOPS domain (awind). Compared are values from each cell in the first model layer, at every output time step. Note: the scale is adjusted to the maximum ozone concentration on a given day, therefore differs in each graph.

3.3 Effect of a domain size

Simulation results obtained with the STOPS system were validated against CMAQ calculated concentration fields for various STOPS domain sizes. The area of interest was always the same and consisted of nine inner cells in the STOPS domain. Therefore, by changing the STOPS domain size, the number of boundary layers around the area of interest differs.

Six different simulations with different domain sizes of 3x3, 5x5, 7x7, 9x9, 15x15, and 21x21 cells were performed. In each case the starting position was the same, with the middle column of the STOPS domain corresponding to the 21st column and 30th row in the CMAQ domain (urban sub-domain). Although the STOPS simulations were performed for the different domains, the final analysis was carried out based on the concentrations in the inner 9 cells of the first layer. Additional analysis, based on the

446 averaged concentration in the area of interest, was also performed. The averaging
447 eliminates concentration differences caused by uncertainties in the horizontal transport.
448 All simulations were carried out for August 25, 2000, for the stationary and moving
449 mode. In case of the moving mode, the STOPS trajectory was determined based on the
450 wind in the middle column. For the purpose of the CMAQ-STOPS comparison the
451 STOPS grid was shifted to coincide with the CMAQ grid.
452 Statistical parameters of the CMAQ-STOPS ozone comparison results from simulations
453 with different domain sizes are shown in Table 5 for the stationary case and in Tables 6
454 and 7 for the moving cases. It can be seen that increasing the number of boundary layers
455 around the domain of interest improves the correlation between CMAQ and stationary
456 STOPS results. In case of the moving mode, the simulations with bigger domains
457 reached the boundary of the CMAQ domain earlier than the intended simulation ending
458 time, therefore, it is not very practical.
459

Table 5. Statistical parameters of simulations with different STOPS domain sizes. In each case only 9 inner cells were taken for the analysis. The results correspond to the stationary case.

CASE	N	HMAX	SMAX	MB	MAE	RMSE	RMSE avg
3x3	117	162.1	158.5	-1.0496	1.9374	3.1827	2.4100
5x5	117	162.1	161.4	-0.9025	1.3159	2.1476	1.7210
7x7	117	162.1	159.0	-0.2914	1.0090	1.7355	1.4075
9x9	117	162.1	160.4	-0.1232	0.6343	1.2566	0.9400
15x15	117	162.1	160.8	0.0818	0.2696	0.4597	0.2346
21x21	117	162.1	162.8	-0.0315	0.2634	0.4579	0.3491

Table 6. Statistical parameters for simulations with different STOPS domain size, where only 9 inner cells were chosen for the analysis. The results correspond to the moving case, when the trajectory starting position corresponds to the 21 and 30 CMAQ column and row, respectively.

CASE	N	HMAX	SMAX	MB	MAE	RMSE	RMSE avg
3x3	117	105.4	106.4	-0.3768	1.6632	2.5934	1.7774
5x5	117	105.4	105.2	-0.2481	1.4438	2.2264	1.3617
7x7	117	105.4	105.1	-0.3131	1.4116	2.1408	1.2725
9x9	108	105.4	104.7	-0.4253	1.2482	1.8741	1.0929
15x15	99	105.4	104.3	-0.1542	1.0885	1.5237	0.6736
21x21	81	84.4	84.4	-0.3360	1.1220	1.7900	0.8787

Table 7. As above, but with different starting position corresponding to the 25 and 30 CMAQ column and row, respectively.

CASE	N	HMAX	SMAX	MB	MAE	RMSE	RMSE avg
3x3	117	143.0	138.1	-1.1138	3.2706	4.9511	3.3688
5x5	117	143.0	133.7	-0.3396	3.0431	4.7310	3.1896
7x7	117	143.0	133.4	-0.1603	2.9672	4.6991	3.2204
9x9	117	143.0	134.0	-0.0864	2.9405	4.6791	3.2066
15x15	108	143.0	134.2	-0.0661	3.0548	4.8358	3.3063
21x21	99	143.0	133.8	0.2430	3.0527	5.1374	3.7556

4. Example of application

Here, we present an example of STOPS application for a source-receptor relationship analysis. Many industrial petrochemical and chemical manufacturing facilities are located in the Houston Ship Channel. In addition to emissions associated with regular

operations, they frequently release additional, so called ‘upset emissions’ [Murphy and Allen, 2005]. Such emission releases can dominate local emissions and result in very high ozone concentrations [Zhang *et al*., 2004; Nam *et al*., 2006]. Impact of such releases can be simulated by STOPS.

We performed the base case simulations as described in Czader et al. (2008) in which we used the extended version of SAPRC-99 that explicitly represents emissions and chemistry of many individual VOCs. In addition to the base case simulation we performed STOPS re-simulations in which additional emission spike of several individual VOCs was added to STOPS one at the time, imitating ‘upset emission’ release. The additional emission was added between 12 and 13 UTC at the location of the middle cell of STOPS domain at its starting position. Figure 6 show snapshots of ozone mixing ratios in the STOPS domain on August 25, 28, and 30 of 2000 along trajectories shown in Fig. 4. The results are from the base case simulation. Figure 7 shows changes in ozone mixing ratios occurring along trajectory downwind from an emission source on August 25 that are caused by additional emissions of VOCs injected into a STOPS domain. It can be seen that different compounds affect ozone concentration to a different extent. The low reactive isobutane (I_BUTA) has a small effect on ozone, which is in contrast to *trans*-2-butene (BUTE2T) that due to its high reactivity has a potential of increasing ozone mixing ratio locally, close to the emission source, and with higher magnitude.

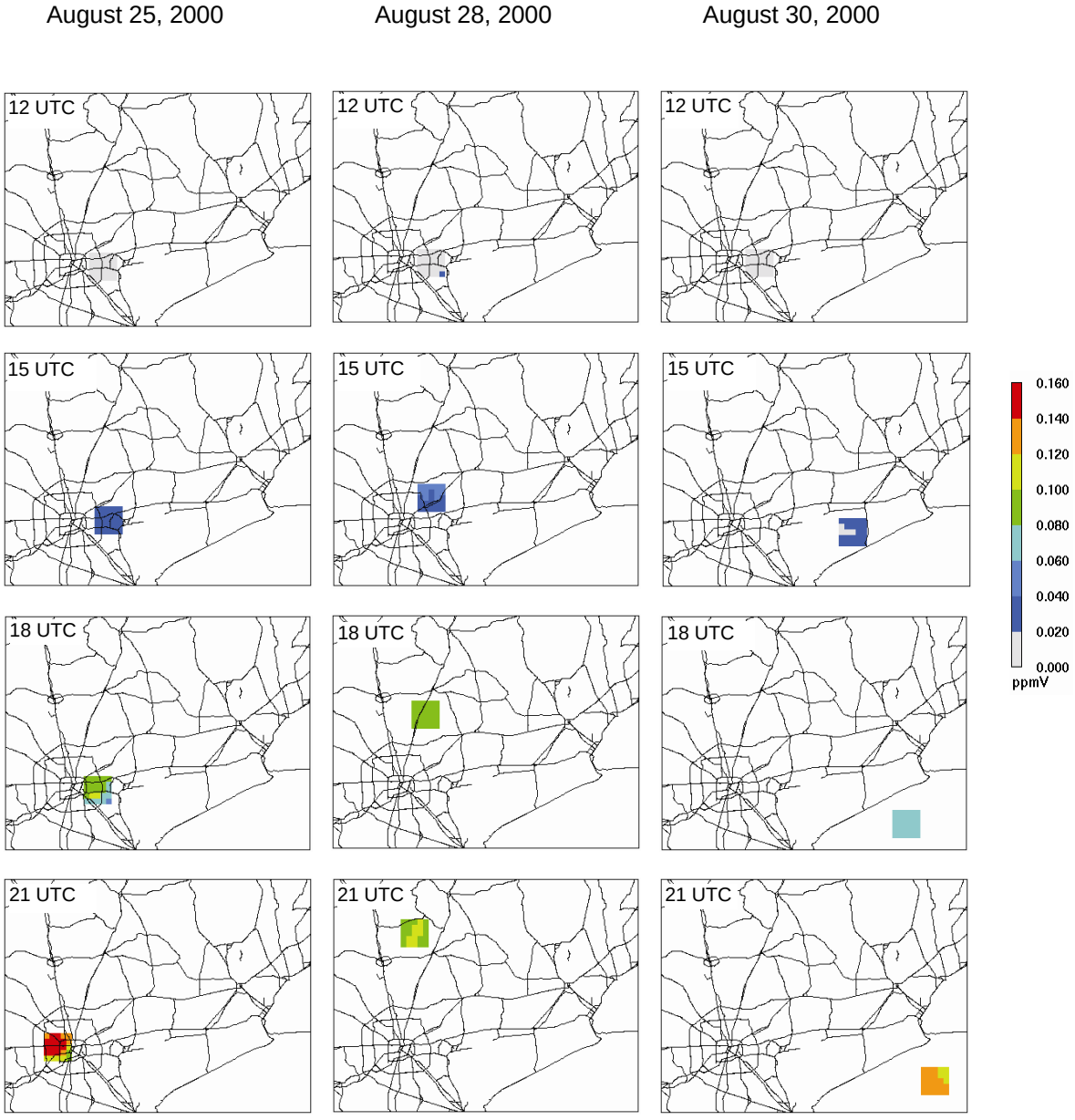
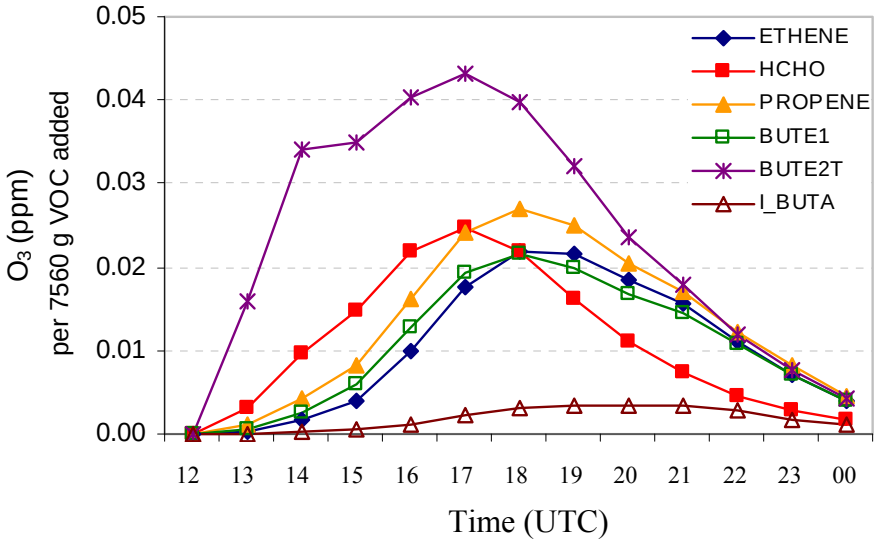


Figure 6. Snapshots of ozone concentrations along STOPS trajectories on August 25 (left), August 28 (middle), and August 30 (right) when the STOPS simulation started from the industrial sub-domain.



505
506

507 Figure 7. Changes in ozone along STOPS trajectory on August 25 due to emission spike
508 of different individual VOCs. The values are integrated in the surface layer of the
509 STOPS domain.

510

511 ## 5. Summary

512 A hybrid Lagrangian-Eulerian based modeling tool (called STOPS) was developed as a
513 computationally efficient 3-D grid sub-model for the purpose of evaluations of the
514 source-receptor relationship upon release of new emissions. It is suitable to track a
515 pollutant plume emitted in the morning then undergoing physical and chemical
516 transformation in the well-mixed convective conditions. The correctness of its
517 algorithms and the overall performance was evaluated against CMAQ simulation results
518 and it was shown that STOPS is capable of predicting ozone mixing ratios in close
519 agreement with CMAQ predictions. Its performance however depends on the trajectory
520 calculations and the atmospheric conditions occurring during the simulation period.
521 Better agreement between CMAQ-STOPS concentration pairs was found when the

STOPS trajectory was calculated based on the winds in the middle column as compared to calculation based on the value averaged in the whole STOPS domain. Under some atmospheric conditions, such as uniform winds on August 28, its performance was very satisfactory, with the mean bias for ozone mixing ratios varying between -0.03 ppbV and -0.78 ppbV and the slope between 0.99 and 1.01 for different analyzed cases. However, for complicated meteorological condition, such as on August 25 where recirculation of air occurred, its predictions deviated from CMAQ simulated values, with mean bias varying between 0.07 ppbV and -4.29 ppbV and slope varying between 0.95 and 1.06 for different analyzed cases for ozone surface mixing ratio. Averaging the surface concentration values over a STOPS domain resulted in the smaller bias between STOPS and CMAQ results. This technique is appropriate since STOPS is designed to be used for the chemical analysis rather than for the analysis of individual cells in which concentration values are strongly affected by fine uncertainties in the horizontal transport. The limitation of STOPS is due to the Lagrangian movement when applied for non-uniform winds for which the plume might be dispersed outside of STOPS domain. This is a limitation of every Lagrangian approach. The advantages of STOPS compared to Lagrangian type models is usage of realistic boundary conditions at every simulations time step as well as using detailed chemistry.

Code availability

The STOPS source code can be obtained by contacting the leading author at bczader@uh.edu

Acknowlegments

This work is dedicated to the memory of Dr. Daewon Byun (1956-2011), whose pursuit of scientific excellence as a developer of the CMAQ model continues to inspire us.

References

Appel, K., Chemel, C., Roselle, S. J., Francis, 5 X. V., Hu, R.-M., Sokhi, R. S., Rao, S. T., and Galmarini, S.: Examination of the Community Multiscale Air Quality (CMAQ) model performance over the North American and European domains, Atmos. Environ., 53, 142–155, 2012.

Arnold, J.R. and Dennis, R.L.: Testing CMAQ chemistry sensitivities in base case and emissions control runs at SEARCH and SOS99 surface sites in the southeastern US, Atmos. Environ., 40, 5027-5040, 2006.

Byun, D. and Schere, K.L.: Review of the Governing Equations, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, Appl. Mech. Rev., 59, 51-77, 2006.

Byun, D.W., Kim, S.-T., and Kim, S.-B.: Evaluation of air quality models for the simulation of a high ozone episode in the Houston metropolitan area, Atmos. Environ., 41, 837-853, 2007.

Czader, B. H., Byun, D. W., Kim, S.-T., and Carter, W. P. L.: A study of VOC reactivity in the Houston-Galveston air mixture utilizing an extended version of SAPRC-99 chemical mechanism, Atmos. Environ., 42, 5733–5742, 2008.

Eder, B. and Yu, S.: A performance evaluation of the 2004 release of Models-3 CMAQ. Atmos. Environ., 40, 4811-4824, 2006.

Henderson, B.H., Kimura, Y., McDonald-Buller, E., Allen, D.T., and Vizuete, W.: Comparison of Lagrangian process analysis tools for Eulerian air quality models, Atmos. Environ., 45, 5200-5211, 2011.

Kimura, Y., McDonald-Buller, E., Vizuete, W., and Allen, D.T.: Application of a Lagrangian process analysis tool to characterize ozone formation in Southeast Texas, Atmos. Environ., 42, 5743-5759, 2008.

Murphy, C.F. and Allen, D.T.: Hydrocarbon emissions from industrial release events in the Houston-Galveston area and their impact on ozone formation, Atmos. Environ., 39, 3785-3798, 2005.

Nam, J., Kimura, Y., Vizuete, W., Murphy, C., and Allen, D.T.: Modeling the impacts of emission events on ozone formation in Houston, Texas. Atmos. Environ., 40, 5329-5341, 2006.

Smyth, S.C., Jiang, W., Yin, S., Roth, H., and Giroux, E.: Evaluation of CMAQ O3 and PM2.5 performance using Pacific 2001 measurement data, Atmos. Environ., 40, 2735-2749, 2006.

Stein, A.F., Isakov, V., Godowitch, J., and Draxler, R.R.: A hybrid modeling approach to resolve pollutant concentrations in an urban area, Atmos. Environ., 41, 9410-9426, 2007.

Zhang, R., Lei, W., Tie, X., and Hess, P.: Industrial emissions cause extreme diurnal urban ozone variability, P. Natl. Acad. Sci. USA, 101, 6346-6350, 2004.