Response to the Executive Editor of GMD

Dear authors,

In my role as Executive editor of GMD, I would like to bring to your attention our Editorial: http://www.geoscientific-model-development.net/gmd_journal_white_paper.pdf http://www.geosci-model-dev.net/6/1233/2013/gmd-6-1233-2013.html *This highlights some requirements of papers published in GMD, which is also available on the GMD website in the 'Manuscript Types' section:* http://www.geoscientific-model-development.net/submission/manuscript_types.html

In particular, please note that for your paper, the following requirements have not been met in the Discussions paper – please correct this in your revised submission to GMD. "– The paper must be accompanied by the code, or means of accessing the code, for the purpose of peer-review. If the code is normally distributed in a way which could compromise the anonymity of the referees, then the code must be made available to the editor. The referee/editor is not required to review the code in any way, but they may do so if they so wish. "

"– All papers must include a section at the end of the paper entitled "Code availability". In this section, instructions for obtaining the code (e.g. from a supplement, or from a website) should be included; alternatively, contact information should be given where the code can be obtained on request, or the reasons why the code is not available should be clearly stated. "

Yours,

Dan Lunt

Dear Dan Lunt,

In response to your comments I would like to inform you that that I have sent the STOPS package, which include the source code along with run scripts, to the editor. I will also include it in the revised submission of the manuscript to GMD. Also, I will add to the manuscript the required section on code availability, in which the reader will be informed that the STOPS source code can be obtained by contacting the leading author at bczader@uh.edu

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Response to the Referee #1

Dear Reviewer,

Thank you for your time and effort put into reviewing the paper. Please find below our responses to your comments.

This publication is timely and well done. The STOPS system could be an important tool for scientists policy makers, and consultants alike. The tool uses a moving CMAQ simulation that dynamically interfaces with archived CMAQ simulations. The tool is well described and the basic performance is well described for the no emissions modification case. I would have liked to see an evaluation of the response to additional emissions, which will stress the boundary assumptions further. I look forward to more application papers (e.g., chemistry updates that would influence boundaries, other emission additions).

--- We evaluated STOPS against CMAQ results for the base case showing that it is capable of predicting mixing ratios in close agreement with CMAQ predictions. It is not possible to evaluate STOPS for the countless possibilities of emission perturbations as the response depend on the choice of emitted species, strength of the perturbation and also, since STOPS accounts for horizontal transport through domain boundaries and some material would be transported outside domain, it also depends on domain size. Because of the latest reason we did not use 1x1 grid domain as it is more likely to quickly lose the effect from a perturbation in the domain.

--- We do plan to work on STOPS applications and hopefully a paper would result from that.

The model description section is clear and detailed. The author first introduces the two basic approaches which air pollution models are based on: Eulerian and La-grangian. The author then points out the limitations of modeling with either approach exclusively. The nested-moving approach in STOPS is described as a useful hybrid Eulerian–Lagrangian modeling approach. This paper provides sufficient description of the modifications to CMAQ. Finally, I would not call this Lagrangian. STOPS is actually a series of Eulerian models strung together at the computational time-step. It is more of a pseudo or quasi-Lagrangian approach.

--- We used the term “Lagrangian” because of STOPS movement with a local flow. 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 (we added this statement in lines 129-132) and we think it is valid to use the term Lagrangian for descriptive purposes . To indicate that it is not exactly Lagrangian tool we will replace the wording “Lagrangian-Eulerian tool” with "Lagrangian-Eulerian based tool" or "Lagrangian-Eulerian approach".

The tables used in the paper are not clear and need improvement. In all tables, what are MAXD and MIND?

--- The MAXD and MIND will be removed from the caption of tables 2, 3, and 4 as they are not shown in tables.

In Table 2, there are three sets of results with identical "NAME" values. I assume this is related to the domain, but the table is unclear.

--- The first set corresponds to results from static simulations for Houston domain (please see figure 2 and table 1 for domains locations and sizes), the second set for industrial domain, and the third for urban domain. Indeed the naming in the table does not show that, we will correct the names to make it clear.

In Table 4, the domain was starting in the industrial domain, but the nomenclature is identical to Table 3 that started in the urban (urb) domain. Why is that appropriate?

--- This is a mistake, thank you for pointing it out; all names in table 4 should have 'ind' instead of 'urb'. We will correct that.

Tables 5, 6 and 7 are referenced by number without the word "table".

--- We will add the word table into a text where the tables are referenced.

Minor comments: - Abstract, add units to the bias in the abstract.

--- We will add the units of ppbV.

Page 7631, why not include a 1x1 simulation?

--- 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. We added this information in manuscript in lines 317-319.

Figure 1, Conceptual model should include multiple columns to be consistent with implementation?

--- We will modify the figure to include 3x3 columns in the conceptual model.

Make it clear that you are comparing instantaneous concentrations (not time interval averaged).

--- We will add the following at the end of section 3 on page 7627 (please see lines 283-4): “where H_i and S_i corresponds to instantaneous mixing ratios obtained with CMAQ and STOPS, respectively.”

Overall, this is a good manuscript that needs minor improvements. More discussion of the differences, or potential for differences, between CMAQ and STOPS with emission modifications would improve the manuscript. Table clarifications are necessary before publication.

--- The evaluation of STOPS against CMAQ results shown that STOPS is capable of predicting mixing ratios in close agreement with CMAQ predictions. As already mentioned, there are endless possibilities for emission modifications and it is not practical to evaluate them here. The scope of the paper was presentation of the model and its evaluation, the emission modification in section 4 shows just a potential application.

--- We will modify the tables according to reviewer suggestions.

Response to Referee #2

Dear Reviewer,

Thank you for your time and effort put into reviewing the paper and for your helpful suggestions. Please find below our response to your comments.

Major comments:

1. More detailed description of STOPS

I recommend a more detailed description of the STOPS model. A work flow could be helpful. This description should include:

--- STOPS is based on the CMAQ model and the structure and science processed are the same as in CMAQ as mentioned in line 10-11 of the abstract and lines 7-10 on page 7624. Since CMAQ is well documented we did not see a need of repeating its description, instead we provided a reference to Byun and Schere 2006 in the manuscript (please see also the following link for additional CMAQ documentation https://www.cmascenter.org/help/documentation.cfm). Therefore, in the manuscript we provided only information on modifications of CMAQ code related to STOPS structure and movement of its domain.

• When are which boundary conditions taken for STOPS. Is there only temporal or also some horizontal interpolation?

--- In the original CMAQ there is temporal interpolation of boundary conditions; this is, hourly boundary values are interpolated to match smaller calculation time steps. In STOPS, in addition to this temporal interpolation, we also added spatial interpolation. It was needed for cases when the STOPS grids do not align with the grid cells of a CMAQ file used for boundary conditions. We added this information in lines 165-173.

• The transport is described in the text. If a column is taken. Is it one box or still divided into several cells?

--- We keep the same vertical structure as in CMAQ, so the column(s) are divided into many vertical layers. Please also see a modified figure 1 and description in lines 19 and 145-155.

• What other processes are calculated? Chemistry? Convenction? Depositon? Rainout? Lightning? ..... How is this done in detail?

--- For each grid cell in a domain the following processes are calculated: 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. We added this information in lines 149-152. Please also refer to Byun and Schere (2006) referenced in the manuscript as well as the following documentation of CMAQ https://www.cmascenter.org/help/documentation.cfm.

• Are the processes calculated in the column or flux changes taken from CMAQ?

--- The processes are calculated for each grid cell inside STOPS domain. In and out-flow of pollutants between grid cells as well as at the STOPS domain boundaries is accounted for.

• Is there a difference between the output time and the calculation timestep? How is this organised?

--- Usually outputs are saved as hourly values but a user has an option to change that. 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. We added this information in lines 165-173.

2. Units and equations

Please add units to all variabes. It makes it easier to understand. If the word "mass" is used I would expect something like a unit "kg". But here it seems not to have a unit at all. Some variables, like delta sigma are not explained properly and some equations have little errors.

--- We will modify the manuscript as follow:

Page 7622 lines 23-25 was: "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."

Will be (see lines 99-102 in the below manuscript): "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 evolution of plumes."

Text from page 2624 line 16 to page 2425 line 4 will be replaced with the following and eq. 2 will be removed (see lines 198 – 223):

"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: (eq 3).

3. Chosen performance metrics

I know that these metrics are often used. It would be helpful to first clarify for what purpose they can be used. What is the scientific question? And which metric is answering this question best? E.g. total ozone for maintaining the mass budget. Or difference in ozone mixing ratios for ... ? There are some issues if the mass or volume in the individual CMAQ cells are differing, e.g. in the vertical. That should be clarified in more detail. MAX and MIN is used in the text but is missing in the description.

--- Our goal was to evaluate STOPS performance for surface ozone mixing ratios (ppbV). The mean absolute error (MAE) would be the most rigorous metric for this purpose but the mean bias is useful too as it shows if model under or overpredicts values. We will remove equation 5 and 6 since we realized that the mean values are not shown in the manuscript and add units.

--- The MAXD and MIND will be removed from the caption of tables 2, 3, and 4 as they are not shown in tables.

4. Concept of verification.

The chosen stepwise verification seems to be valid, but hard to judge, since I couln'd follow in detail. Please include a subsection explaining the procedure, motivation, and interpretation of the experimental set-up in more detail. (see also questions below). The fact that there is only one subsection in 3.2 might indicate that there are some more thoughts on the structure necessary.

--- The following will be added before section 3.1 (lines 295-300): "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."

--- Section 3.1 will be renamed to: "Effect of boundary conditions". Section 3.2: "Uncertainties related to movement of STOPS". Section 3.2.1 will be changed to 3.3: "Effect of domain size".

Some more general comments:

• Please explain rows and columns in CMAQ. And make a difference between the CMAQ column and STOPS vertical column. This is confusing.

--- We will replace the sentences on page 2623 lines 24 through page 7624 line 5 with a new description. Please see lines 145-173 in the modified manuscript.

• Title: "Development" -> " Description"? (also section name)

--- The whole manuscript is a description of STOPS and its evaluation, in the title we wanted to underline that we both developed and evaluated this model.

• The authors sometimes use ppb and ppbV. Is there a difference? Is it mass versus volume mixing ratio? Please use it uniformly.

--- We will modify the text to uniformly use unit “ppbV” which indicates volume mixing ratio.

• Similar with CST and UTC. The use is correct, but it makes it harder to read. Please use one time only.

--- We will modify the manuscript to consistently use UTC.

More detailed comments:

l 17: Why "columns of air.", I rather would say air parcels. Most are only points in the atmosphere, but some actually have an extension.

--- We will replace “columns” to “parcels” in the description of Lagrangian approach in the Introduction.

l19 : "so there are no advection terms in the set of governing equations.". Please rephrase. It sounds like there is no advection. Of course the air parces are advected.

--- What we meant is that there is only parcel movement with wind but there is no advection in and out of the column, a process that is considered in Eulerian models. We will modify it as follow (see lines 58-61):

“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.”

l23-28: Please rephrase, since there are Lagrangian chemistry models!

--- We will rephrase as follow: “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.” (See lines 67-70)

p7622 l8 "... tool to study a source-receptor relationship". Still there are diffusion processes, turbulence ets, which is normally not resolved, but which leads to a inter parcel exchange. This normally limits the efficiency of source-receptor relationships. This might come later, but should somehow be mentioned here. On top, there are other methods to resolve source receptor relationships, such as tagging. That should be mentioned somehow. Lagrangian methods are not the only one. Probably a combination would be most favourable?

--- While tagging could be used in source-receptor applications it increases number of model species and therefore computation time. It also does not give information about response to emission changes. Our goal was to get a fast tool that simulates effects of perturbation in emissions and STOPS is just one approach that we thought might be useful for that purpose.

p7623 l 6 Delete "A"

--- we will delete it.

p7624 l 8 "physical and chemical processes in STOPS are the same as in the full domain CMAQ model" Please clarify: Are the same subroutines used, i.e. there is a call of the e.g. chemistry package from STOPS or are chemical changes from the CMAQ simulation extracted and applied to the column in STOPS?

--- You could think of STOPS as being CMAQ with a much smaller domain that moves with wind. As STOPS is based on CMAQ’s code, the same subroutines are used and the same process included. We modified only several subroutines to account for movement of a domain and added a new module as described in the manuscript.

l 19 Correct "l=PBL" -> "PBL" (in all equations!)

--- It will be corrected.

l 20ff add units to the variables. Every sentence is a pragraph that is irritating. Clarify pl (midlevel pressure?). Explain why the mean wind has units m/s/kg. (sigma is a number; w in m/s?; and M in kg?) In eq. (2) M is defined as a unitless number, namly the ratio of the PBL-mass to the total model atmosphere mass uin the respective column. Please revise either the equations or the naming. A mass shoulds have kg as a unit. Explain delta sigma. Difference of what?

--- The idea was to calculate average wind based on wind values in each layer and weight it by mass of air in each layer. We approximated mass by calculating differences in pressure which in sigma coordinate system could be calculated from differences in sigma layers. We will remove eq. 2, modify equations 1 as well as rephrase the text as follow (see lines 198-223) : "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: (eq 3).

Section 3:

1. *Please clarify the Si - Hi matching: It is unclear to me how the matching of Hi and Si is done, e.g. to calculate the mean bias in eq. (7). Background: The column in STOPS is moving. Hence after a couple of timesteps the column is covering in the horizontal fractions of different CMAQ grid cells. Which of these gird cells is taken? Or is a wheighted mean calculated? See e.g. also Grewe et al., 2014 (Meteorologische Zeitschrift) for problems in Eulerian-Lagrangian cell matching.*

--- Explanation will be added on page 7627 just before section 3.1 (lines 291-3): "For cases when STOPS grids do not align with CMAQ grids the CMAQ values from grid cell corresponding to a STOPS cell are interpolated by means of weighted averaging of CMAQ values."

2. Please clarify units of Si Hi

--- We will add the following at the end of section 3 on page 7627 (lines 283-4): "where H_i and S_i corresponds to instantaneous mixing ratios (ppbV) obtained with CMAQ and STOPS, respectively."

3. *Please motivate more the use of the performance metrics. Clarify why the difference of (i guess) ppbvs is taken in eq. (7)? For example, if the mass or volume of the individual cells in the column is differs largely, then one cell might dominate the total mass in the column. The mass of a specied simulated with STOPS and CMAQ might be basically equal, but the performance metrics might show a large discrepancy because it is dominated by cells with low mass.*

--- Our goal was to evaluate STOPS performance for surface ozone, so the statistics is calculated for ozone mixing ratios in ppbV. The mean bias is the most widely used in atmospheric model evaluations

therefore it is useful for comparing our results with other studies, also it provides information on model under or over predicts values. MAE and RMSE are the more rigorous metrics that we thought would be useful to show.

4. *Explain what stationary mode means. What is stationary? windfield? chemistry? zero wind?*

--- When we use “stationary” we refer to a domain that is not moving. To clarify this we will modify the first sentence in that paragraph (p. 7627 lines 14-16) as follow (see lines 302-306):”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.”

5. *The naming "output time step" is confusing. Is this the time step of STOPS? or is STOPs run with shorter time steps and just the output of STOPS is at a different intervall?*

--- The calculation time step is in order of minutes. The output time step is usually 1 hour. We will rephrase the paragraph on page 7628 (lines 7 – 18) to provide clarification on that. Please see lines 331-349.

6. Throughout the text "interpolation" is used. I first thought a horizontal interpolation is meant to match locations. Please clarify.

--- Temporal interpolation of hourly input values is performed for most of the input data, including boundary conditions, in CMAQ as well as STOPS. In addition, for the case when STOPS domain travels; thus, may not necessarily align with the grid cell of input data, we also perform spatial interpolation. Please see modified lines 165-173.

p 7627 l 16/17 "of either initial or boundary values", but what is with the Hi values?

--- The following will be added at the end of section 3 on page 7627: “where H_i and S_i corresponds to instantaneous mixing ratios (ppbV) obtained with CMAQ and STOPS, respectively.”

p 7628 l12 Please define what is meant by "from the corresponding grid cells".

--- Here STOPS was just like a sub-domain of CMAQ, so its grid is located inside CMAQ grid and aligns with CMAQ grid cell. We were comparing values from STOPS grids cells with the corresponding grid cells in CMAQ. We will rephrase this sentence to make it clear. Please also refer to a modified figure 1 and modified explanation in lines 145-173 and 302-306 and 331-349.

Section 3.2

Here a couple of things are now explained, which partly clarify some of the questions above. Please give this information earlier. (stationary/alignment of cells/ ...)

--- As mentioned above we will provide some additional information in lines 145-173, 291-300.

p 7629 l8-9. How is the interpolation done?

--- We will rephrase it as follow (please see lines 381-389): “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.”

p 7630 l 21 "STOPS concentrations were spatially ..." I would have thought that the CMAQ values have to be interpolated (or redistributed?). How many STOPS values are available? Where are they located?

---You are right, that was a mistake and we will rephrase it as follow (lines 385-7): “we performed spatial interpolation by calculating weighted average from several CMAQ grid cells that overlap with the STOPS grid cell.”
--- Number of STOPS values depends on the size of its domain; since we compared only surface values there were N values as specified in equation 4, where NCOL and NROW is number of columns and rows in STOPS domain, and NTSTEP number of output time steps (please see this added explanation in lines 284-5. As STOPS moves it calculates mixing ratios for different locations.

Figure 5: triangles and crosses cannot be identified. Please plot differently, e.g. in color.

--- We will replace the graphs with color ones.

"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" What is looked at? Surface ozone? 1.3 and 1.5 ppbv? kg?

--- yes, we looked at surface ozone (ppbV). We will add this information into a manuscript at lines 27, 31, 449.

p 7631 "STOPS system were validated against CMAQ calculated concentration". This assumes that CMAQ is the reference. But what is if the lagrangian transport is just more accurate, with less diffusion. Then a

difference would be an enhancement of the model. But the statistics would show the opposite effect. Please explain.

--- Since STOPS is based on CMAQ model so in this context we believe it is right to evaluate STOPS against CMAQ.

Table 5 and 6 could be converted into a figure to better show the convergence?

--- We prefer to show all the statistics currently listed in those tables and therefore would like to keep the tables.

Section 4:

At what time is the release? 6:00? And where exactly?

--- We will add the following explanation in line 19 p. 7632 (see lines 535-6): "The additional emission was added between 12 and 13 UTC at the location of the middle cell of STOPS domain at its starting position."

Why did you change between UTC and CST?

--- We will modify to UTC to be consistent with other parts of the manuscript.

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

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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 1.063 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 ~~columns of~~ 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~~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. 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.~~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.~~

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~~ Lagrangian movement~~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 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~~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 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 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. ~~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.~~

~~In the simplest application, the STOPS domain can consist of only one cell in the horizontal direction, which corresponds to a 2D column shown in Fig. 1. The modeling domain can be extended with a few horizontal layers of cells padding the targeted analysis domain.~~

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.

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 a Lagrangian column models.

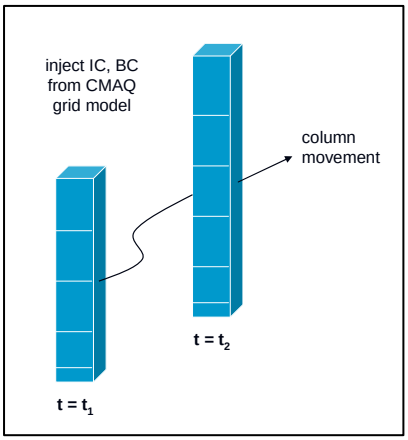

Figure1. The conceptual model for STOPS trajectory movement.

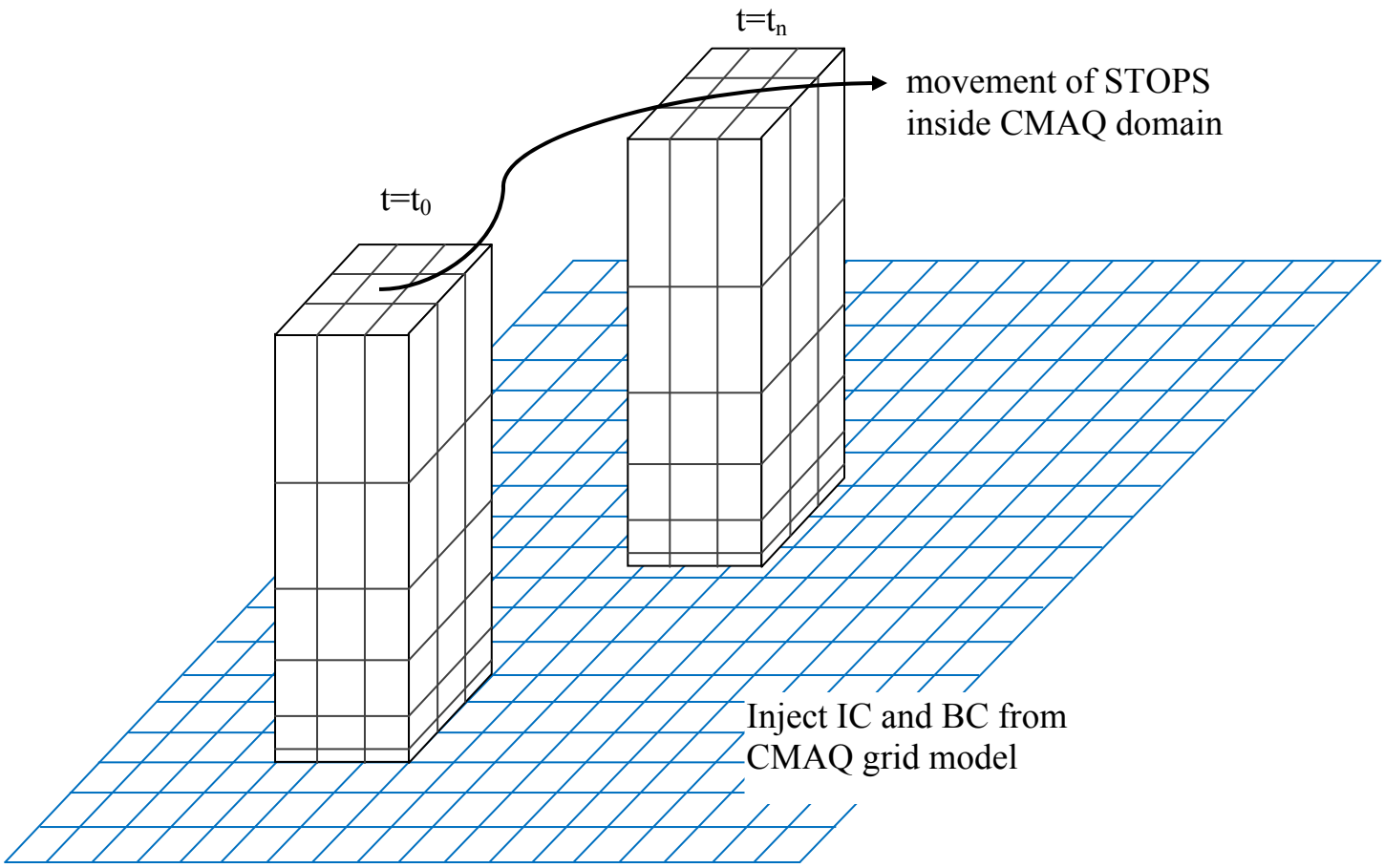

Figure1. The conceptual model ~~for~~ of STOPS and its ~~trajectory~~ movement.

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:

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

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

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

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

~~$$\bar{w}_{PBL} = \frac{1}{M_{PBL}} \sum_{l=1}^{l=PBL} w_l \cdot \Delta\sigma_l \quad (1)$$~~

~~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:~~

~~$$M_{PBL} = \sum_{l=1}^{l=PBL} \Delta\sigma_l \quad (2)$$~~

~~where σ is defined as:~~

$$\sigma = \frac{(p - p_t)}{(p_s - p_t)} \quad (\text{~~3~~}2)$$

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~~s~~ 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*
~~Daily ozone maximum from STOPS simulations —~~ and *SMAX*, respectively.

For cases when STOPS grids do not align with CMAQ grids the CMAQ values from several grid cells 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 ~~STOPS in the stationary mode~~ 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.~~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 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~~ column in each STOPS domain relatively to the CMAQ (host) grid, number of padding cells in each direction around ~~a~~ STOPS middle ~~cell~~column, and a number of total STOPS columns and rows ~~of the host domain~~ 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.

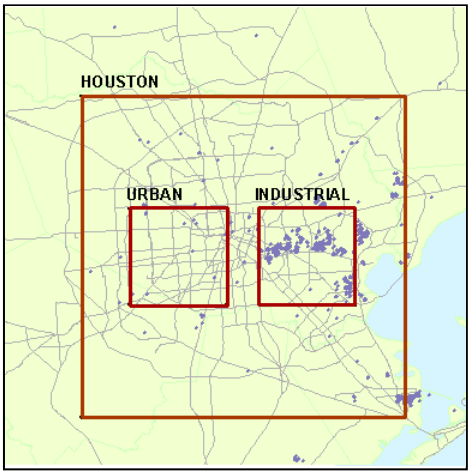

Figure.2. Starting locations of STOPS domains. Points indicate location of emission point sources in Houston.

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 ~~modeled~~ CMAQ calculated mixing ratios ~~concentrations~~ are also saved ~~with an~~ at hourly ~~output time step~~intervals. However, ~~Because a model performs~~ 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 ~~, which~~ is also a case for STOPS. ~~that uses CMAQ hourly concentrations for boundary conditions.~~ For the comparison of STOPS results with CMAQ values we used CMAQ concentrations from the ~~corresponding~~ 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 ~~to calculate~~ advection is calculated ~~CMAQ uses~~ based on values from adjacent cells at every synchronization time step. In ~~as opposed to~~ 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. ~~and t~~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 Table2. 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~~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~~concentration pairs~~, when STOPS was used in the stationary mode ~~(the values of MAXD and MIND are given in ppbv)~~. "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

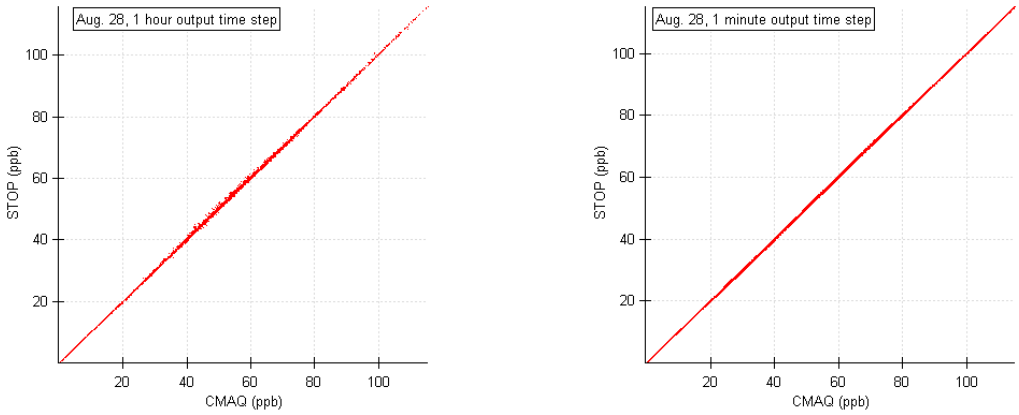

Figure 3. Comparison of CMAQ and static STOPS simulation results for Aug. 28 for 1 hour (left) and 1 minute (right) output time step. Both graphs correspond to simulation from the Houston domain.

3.2 ~~STOPS in the moving mode~~ Uncertainties related to movement of STOPS

The next step in the STOPS verification was to analyze uncertainties related to the movement of a ~~nest~~ 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. ~~In order to overcome this problem, f~~For the ~~comparison~~ 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 ~~either~~ the STOPS ~~domain~~ grid cells ~~had to be aligned~~ 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 ~~or~~ weighted average from several CMAQ grid cells that overlap with the STOPS grid cell~~STOPS values from several cells have to be interpolated to the corresponding CMAQ cell~~. The performance evaluation was tested for these two ~~possibilities~~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.

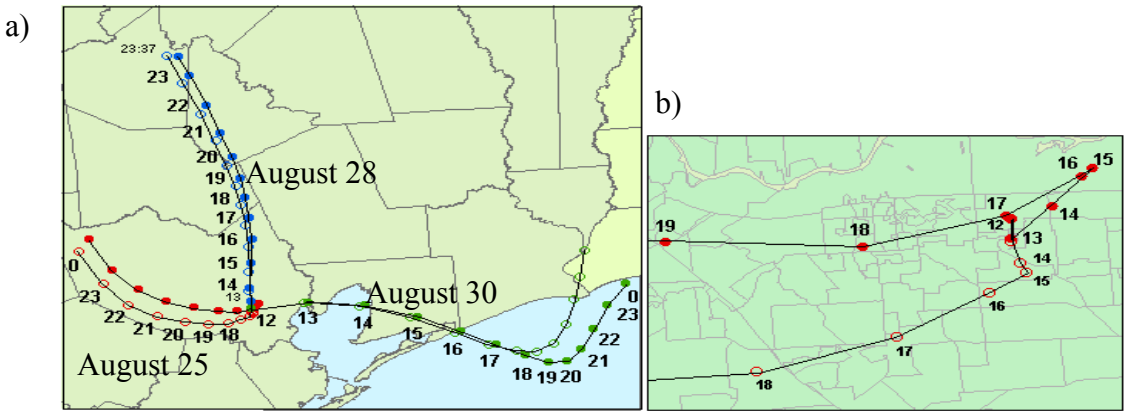

Figure 4. a) STOPS trajectories starting from the industrial sub-domain. Trajectories determined based on the winds in the STOPS middle column are indicated by filled circles, and those determined based on the average winds in the whole STOPS domain with open circles. Trajectories for Aug. 25 are indicated 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 ~~STOPS~~ CMAQ concentrations were spatially interpolated to be compared with ~~CMAQ~~ STOPS ~~concentrations~~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 ~~simulations~~ when ~~the~~ 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 ~~concentrations~~ 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 ~~STOPS~~ CMAQ values had to be ~~were~~ interpolated.

458 Table 3. A summary of statistical parameters for STOPS-CMAQ concentrations, when STOPS was used
459 in the moving mode, with the starting position at the urban sub-domain ~~(the values of MAXD and MIND~~
460 ~~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	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

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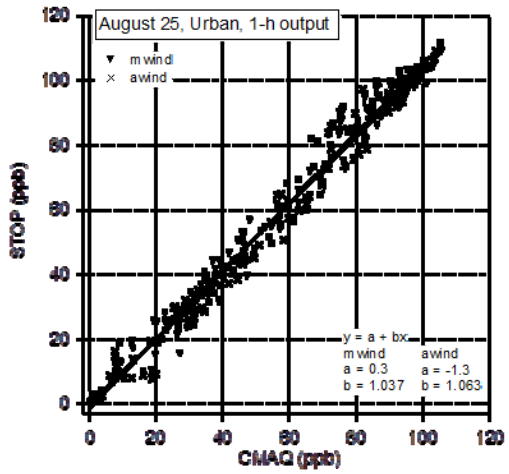
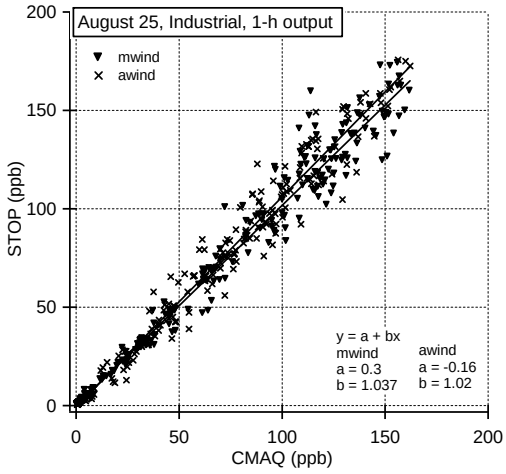
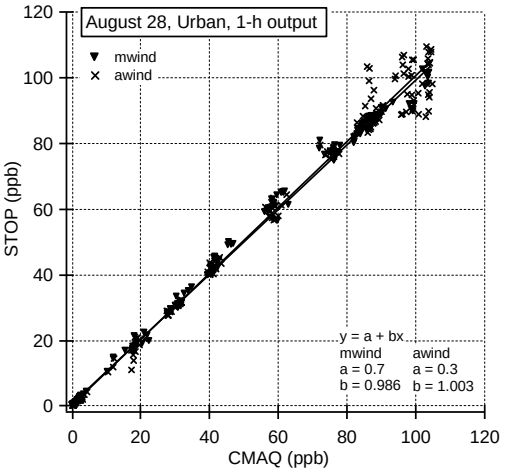
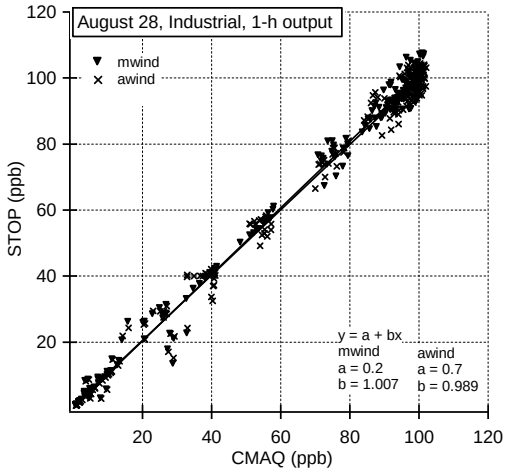
Table 4. A summary of statistical parameters for STOPS-CMAQ concentrations, when STOPS was used in the moving mode, with the starting position at the industrial sub-domain ~~(the values of MAXD and MIND are given in ppb)~~.

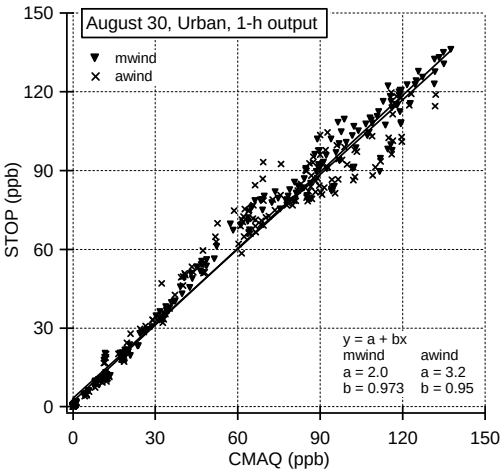
NAME	N	HMAX	SMAX	MB	MAE	RMSE
awind_~~urb~~ind_1h.0825	217	162.1	175.6	-3.7049	6.667	9.7334
awind_ind~~urb~~_1h.0828	201	102.0	104.5	-0.0743	2.7724	3.6884
awind_ind~~urb~~_1h.0830	217	141.4	140.1	0.5727	2.2085	3.4874
awind_ind~~urb~~_5m.0825	2329	166.2	179.9	-4.2896	6.9033	10.246
awind_ind~~urb~~_5m.0828	2281	102.0	105.4	-0.0317	2.8724	3.7569
awind_ind~~urb~~_5m.0830	2329	141.7	140.5	0.7063	2.4671	3.9274
awind_ind~~urb~~_1m.0825	11545	166.0	178.6	-4.0882	7.0306	10.1471
awind_ind~~urb~~_1m.0828	11373	101.5	106.2	-0.2101	2.9622	3.8751
awind_ind~~urb~~_1m.0830	11545	140.4	139.7	0.6337	2.3704	3.7275
mwind_ind~~urb~~_1h.0825	217	162.1	174.0	-1.2557	6.3057	9.6064
mwind_ind~~urb~~_1h.0828	201	101.6	107.3	-0.6898	2.3871	3.4938
mwind_ind~~urb~~_1h.0830	217	138.0	136.8	0.125	1.4439	1.9605
mwind_ind~~urb~~_5m.0825	2329	166.4	178.7	-1.0198	6.3622	9.4587
mwind_ind~~urb~~_5m.0828	2217	101.7	105.6	-0.2336	2.3862	3.3116
mwind_ind~~urb~~_5m.0830	2329	141.8	137.4	0.9498	2.0799	2.8743
mwind_ind~~urb~~_1m.0825	11545	166.0	177.7	-0.6788	6.2981	9.3914
mwind_ind~~urb~~_1m.0828	11017	101.1	105.7	-0.3779	2.2792	3.2517
mwind_ind~~urb~~_1m.0830	11545	140.0	136.6	0.743	1.9787	2.6921
awind_ind~~urb~~_1h_sh.0825	325	162.1	175.6	-2.7155	4.1153	6.5406
awind_ind~~urb~~_1h_sh.0828	300	102.6	104.5	-0.0949	1.5528	2.2241
awind_ind~~urb~~_1h_sh.0830	325	141.5	141.3	-0.0785	1.6427	2.3778
awind_ind~~urb~~_5m_sh.0825	3625	166.4	179.9	-1.0475	3.9286	6.2411
awind_ind~~urb~~_5m_sh.0828	3550	102.4	105.4	-0.0618	1.4688	2.0437
awind_ind~~urb~~_5m_sh.0830	3625	142.4	142.2	-0.1354	1.6548	2.502
awind_ind~~urb~~_1m_sh.0825	18025	166.0	178.6	-1.0034	4.0013	6.2608
awind_ind~~urb~~_1m_sh.0828	17750	101.9	106.2	-0.3176	1.4425	2.0392
awind_ind~~urb~~_1m_sh.0830	18025	141.0	141.1	-0.1505	1.6257	2.3916
mwind_ind~~urb~~_1h_sh.0825	325	162.1	174.0	-2.4646	3.9385	6.1064
mwind_ind~~urb~~_1h_sh.0828	300	101.9	107.3	-0.782	1.5209	2.1193
mwind_ind~~urb~~_1h_sh.0830	325	141.1	141.3	-0.224	1.3034	1.6851
mwind_ind~~urb~~_5m_sh.0825	3625	166.4	178.7	-1.0628	4.012	6.134
mwind_ind~~urb~~_5m_sh.0828	3450	101.7	105.6	-0.3803	1.3697	1.8761
mwind_ind~~urb~~_5m_sh.0830	3625	142.4	143.1	-0.1763	1.4963	2.0331
mwind_ind~~urb~~_1m_sh.0825	18025	166.0	177.7	-0.8412	3.9665	6.0567
mwind_ind~~urb~~_1m_sh.0828	17200	101.2	105.7	-0.6202	1.4004	1.9443
mwind_ind~~urb~~_1m_sh.0830	18025	140.8	141.6	-0.355	1.4364	1.9099

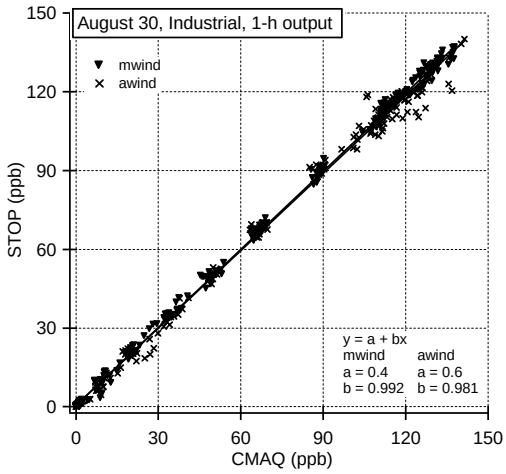

August 25, Urban, 1-h output
mwind
awind
STOP (ppb)
CMAQ (ppb)
y = a + bx
mwind awind
a = 0.3 a = -1.3
b = 1.037 b = 1.063

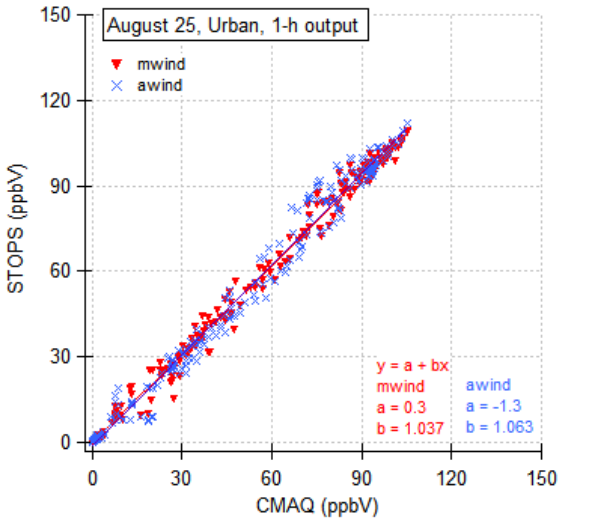

August 25, Industrial, 1-h output
mwind
awind
STOP (ppb)
CMAQ (ppb)
y = a + bx
mwind awind
a = 0.3 a = -0.16
b = 1.037 b = 1.02

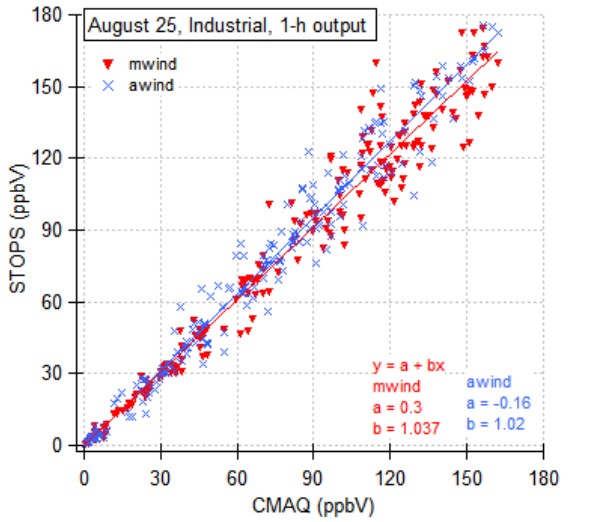

August 28, Urban, 1-h output
mwind
awind
STOP (ppb)
CMAQ (ppb)
y = a + bx
mwind awind
a = 0.7 a = 0.3
b = 0.986 b = 1.003

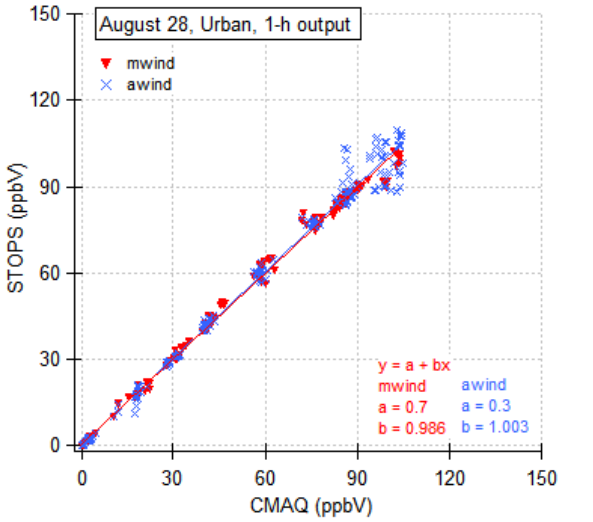

August 28, Industrial, 1-h output
mwind
awind
STOP (ppb)
CMAQ (ppb)
y = a + bx
mwind awind
a = 0.2 a = 0.7
b = 1.007 b = 0.989

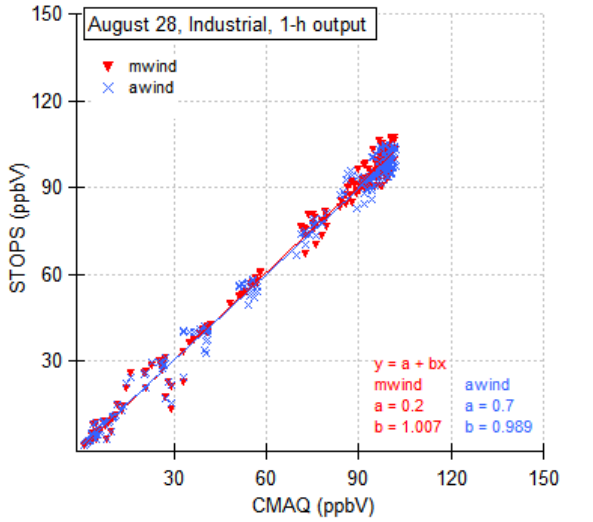

August 30, Urban, 1-h output
mwind
awind
STOP (ppb)
CMAQ (ppb)
y = a + bx
mwind awind
a = 2.0 a = 3.2
b = 0.973 b = 0.95


August 30, Industrial, 1-h output
mwind
awind
STOP (ppb)
CMAQ (ppb)
y = a + bx
mwind awind
a = 0.4 a = 0.6
b = 0.992 b = 0.981


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


August 25, Industrial, 1-h output
mwind
awind
STOPS (ppbV)
CMAQ (ppbV)
y = a + bx
mwind
a = 0.3
b = 1.037
awind
a = -0.16
b = 1.02


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


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

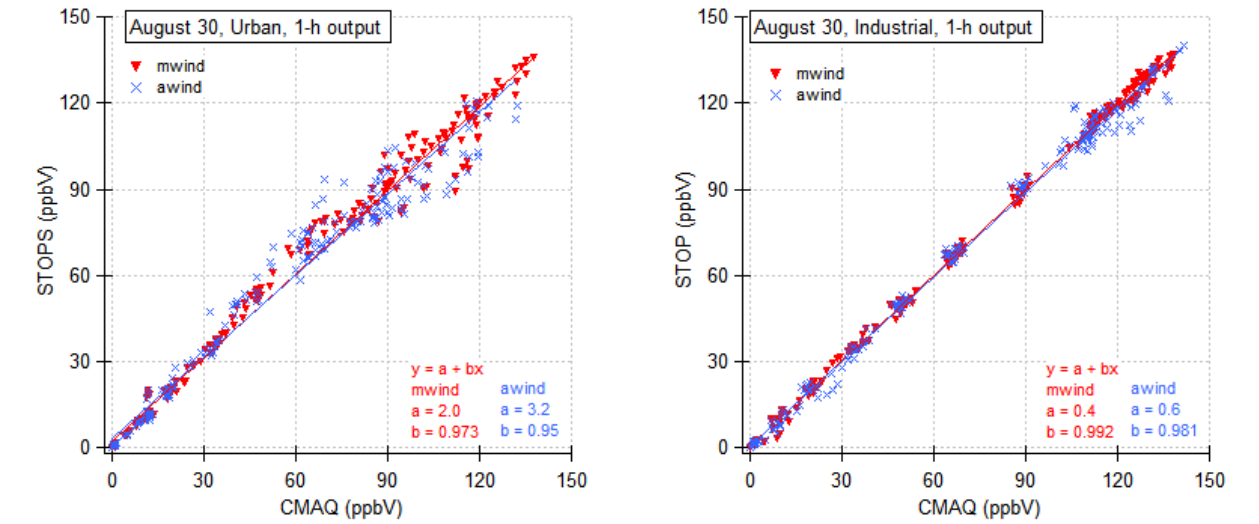


Figure 5. Comparison of ozone concentrations obtained with STOPS and CMAQ for Aug. 25, 28, and 30 for the STOPS starting position at the urban sub-domain (left figures) and the industrial sub-domain (right figures). Triangles correspond to the trajectory determined from winds in the middle column (mwind), 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 ~~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 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 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 August 25, 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 Table 5 for the stationary case and in Tables 6 and 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.

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 ~~the~~ ozone mixing ratio locally, close to the emission source, and with higher magnitude.

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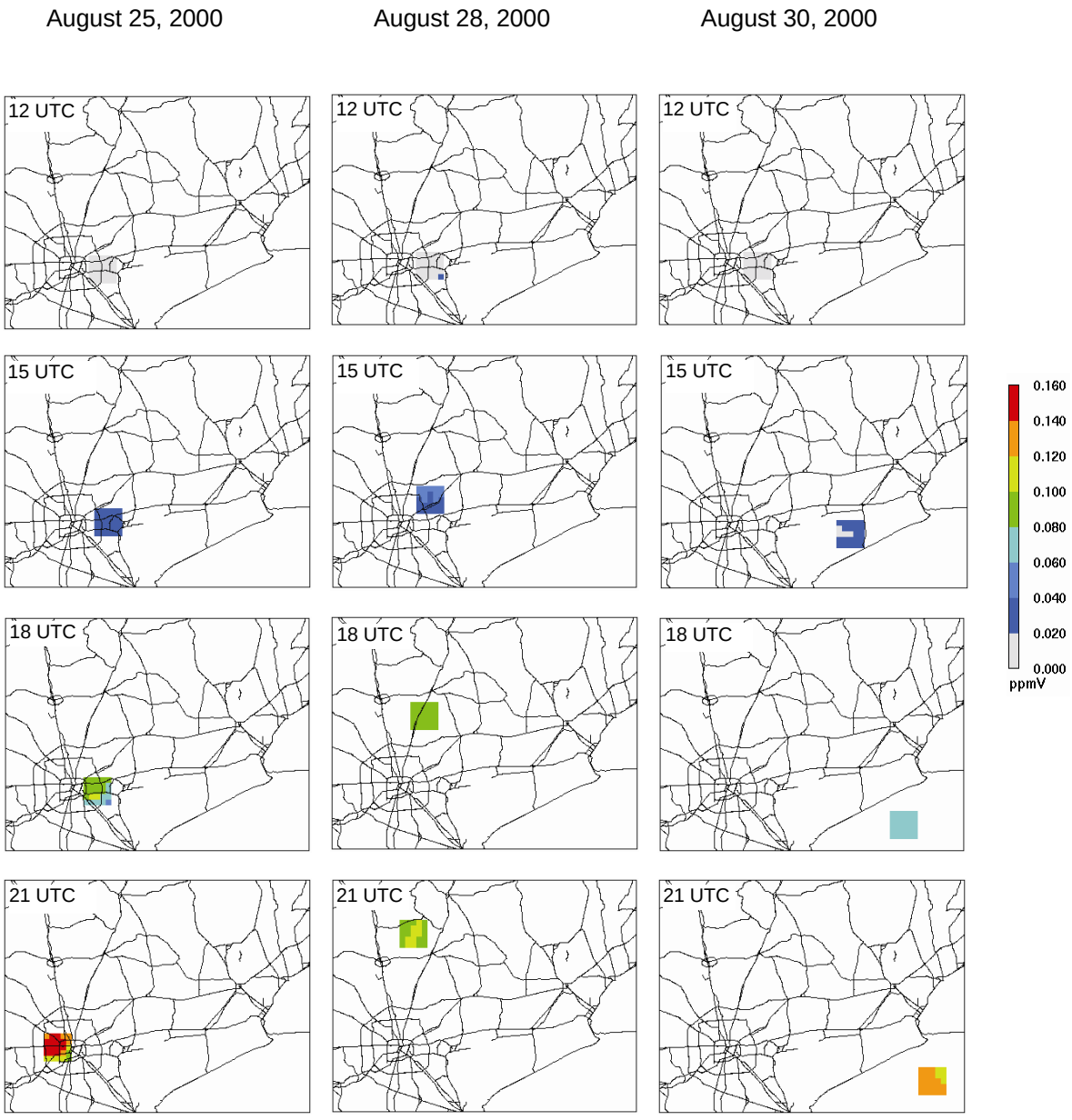

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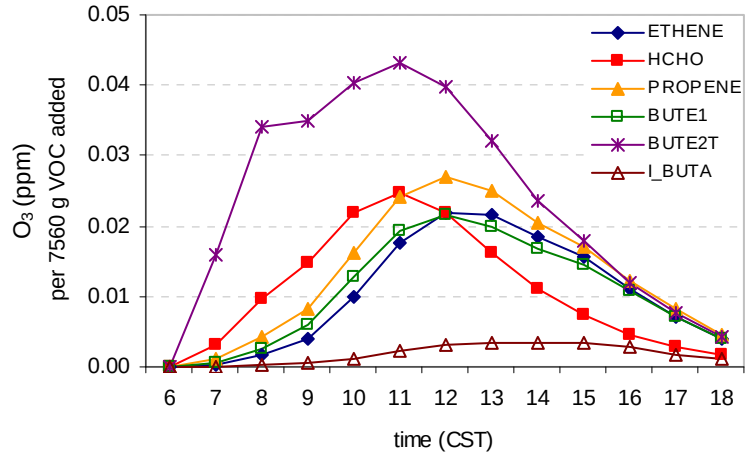
549 Figure 6. Snapshots of ozone concentrations along STOPS trajectories on August 25
550 (left), August 28 (middle), and August 30 (right) when the STOPS simulation started
551 from the industrial sub-domain.



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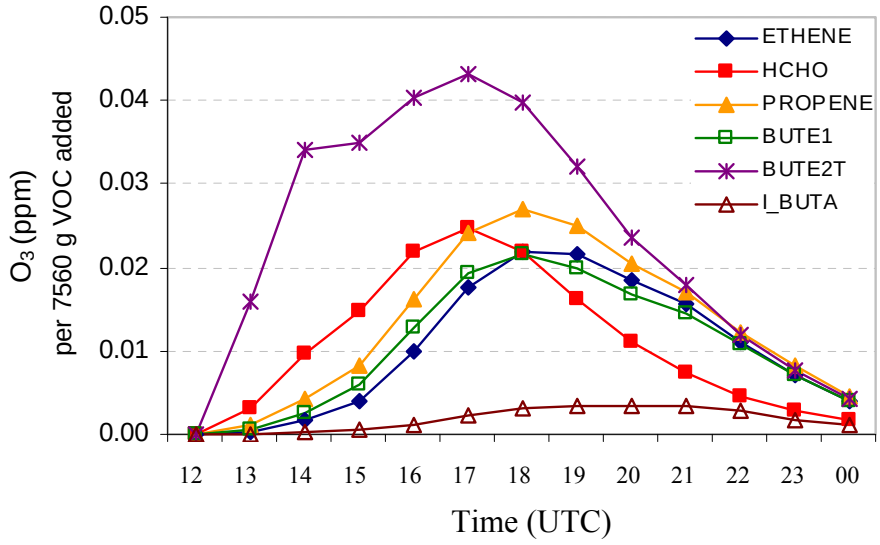

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555 Figure 7. Changes in ozone along STOPS trajectory on August 25 due to emission spike
556 of different individual VOCs. The values are integrated in the surface layer of the
557 STOPS domain.

558

5. Summary

A hybrid Lagrangian-Eulerian based 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 and it was shown that STOPS is capable of predicting ozone mixing ratios in close agreement with CMAQ predictions. ~~STOPS~~ Its performance however 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 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~~3~~ 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.

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