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

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Abstract

A hybrid Lagrangian–Eulerian 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 varies between −0.03 and −0.78 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 varying between 0.07 and −4.29 and slope varying between 0.95 and 1.063 for different analyzed cases. For historical reasons this hybrid Lagrangian – Eulerian 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.

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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 (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 columns of air. The air columns move along with the prevailing winds, so there are no advection terms in the set of governing equations. Primary pollutant emissions are injected into the columns when they 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. However, they do not account for chemical transformations as the chemistry is modeled as first order decay (pseudo second-order) of pollutants; and therefore, 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.

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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 dispersion model. 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 modeling tool was designed to re-simulate only a part of a 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 affecting pollutant mixing ratios, 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 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 (mass budget and integrated chemical reaction rates) 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

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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. A Lagrangian approaches were also developed for the purpose of detailed analysis of chemical interactions inside a plume. For example, Kimura et 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 modeling approach

A hybrid Eulerian–Lagrangian 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. 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. STOPS can be considered as a moving nest window model, where the domain moves with the mean wind speed of the target air column in which the dynamic boundary conditions are obtained from saved original CMAQ simulation results.

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In the simplest application, the STOPS domain can consist of only one cell in the horizontal direction, which corresponds to a 2-D column shown in Fig. 1. The modeling domain can be extended with a few horizontal layers of cells padding the targeted analysis domain. 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. 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. The trajectory used for moving the STOPS domain, in fact, should be viewed as the window of analysis. STOPS is essentially a moving nest CMAQ that utilizes the saved original CMAQ simulation results to provide boundary conditions, initial conditions, emissions and meteorological parameters necessary for the simulations. Use of the dynamic boundary conditions is one of the advantages of STOPS compared to Lagrangian column models.

The trajectory for STOPS movement is calculated based on the mean wind $\overline{w}_{\mathrm{PBL}}$ in the middle column (thereafter mwind) that is mass averaged up to the Planetary Boundary Layer (PBL) height according to the following equation:

$$\bar{w}_{\mathrm{PBL}} = \frac{1}{M_{\mathrm{PBL}}} \sum_{l=1}^{l=\mathrm{PBL}} w_l \cdot \Delta\sigma_l \tag{1}$$

where l is a layer number, M_{PBL} is the total mass of air column from the surface to the PBL height, and w_l is a wind in the layer l.

The total mass of air from the surface to the PBL height (M_{PBL}) is calculated as follows:

$$M_{\mathrm{PBL}} = \sum_{l=1}^{l=\mathrm{PBL}} \Delta\sigma_l \tag{2}$$

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where σ is defined as:

$$\sigma = \frac{(p - p_t)}{(p_s - p_t)} \quad (3)$$

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 concentrations.
- 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.

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– 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

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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
$$N = \mathrm{NCOL} \cdot \mathrm{NROW} \cdot \mathrm{NTSTEP} \quad (4)$$

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

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

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

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

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

Daily ozone maximum from CMAQ simulations HMAX
Daily ozone maximum from STOPS simulations SMAX

3.1 STOPS in the stationary mode

First, the correctness of the STOPS code implementation was verified by performing STOPS simulations in the static mode in which the grid cells were aligned with CMAQ grids and directly compared to CMAQ values. 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

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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 cell in each STOPS domain relatively to the CMAQ (host) grid, number of padding cells in each direction around a middle cell, and a column and row of the host domain are presented in Table 1.

Usually boundary conditions are provided at hourly intervals and modeled concentrations are saved with an hourly output time step. Because a model performs calculations at time intervals on the order of minutes the boundary values are interpolated to match a specific computation time step, which is also a case for STOPS that uses CMAQ hourly concentrations for boundary conditions. For the comparison of STOPS results we used CMAQ concentrations from the corresponding grid cells. These gird cells are not at the domain boundaries but inside domain; therefore, to calculate advection CMAQ uses values from adjacent cells every synchronization time step as opposed to STOPS hourly values. Because of that, we expect some differences between STOPS and CMAQ calculated mixing ratios and to justify them, CMAQ and STOPS simulations were performed for different output time steps, which were set to 1 h, 5 min, and 1 min, which is close to the synchronization time step.

Three sample days out of the TexAQS 2000 episode were chosen for simulations: 25, 28, and 30 August. For all cases the STOPS simulation started at 12:00 UTC and lasted 12 h. 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 min 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 h output time step (left) and 1 min time step (right)

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with less scattering from 1 min output time step, confirming that shortening the output time step makes STOPS results closer to CMAQ.

3.2 STOPS in the moving mode

The next step in the STOPS verification was to analyze uncertainties related to the movement of a nest domain. A direct comparison between CMAQ and STOPS result was complicated due to the fact that STOPS grids do not necessarily align with CMAQ grid. In order to overcome this problem, for the comparison purpose either the STOPS domain had to be aligned with the CMAQ grid (shifted) or STOPS values from several cells have to be interpolated to the corresponding CMAQ cell. The performance evaluation was tested for these two possibilities.

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 Fig. 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. 25 August 2000 was the day with complicated, circular wind patterns; on 28 August 2000 strong, but uniform southerly winds were observed, and on 30 August 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 Fig. 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 12:00 UTC and end the next day at 00:00 UTC, except trajectories on 28 August that ended at 23:00 UTC due to subdomain reaching the boundaries of CMAQ domain earlier as an effect of strong winds on that day. On 28 and 30 August there are little differences in trajectories determined by the two different

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methods. However, as can be seen from Fig. 4b, there are differences in trajectories for 25 August, especially during the first couple of hours of simulations. Both trajectories move south between hour 12:00 and 13:00 UTC. After that, the trajectory determined by the winds in the middle column moves east until 15:00 UTC and then west, making a circular pattern; at 17:00 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.

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 h, the simulation was performed for 25 August, 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 STOPS concentrations were spatially interpolated to be compared with CMAQ concentrations. 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 25, 28, and 30 August for the STOPS starting position at the urban sub-domain (left graphs) and industrial sub-domain (right graphs). Triangles correspond to STOPS simulations when the trajectory was determined based on the winds in the middle column (mwind), crosses to

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the trajectory obtained from the average winds in the whole STOPS domain (awind). Plotted are concentrations from all cells in the first model layer, at every output time step. Very good performance was found on 28 August with the averaged mean absolute error of 1.3 and 1.5 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 STOPS values were interpolated.

3.2.1 Effect of a domain size on the STOPS performance

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 3×3, 5×5, 7×7, 9×9, 15×15, and 21×21 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 averaged concentration in the area of interest, was also performed. The averaging eliminates concentration differences caused by uncertainties in the horizontal transport. All simulations were carried out for 25 August 2000, for the stationary and moving mode. In case of the moving mode, the STOPS trajectory was determined based on the wind in the middle column. For the purpose of the CMAQ-STOPS comparison the STOPS grid was shifted to coincide with the CMAQ grid.

Statistical parameters of the CMAQ-STOPS ozone comparison results from simulations with different domain sizes are shown in 5 for the stationary case and in 6 and

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7 for the moving cases. It can be seen that increasing the number of boundary layers around the domain of interest improves the correlation between CMAQ and stationary STOPS results. In case of the moving mode, the simulations with bigger domains reached the boundary of the CMAQ domain earlier than the intended simulation ending time, therefore, it is not very practical.

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. Figure 6 show snapshots of ozone mixing ratios in the STOPS domain on 25, 28, and 30 August of 2000 along trajectories shown in Fig. 4. The results are from the base case simulation. Figure 7 shows changes in ozone occurring along trajectory downwind from emission source on 25 August that are caused by additional emissions of VOCs. 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 the ozone mixing ratio locally, close to the emission source, and with higher magnitude.

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5 Summary

A hybrid Lagrangian–Eulerian modeling tool (called STOPS) was developed as a computationally efficient 3-D grid sub-model for the purpose of evaluations of the source-receptor relationship upon release of new emissions. It is suitable to track a pollutant plume emitted in the morning then undergoing physical and chemical transformation in the well-mixed convective conditions. The correctness of its algorithms and the overall performance was evaluated against CMAQ simulation results. STOPS performance depends on the trajectory calculations and the atmospheric conditions occurring during the simulation period. 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 28 August, its performance was very satisfactory, with the mean bias for ozone mixing ratios varying between −0.03 and −0.78 and the slope between 0.99 and 1.01 for different analyzed cases. However, for complicated meteorological condition, such as on 25 August where recirculation of air occurred, its predictions deviated from CMAQ simulated values, with mean bias varying between 0.07 and −4.29 and slope varying between 0.95 and 1.063 for different analyzed cases. 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.

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Table 1. 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

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Table 2. Summary of statistical parameters for STOPS-CMAQ concentration pairs, when STOPS was used in the stationary mode (the values of MAXD and MIND are given in ppbv).

NAME	*N*	HMAX	SMAX	MB	MAE	RMSE
stat_1h.0825	5733	162.1	162.9	−0.1894	0.3822	0.6820
stat_1h.0828	5733	115.6	115.8	−0.1160	0.1979	0.3229
stat_1h.0830	5733	158.7	158.7	−0.3089	0.3870	0.5920
stat_5m.0825	63 945	166.4	167.1	−0.1183	0.2067	0.3946
stat_5m.0828	63 945	116.0	115.7	0.0369	0.1213	0.2075
stat_5m.0830	63 945	160.3	160.5	0.0167	0.1297	0.2295
stat_1m.0825	317 961	166.0	166.0	0.0140	0.0456	0.0906
stat_1m.0828	317 961	115.1	115.1	−0.0117	0.0365	0.0744
stat_1m.0830	317 961	158.9	158.9	−0.0138	0.0308	0.0715
stat_1h.0825	325	108.7	113.9	−0.8562	1.0007	1.4691
stat_1h.0828	325	88.5	88.0	−0.7096	0.8004	1.1424
stat_1h.0830	325	145.1	147.8	−1.8936	1.9774	2.6690
stat_5m.0825	3625	111.6	112.8	−0.5794	0.6502	0.9494
stat_5m.0828	3625	88.6	87.7	−0.2883	0.4229	0.6003
stat_5m.0830	3625	148.2	148.4	−0.4536	0.5636	0.7370
stat_1m.0825	18 025	112.0	112.6	−0.1275	0.2107	0.3356
stat_1m.0828	18 025	86.6	86.6	−0.0724	0.1045	0.1426
stat_1m.0830	18 025	146.6	146.7	−0.0974	0.1342	0.2249
stat_1h.0825	325	162.1	161.4	−0.9287	1.3587	2.1596
stat_1h.0828	325	69.2	70.7	−0.5708	0.6402	0.9812
stat_1h.0830	325	145.9	148.0	−1.5667	1.5673	1.9527
stat_5m.0825	3625	165.9	167.1	−0.5115	0.6070	0.9891
stat_5m.0828	3625	70.5	71.0	−0.2271	0.3825	0.6278
stat_5m.0830	3625	145.9	146.8	−0.3074	0.3411	0.4611
stat_1m.0825	18 025	165.4	165.8	0.0214	0.2073	0.3132
stat_1m.0828	18 025	69.9	69.7	−0.0300	0.0875	0.1292
stat_1m.0830	18 025	144.3	144.7	−0.1970	0.2114	0.3607

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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 (the values of MAXD and MIND are given in ppb).

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	11 545	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	11 545	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	11 545	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	11 545	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	18 025	110.7	107.3	0.0743	1.3337	1.9913
awind_urb_1m_sh.0828	14 750	103.6	109.2	−0.0619	1.3074	2.2298
awind_urb_1m_sh.0830	18 025	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	18 025	109.2	106.5	−0.1237	1.3359	1.9914
mwind_urb_1m_sh.0828	14 250	103.0	111.2	0.1064	1.2086	2.0841
mwind_urb_1m_sh.0830	18 025	138.4	138.5	−0.4413	2.4165	3.5173

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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 (the values of MAXD and MIND are given in ppb).

NAME	*N*	HMAX	SMAX	MB	MAE	RMSE
awind_urb_1h.0825	217	162.1	175.6	−3.7049	6.667	9.7334
awind_urb_1h.0828	201	102.0	104.5	−0.0743	2.7724	3.6884
awind_urb_1h.0830	217	141.4	140.1	0.5727	2.2085	3.4874
awind_urb_5m.0825	2329	166.2	179.9	−4.2896	6.9033	10.246
awind_urb_5m.0828	2281	102.0	105.4	−0.0317	2.8724	3.7569
awind_urb_5m.0830	2329	141.7	140.5	0.7063	2.4671	3.9274
awind_urb_1m.0825	11 545	166.0	178.6	−4.0882	7.0306	10.1471
awind_urb_1m.0828	11 373	101.5	106.2	−0.2101	2.9622	3.8751
awind_urb_1m.0830	11 545	140.4	139.7	0.6337	2.3704	3.7275
mwind_urb_1h.0825	217	162.1	174.0	−1.2557	6.3057	9.6064
mwind_urb_1h.0828	201	101.6	107.3	−0.6898	2.3871	3.4938
mwind_urb_1h.0830	217	138.0	136.8	0.125	1.4439	1.9605
mwind_urb_5m.0825	2329	166.4	178.7	−1.0198	6.3622	9.4587
mwind_urb_5m.0828	2217	101.7	105.6	−0.2336	2.3862	3.3116
mwind_urb_5m.0830	2329	141.8	137.4	0.9498	2.0799	2.8743
mwind_urb_1m.0825	11 545	166.0	177.7	−0.6788	6.2981	9.3914
mwind_urb_1m.0828	11 017	101.1	105.7	−0.3779	2.2792	3.2517
mwind_urb_1m.0830	11545	140.0	136.6	0.743	1.9787	2.6921
awind_urb_1h_sh.0825	325	162.1	175.6	−2.7155	4.1153	6.5406
awind_urb_1h_sh.0828	300	102.6	104.5	−0.0949	1.5528	2.2241
awind_urb_1h_sh.0830	325	141.5	141.3	−0.0785	1.6427	2.3778
awind_urb_5m_sh.0825	3625	166.4	179.9	−1.0475	3.9286	6.2411
awind_urb_5m_sh.0828	3550	102.4	105.4	−0.0618	1.4688	2.0437
awind_urb_5m_sh.0830	3625	142.4	142.2	−0.1354	1.6548	2.502
awind_urb_1m_sh.0825	18 025	166.0	178.6	−1.0034	4.0013	6.2608
awind_urb_1m_sh.0828	17 750	101.9	106.2	−0.3176	1.4425	2.0392
awind_urb_1m_sh.0830	18 025	141.0	141.1	−0.1505	1.6257	2.3916
mwind_urb_1h_sh.0825	325	162.1	174.0	−2.4646	3.9385	6.1064
mwind_urb_1h_sh.0828	300	101.9	107.3	−0.782	1.5209	2.1193
mwind_urb_1h_sh.0830	325	141.1	141.3	−0.224	1.3034	1.6851
mwind_urb_5m_sh.0825	3625	166.4	178.7	−1.0628	4.012	6.134
mwind_urb_5m_sh.0828	3450	101.7	105.6	−0.3803	1.3697	1.8761
mwind_urb_5m_sh.0830	3625	142.4	143.1	−0.1763	1.4963	2.0331
mwind_urb_1m_sh.0825	18 025	166.0	177.7	−0.8412	3.9665	6.0567
mwind_urb_1m_sh.0828	17 200	101.2	105.7	−0.6202	1.4004	1.9443
mwind_urb_1m_sh.0830	18 025	140.8	141.6	−0.355	1.4364	1.9099

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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
3 × 3	117	162.1	158.5	−1.0496	1.9374	3.1827	2.4100
5 × 5	117	162.1	161.4	−0.9025	1.3159	2.1476	1.7210
7 × 7	117	162.1	159.0	−0.2914	1.0090	1.7355	1.4075
9 × 9	117	162.1	160.4	−0.1232	0.6343	1.2566	0.9400
15 × 15	117	162.1	160.8	0.0818	0.2696	0.4597	0.2346
21 × 21	117	162.1	162.8	−0.0315	0.2634	0.4579	0.3491

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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
3 × 3	117	105.4	106.4	−0.3768	1.6632	2.5934	1.7774
5 × 5	117	105.4	105.2	−0.2481	1.4438	2.2264	1.3617
7 × 7	117	105.4	105.1	−0.3131	1.4116	2.1408	1.2725
9 × 9	108	105.4	104.7	−0.4253	1.2482	1.8741	1.0929
15 × 15	99	105.4	104.3	−0.1542	1.0885	1.5237	0.6736
21 × 21	81	84.4	84.4	−0.3360	1.1220	1.7900	0.8787

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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
3 × 3	117	143.0	138.1	−1.1138	3.2706	4.9511	3.3688
5 × 5	117	143.0	133.7	−0.3396	3.0431	4.7310	3.1896
7 × 7	117	143.0	133.4	−0.1603	2.9672	4.6991	3.2204
9 × 9	117	143.0	134.0	−0.0864	2.9405	4.6791	3.2066
15 × 15	108	143.0	134.2	−0.0661	3.0548	4.8358	3.3063
21 × 21	99	143.0	133.8	0.2430	3.0527	5.1374	3.7556

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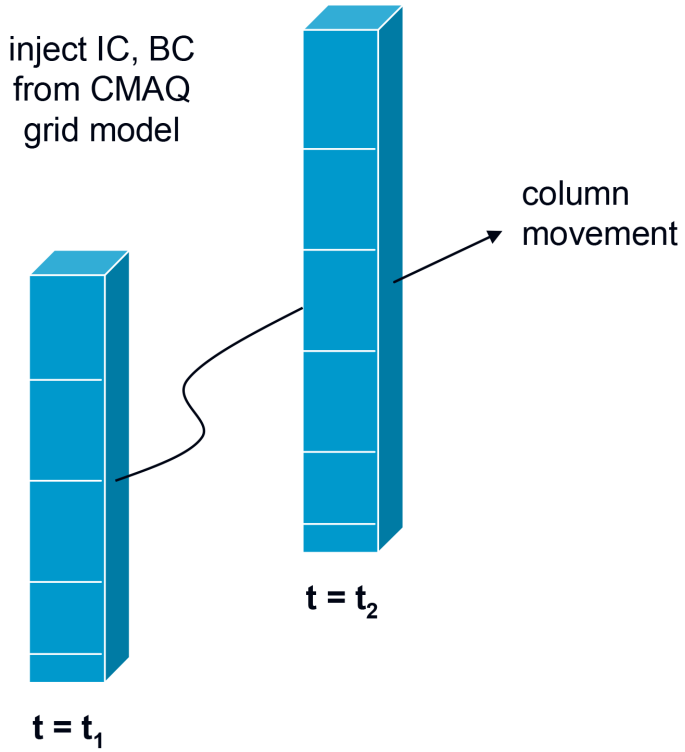

Figure 1. The conceptual model for STOPS trajectory movement.

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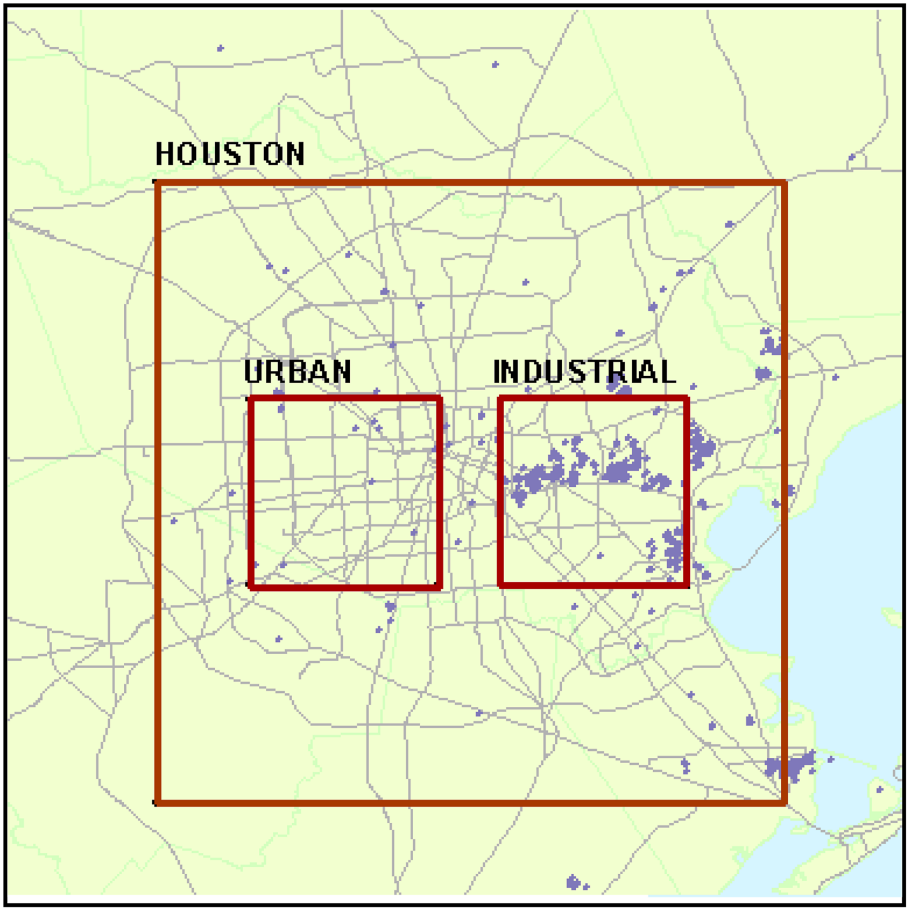

Figure 2. Starting locations of STOPS domains. Points indicate location of emission point sources in Houston.

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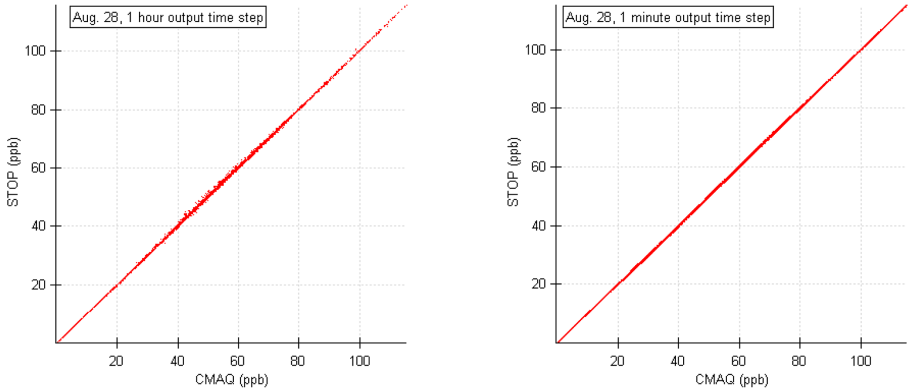

Figure 3. Comparison of CMAQ and static STOPS simulation results for 28 August for 1 h (left) and 1 min (right) output time step. Both graphs correspond to simulation from the Houston domain.

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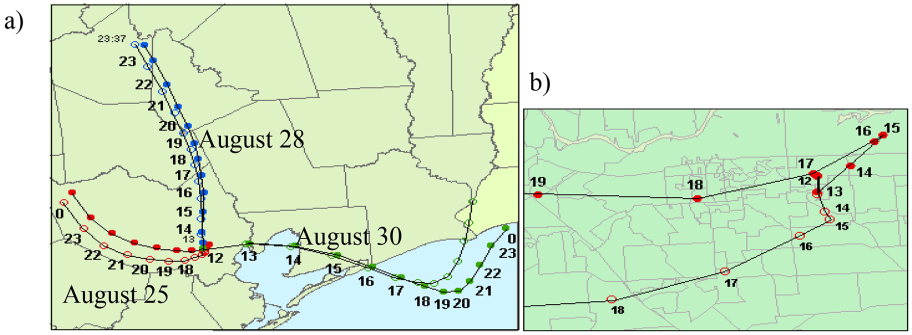

Figure 4. (a) STOPS trajectories starting from the industrial sub-domain. Trajectories determined based on the winds in the STOPS middle column are indicated by filled circles, and those determined based on the average winds in the whole STOPS domain with open circles. Trajectories for 25 August are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

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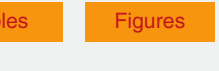

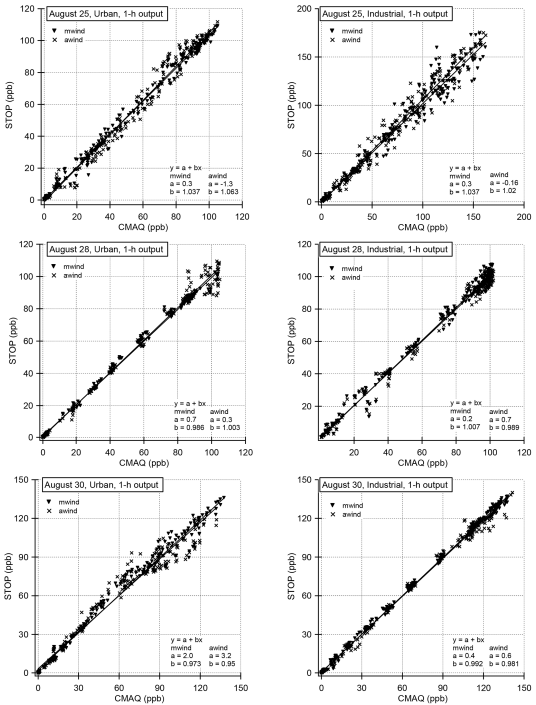

Figure 5. Comparison of ozone concentrations obtained with STOPS and CMAQ for 25, 28, and 30 August for the STOPS starting position at the urban sub-domain (left figures) and the industrial sub-domain (right figures). Triangles correspond to the trajectory determined from winds in the middle column (mwind), 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.

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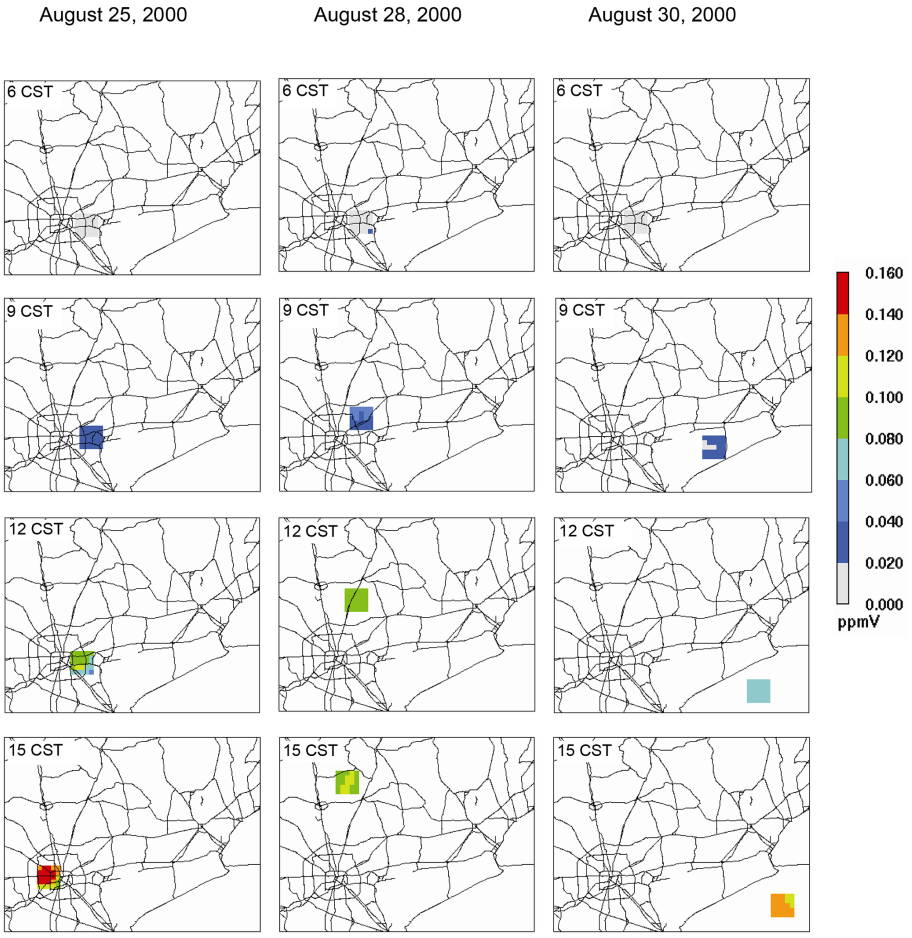

Figure 6. Snapshots of ozone concentrations along STOPS trajectories on 25 August (left), 28 August (middle), and 30 August (right) when the STOPS simulation started from the industrial sub-domain.

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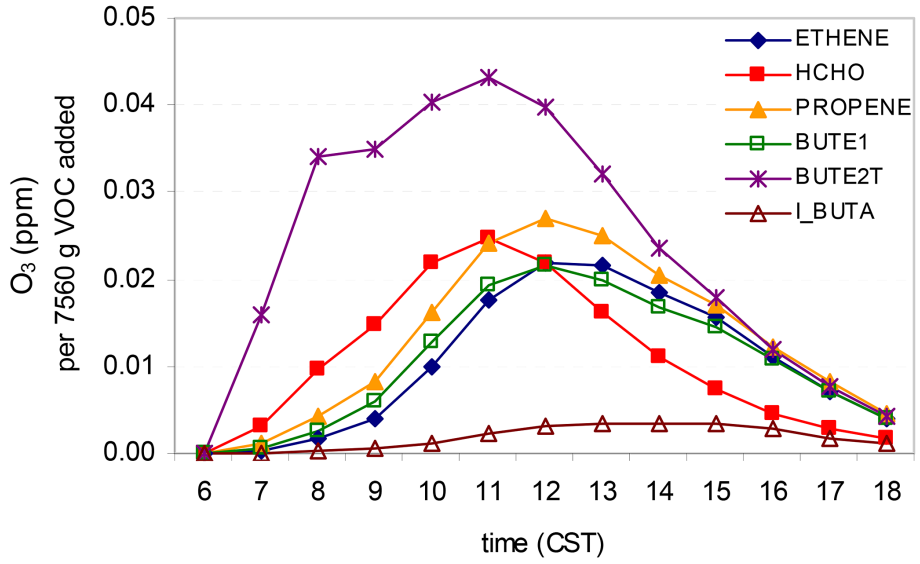

Figure 7. Changes in ozone along STOPS trajectory on 25 August due to emission spike of different individual VOCs. The values are integrated in the surface layer of the STOPS domain.

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