Population Exposures to Ethylene: A Literature Review (and Modeling Exercise)

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Overview

Goal
To conduct a preliminary assessment of inhalation exposure potential in human populations with a specific focus source contribution

Task 1 – evaluate available literature information

Task 2 – conduct preliminary modeling estimates of industrial emissions to ambient concentrations

Inhalation Exposure Determination

General approaches to exposure assessment

1. Direct measurement of the personal exposure for an appropriate sample of the population.

2. Indirect measurement using local ambient air concentrations and general activity patterns in specific microenvironments.

3. Mathematical models using emission and air speed information.

4. Biomonitoring using blood, urine, or exhaled air.

Sources of Ethylene Emissions

Biogenic

- Vegetation
- Leaf litter
- Seawater
- Soil
- Marine sediment

Biomass burning

- Forest fires
- Crop residue burning

Anthropogenic

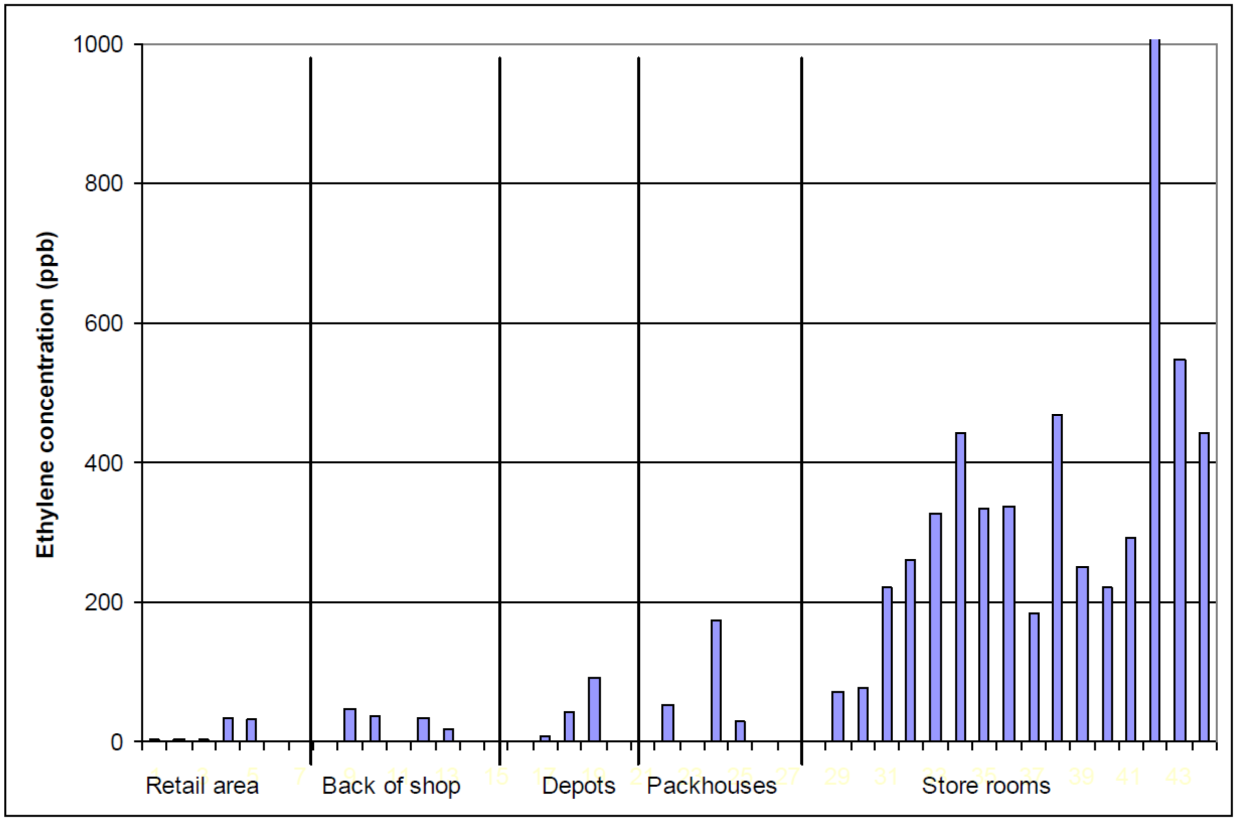
- Olefin plants
- Refineries
- Traffic
- Incineration

Occupational Exposure Levels[1]

Facility	Sample type	Mean (ppb)	Range (ppb)	Author
coke oven battery	personal	231	10.8 – 1288	Lin et al., 2007
green produce distributor	area	---	80 – 1412	Wills et al., 2000
olefin Plant	personal	2600	50 – 49000	Martin & Caldwell, 2004
petrochemical plant	area	---	11 – 193	Lin et al., 2004
horticultural industry[2]	area		0 – 133000	Fraser et al, 1999

[1] ACGIH 8-hr TLV of 200 ppm (200,000 ppb)
[2] operations included growers, wholesalers, retailers and processors of fruits, vegetables and flowers

Ethylene Levels Associated with the Storage and Distribution of Consumer Produce



Rees et al., 2011

Levels Typically Observed in Different Environments

Environment	Range (ppb)
clean air background	0.1 – 0.2
rural	0.4 – 0.7
urban background	0.3 – 1.5
indoor air	2 – 10
busy roadway	5 – 15
industrial fenceline	10 – 100
secondhand smoke	50 – 100
occupational	100 – 50000

Measurements compiled from a review published information

Sources and Airborne Concentrations of Ethylene

Sample type	Conc (ppb)	Author
Waste incinerator flue gas	5600	Carotti et al., 1972
Above land fill	2700	Bogner et al., 2010
Second hand smoke	80	Persson et al., 1988
Expire air	23	Conkle et al., 1975
Home refrigerator[1]	100	Wills et al., 2000
Forest fire plume	37	Rinsland et al., 2005
Incense burning	17,000	Yang et al., 2007
Kitchen using LPG	113	Huang et al., 2011
Charcoal briquette smoke	3500	Olsson, 2003
Below snow pack	0.16	Swanson et al., 2002
Above soybean field	30.3	Kang et al., 2004

[1] ranged as high as 590 ppb

Population Exposures to Ethylene

Far-field sources

- sources located at distances that are many meters or kilometers away from where a person spends time or performs an activity
- may contribute to population exposure depending on overlap between activity
- industrial releases and biomass burning

Near-field sources

- sources located within a few meters of a location where a person spends time
- always contributes to the overall level of exposure
- motor vehicle emissions, cooking, building products off-gassing, consumer products

Personal Exposures to Ethylene

Personal exposures to ethylene in commuters within the city of Dublin, Ireland (McNabola et al., 2008)[1]

Statistic	Car (ppb)	Bicycle (ppb)	Bus (ppb)	Pedestrian (ppb)
mean	7.32	8.83	8.46	6.18
SD	9.67	4.55	4.07	4.50
N	45	42	27	37

[1]three mile route through heavy traffic

Indoor/Outdoor Ratios for Ethylene

Location	Indoor conc. (ppb)	Outdoor conc. (ppb)	I/O ratio	Author
Rio Grand Valley	5.41	1.75	3.86	Mukurjee et al., 1997
Regina, Canada	3.32	1.31	2.53	Health Canada, 2010
Windsor, Canada	5.24	2.79	1.87	Health Canada, 2010
Halifax, Canada	2.44	0.80	3.05	Health Canada, 2009
Nepal	245	1.05	233	Davidson et al., 1986
Beijing, China	10.46	6.75	1.55	Duan et al., 2014
Boise, ID[1]	13.5	12.5	1.08	Lewis , 1991

[1]No obvious indoor sources

Personal Population Exposures to Ethylene

Location (season)	Sample size	Mean (ppb)	Range (ppb)
personal (summer)	207	5.01	1.12 – 105.00
personal (winter)	119	5.73	0.94 – 57.61
indoor (summer)	217	5.03	0.76 – 117.00
outdoor (summer)	216	1.30	0.16 – 3.89
indoor (winter)	91	5.17	0.24 – 63.12
outdoor (winter)	126	2.82	0.66 – 10.21

24-hr samples over two year period
100 participants
Children and Adults

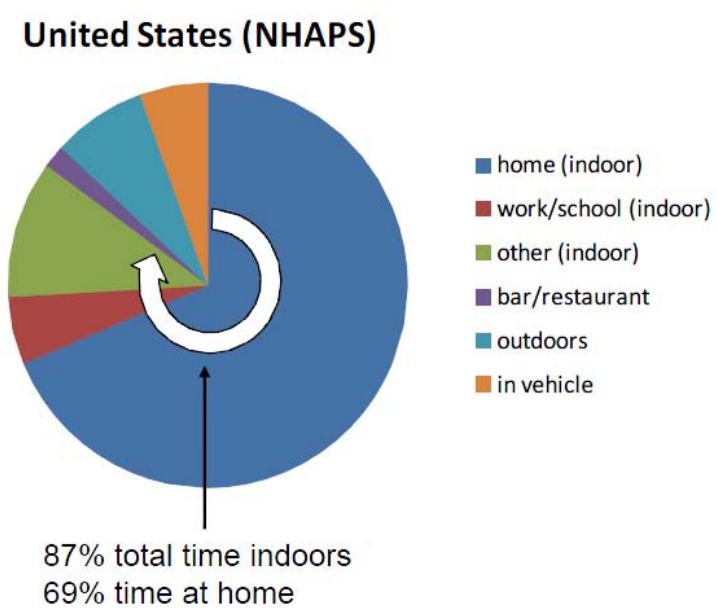
Health Canada, 2010

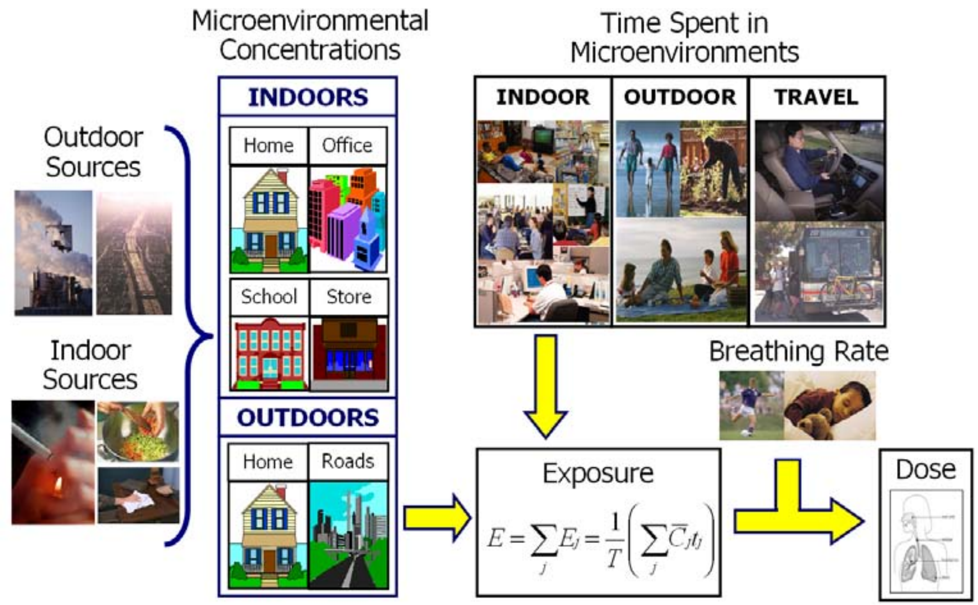
Findings from HEATS

- 27 adults surveyed in two areas in Houston
  - Aldine
  - Houston ship channel
- personal, indoor, and outdoor samples collected for 14 air toxics
- outdoor fixed-site measurements higher in the ship channel than in Aldine
- indoor/outdoor ratios generally ranged from 1.7 – 6.7 (9 of 14)
- personal exposures higher than residential or outdoor exposures in both study areas
- personal exposures in the two areas were similar and did not reflect differences in the type and density of point source emissions or the ambient concentrations at the two sites

Morandi et al., 2009

Time-Activity Patterns for Adults in the US





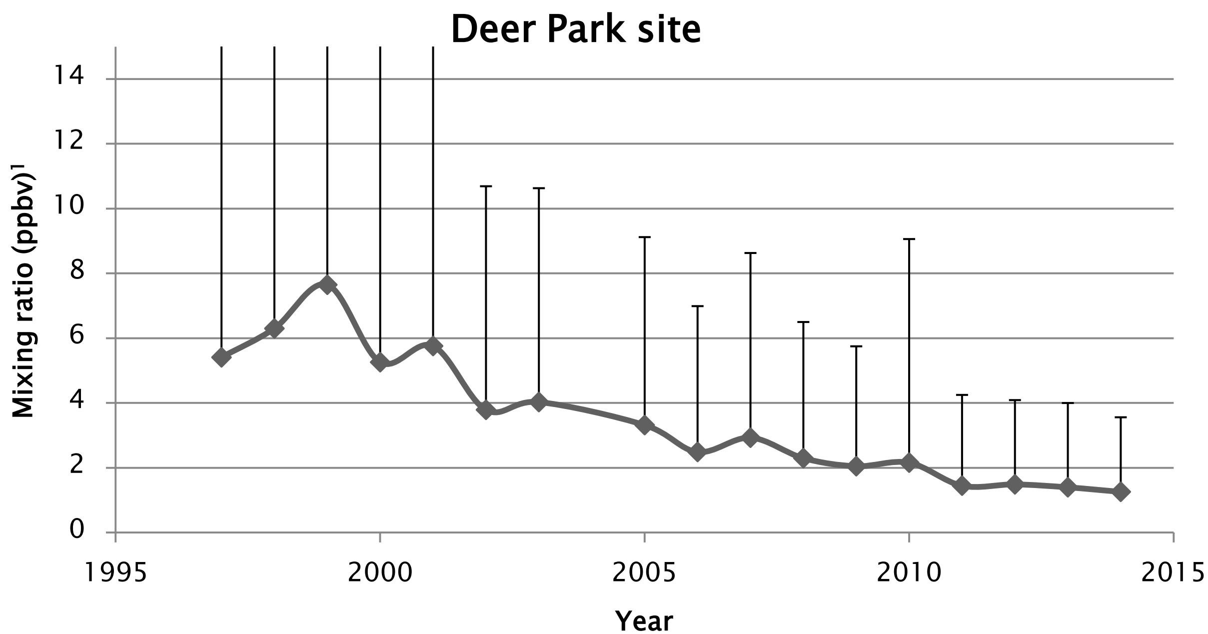
Leech et al., 2002

Indoor Sources of Ethylene Exposure

- house plants
- cooking oils
- fruits and vegetables
- ETS
- vehicle in attached garage
- cooking and heating fuels
- wood burning fireplace

Temporal Trends in Ambient Ethylene

Ethylene measurements at the Deer Park monitoring site located near the Houston Ship Channel



TCEQ. AutoGC Data by Day by Site (all parameters). Accessed 2014. Texas Commission on Environmental Quality

Conclusions from the Literature Review

- Diverse number of sources both indoors and outdoors
- Occupational exposures higher than population exposures but still below applicable limits
- High spatial and temporal differences in ambient air levels
- Indoor air levels generally higher than outdoor levels
- Personal exposures not well correlated with those found outdoors
- Relative contribution from industrial emissions and traffic difficult to exactly determine but evidence suggest that indoor sources far more important
- Microenvironmental modeling using time activity-analysis may yield useful insight on the contributions from specific sources

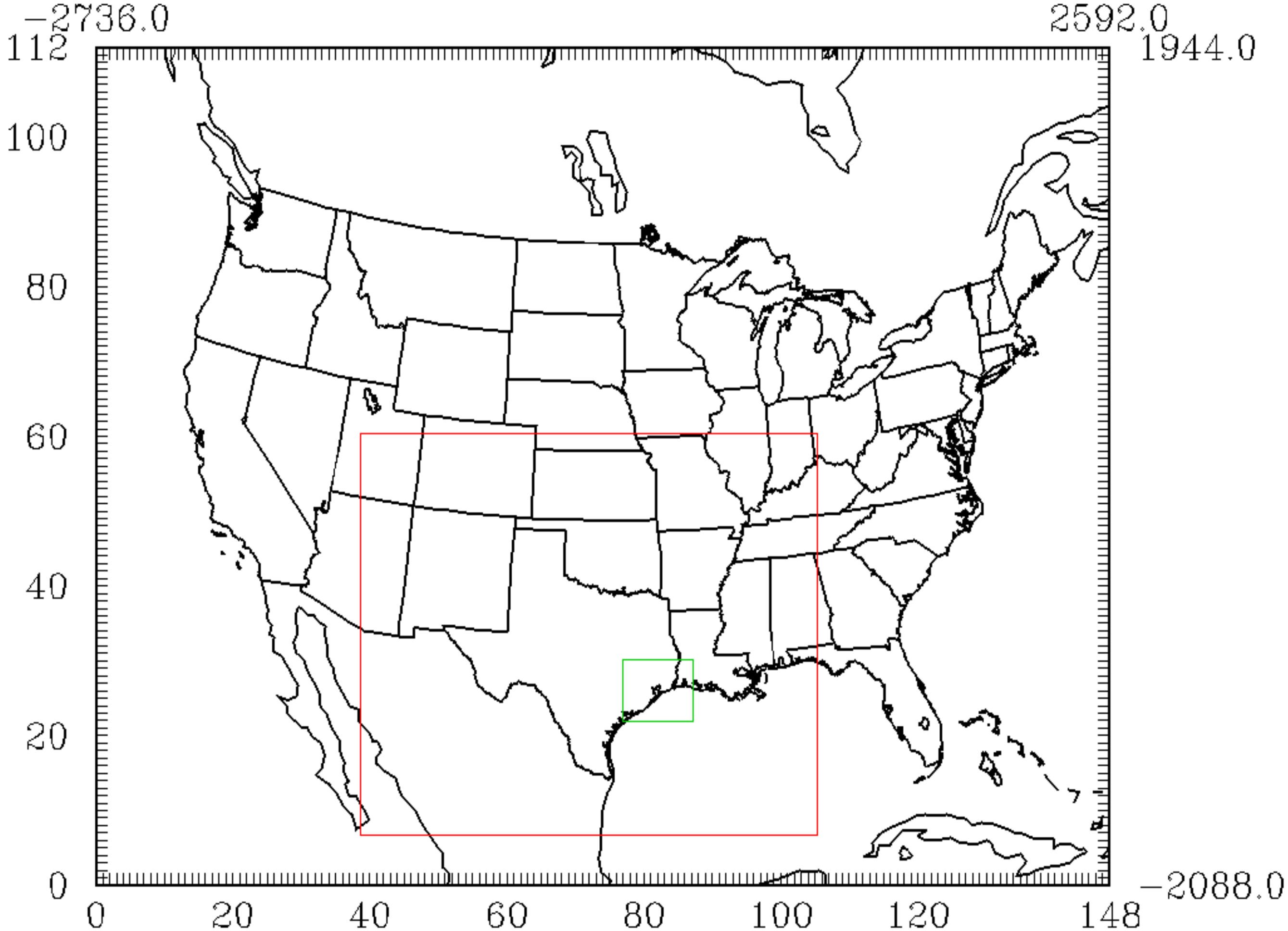
Motivation

- Provide a screening examination at monthly average ethylene concentrations in the Houston/Galveston/Brazoria area

Modeling Approach

- CAMx 6.1
- 36km/12km/4km Nested Application
- Ozone Season 2010 Modeling Platform
- Impacts Simulated Two Ways
  - Zero-Out: Removed industrial ethylene in 4km domain and reran model (Removing emissions changes reactivity)
  - Reactive Tracer (RTRAC): Run model with a reactive tracer to represent ethylene. Accounts for ethylene reacting with ozone, OH and NO_3.

Modeling Domain



Caveats

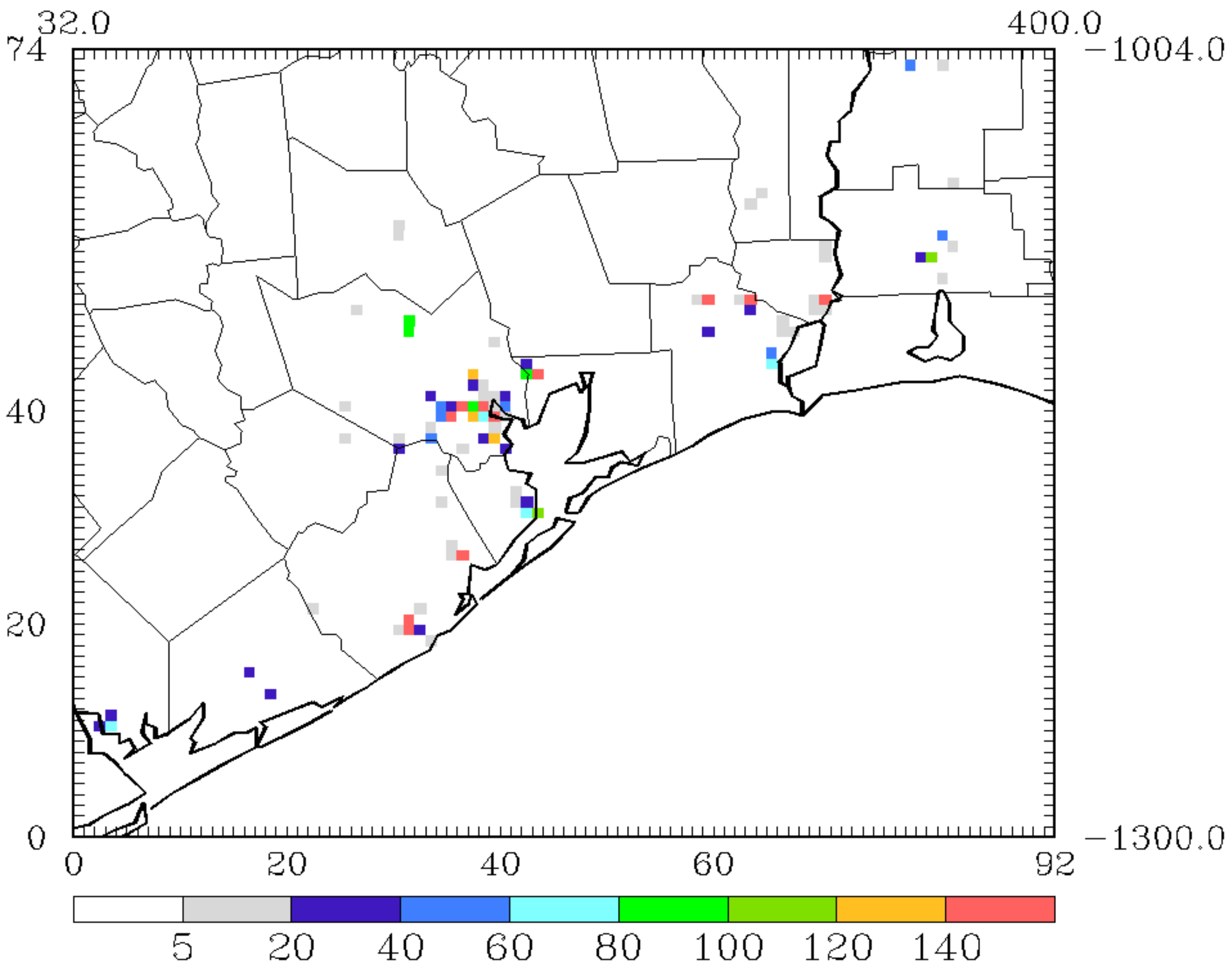
- Used existing CAMx platform for extended ozone season only (May–Oct. 18)
- This model platform has not been evaluated against ethylene observations
- CAMx model has artificial dilution of emissions to 16 km^2 grid volume
- Results must be viewed as “screening” level

Emissions

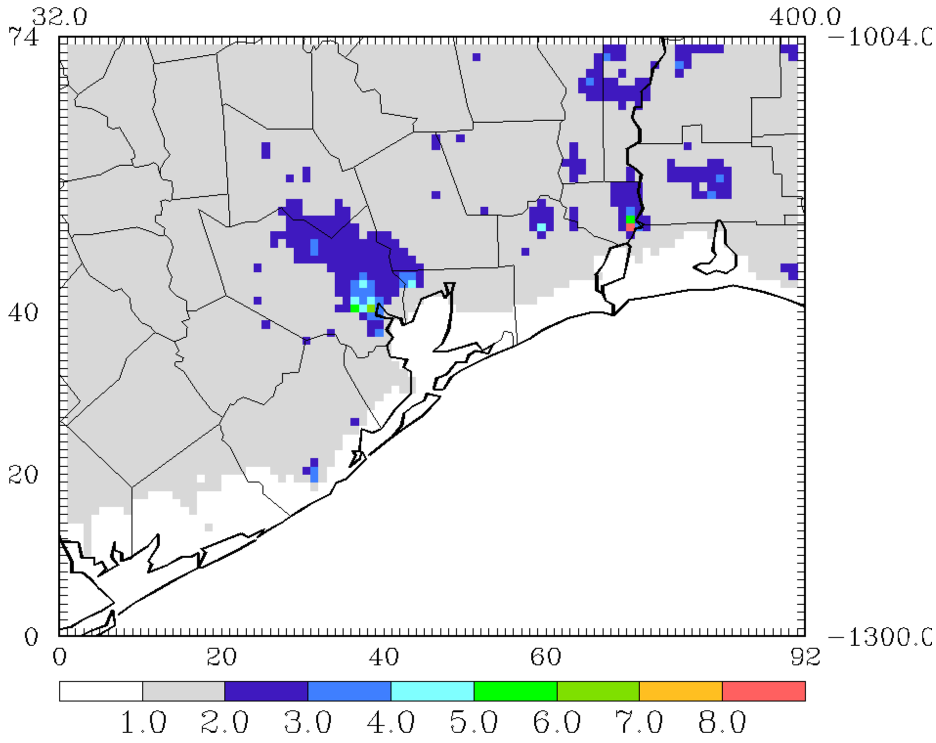
- EPA's National Emissions Inventory (NEI) for 2008,2011
- Continuous Emissions Monitor (CEM) data where available
- Wildfires – EPA 2010 Preformatted for SMOKE
- On-road Mobile – MOVES2010b
- Non-road Mobile – NMIM
- Biogenics – MEGAN

Industrial Ethylene Emissions Full Domain

Peak: 622 tpy
Grid Total: 5,435 tpy

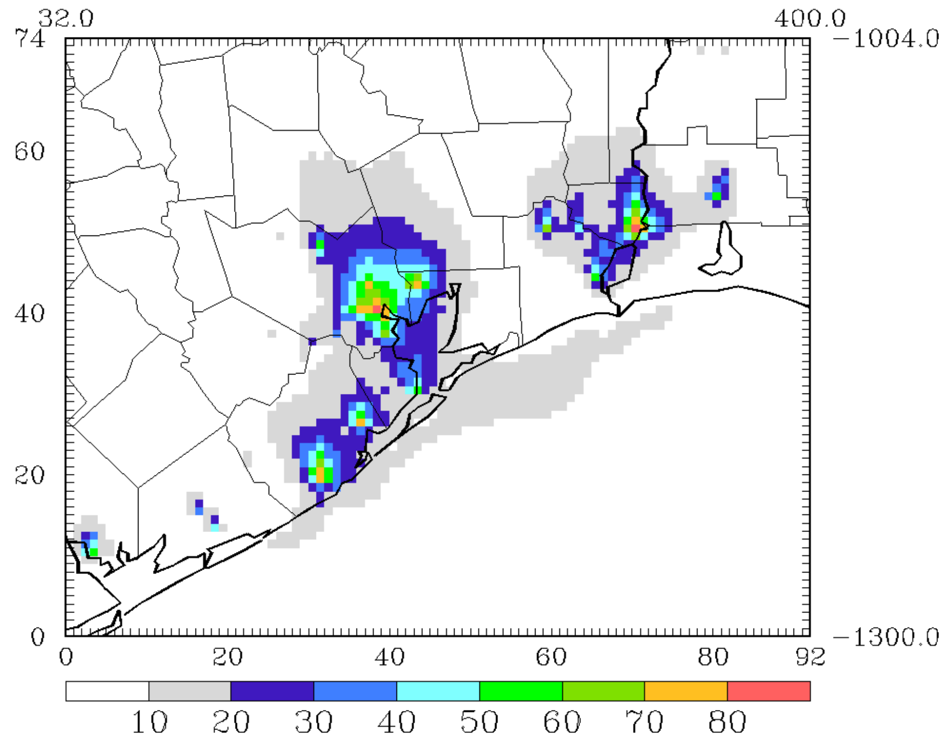


Modeling Results (May)

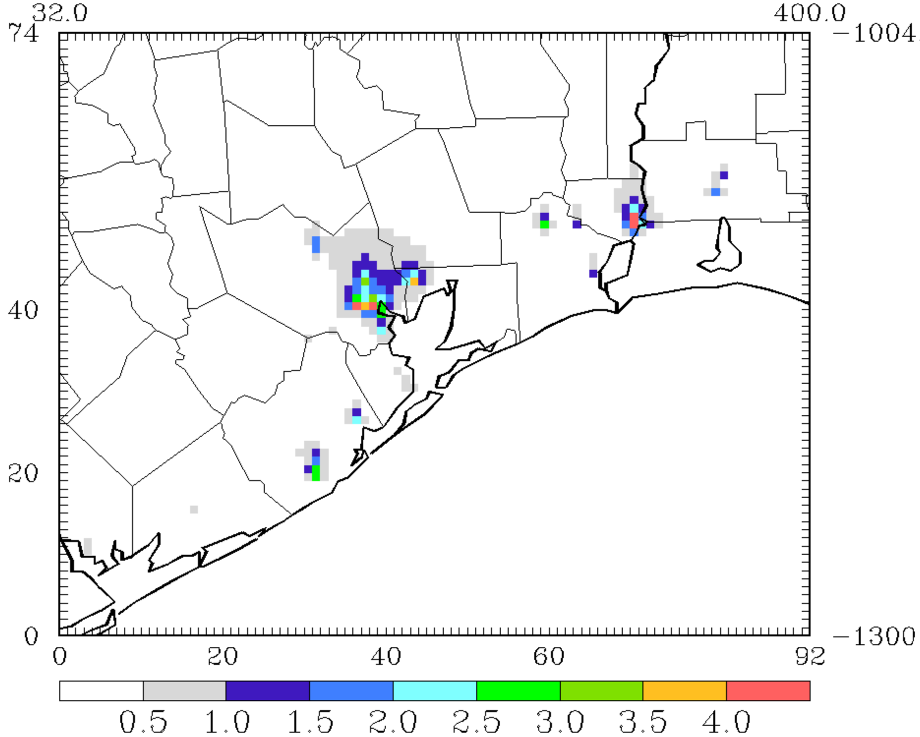


Base Concentration
Peak: 10.0 ppb
99th Percentile: 2.54 ppb
Median: 1.30 ppb

Percentage Contribution
Peak: 89.3%



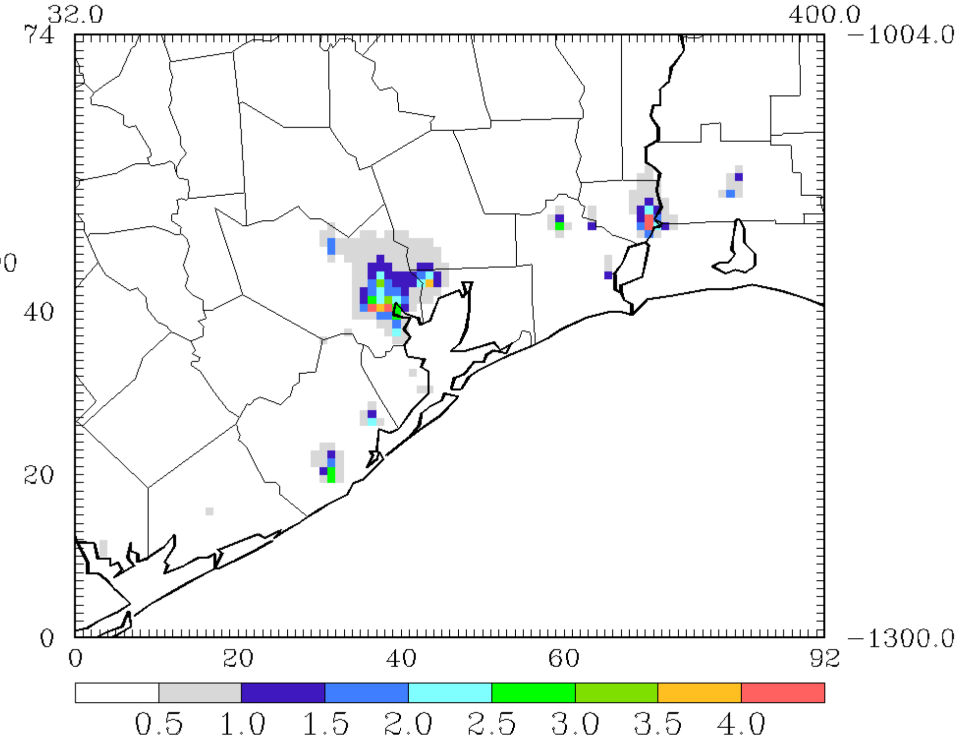
Modeling Results (May Conc.)



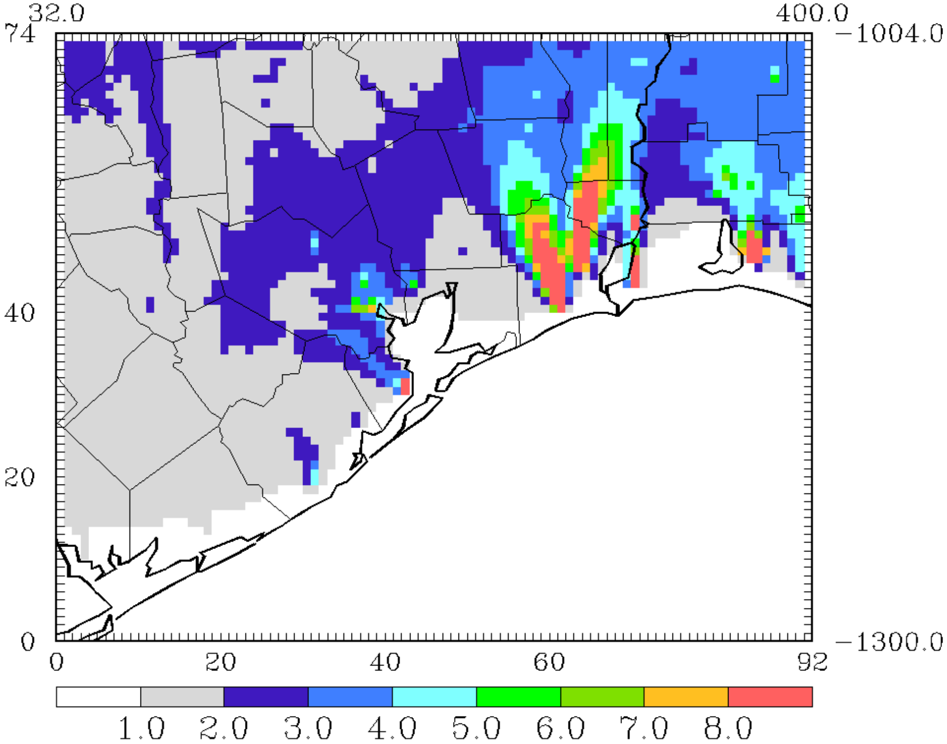
Zero-Out
Peak: 8.94 ppb
99th Percentile: 1.08 ppb
Median: 0.03 ppb

Industrial Contribution (ppb)

RTRAC
Peak: 8.97 ppb

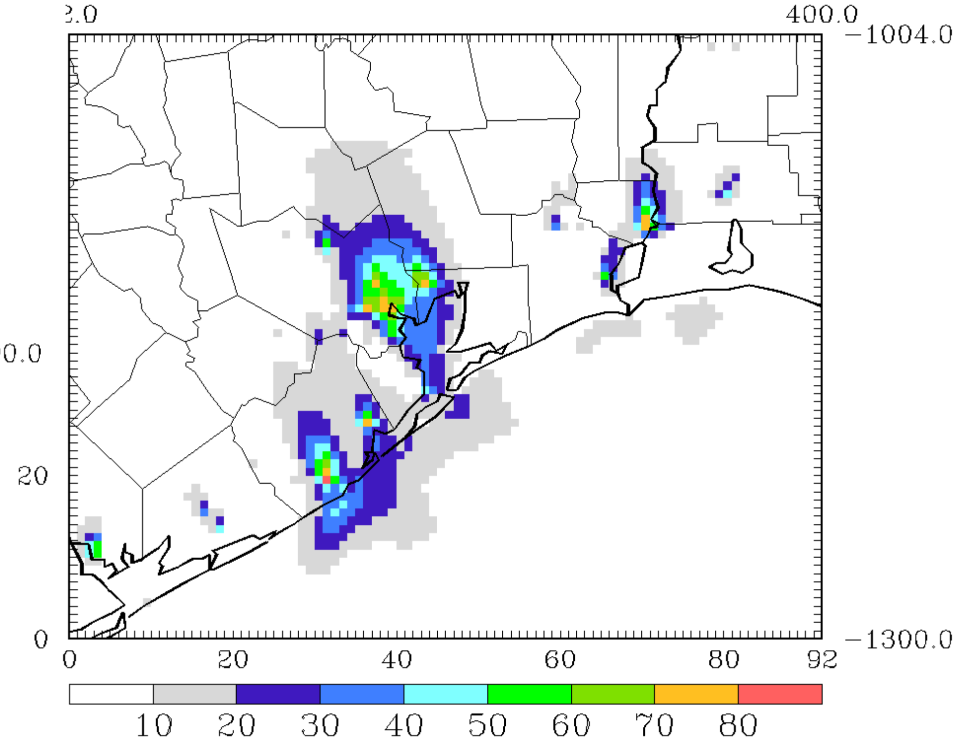


Modeling Results (June)

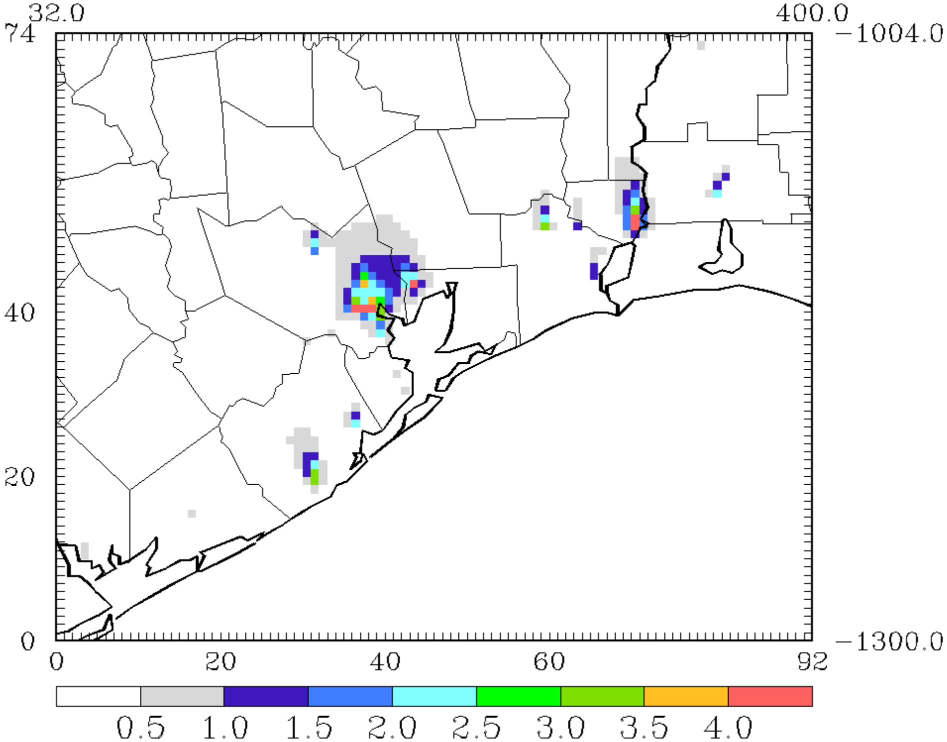


Base Concentration
Peak: 51.6 ppb
99th Percentile: 7.92 ppb
Median: 1.64 ppb

Percentage Contribution
Peak: 80.6%



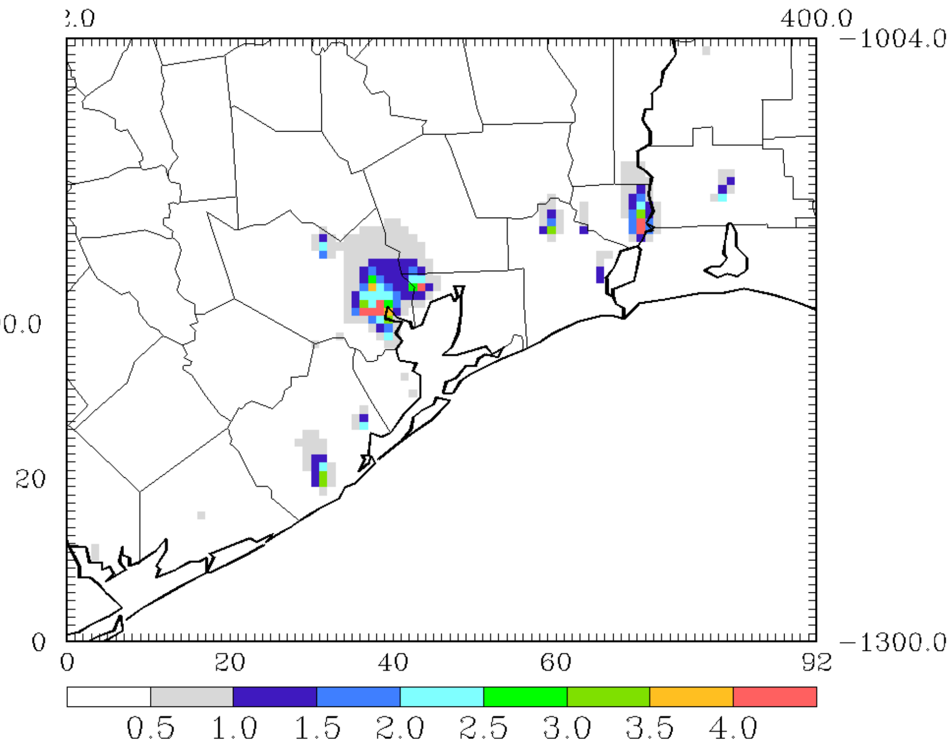
Modeling Results (June Conc.)



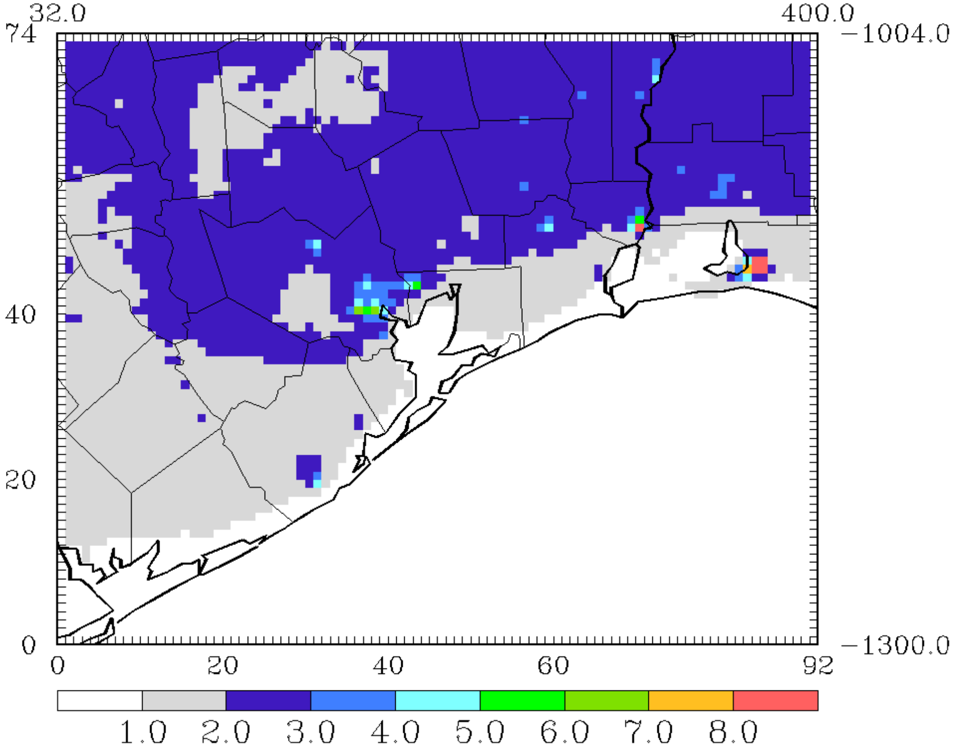
Zero-Out
Peak: 9.68 ppb
99th Percentile: 1.30 ppb
Median: 0.02

Industrial Contribution (ppb)

RTRAC
Peak: 9.68 ppb

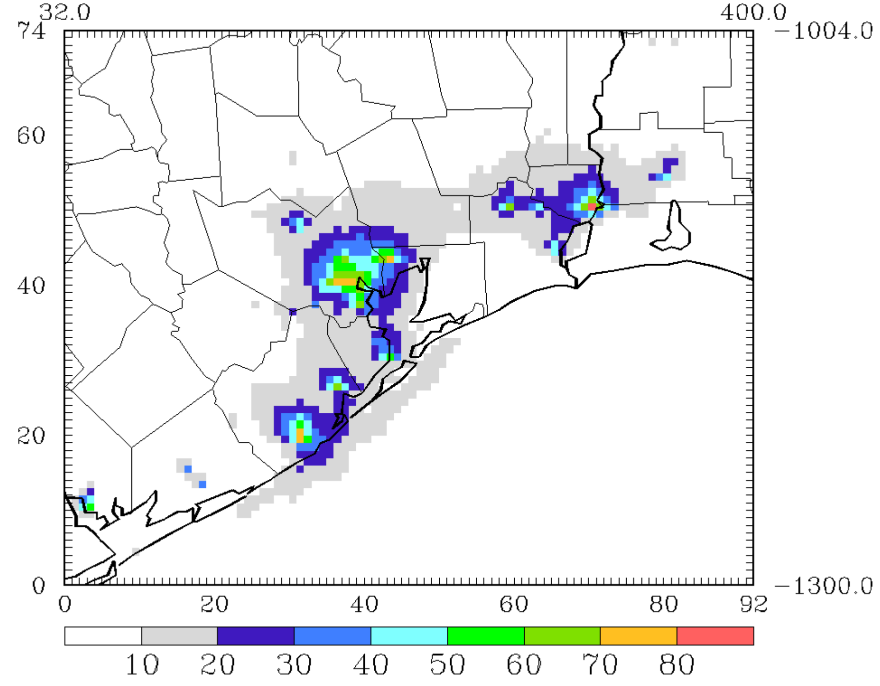


Modeling Results (July)

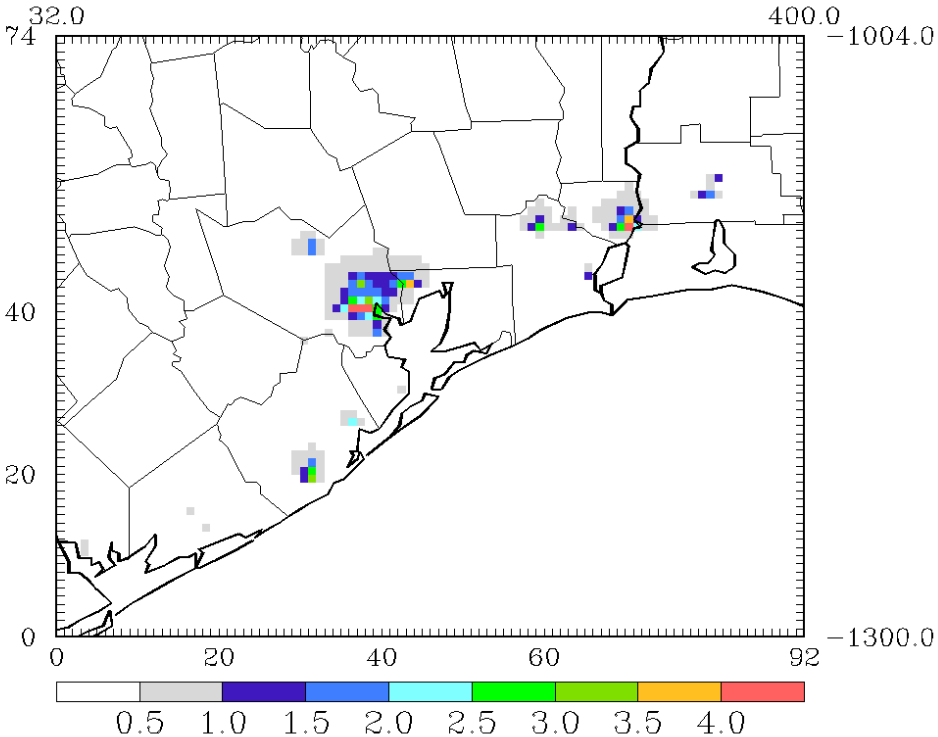


Base Concentration
Peak: 41.3 ppb
99th Percentile: 3.00 ppb
Median: 1.75 ppb

Percentage Contribution
Peak: 87.2%



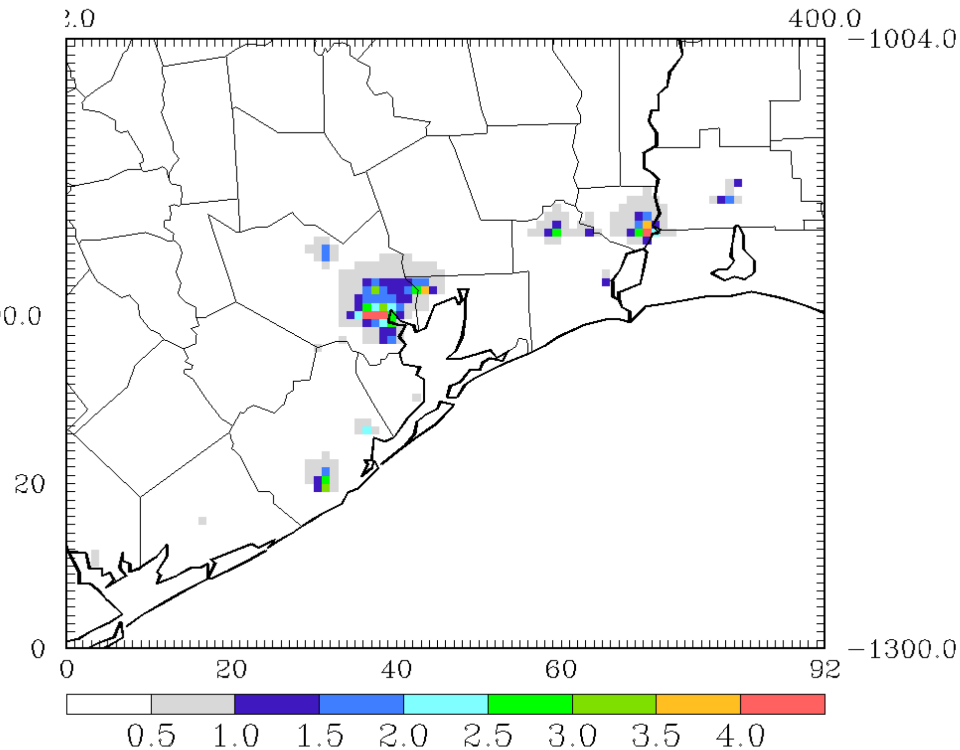
Modeling Results (July Conc.)



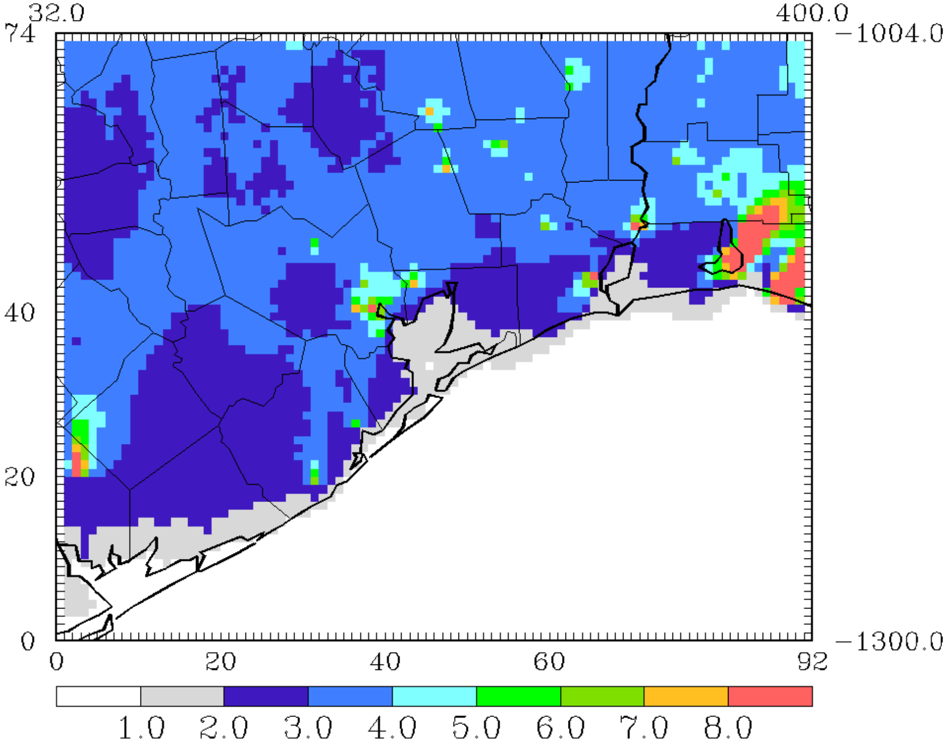
Zero-Out
Peak: 8.88 ppb
99th Percentile: 1.06 ppb
Median: 0.04 ppb

Industrial Contribution (ppb)

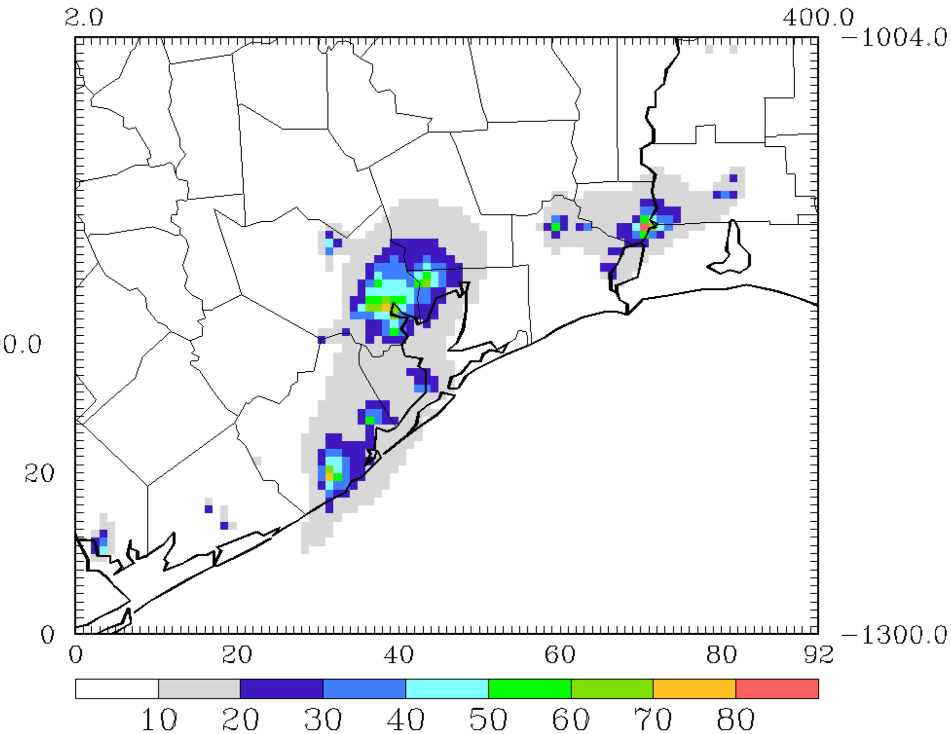
RTRAC
Peak: 8.89 ppb



Modeling Results (August)

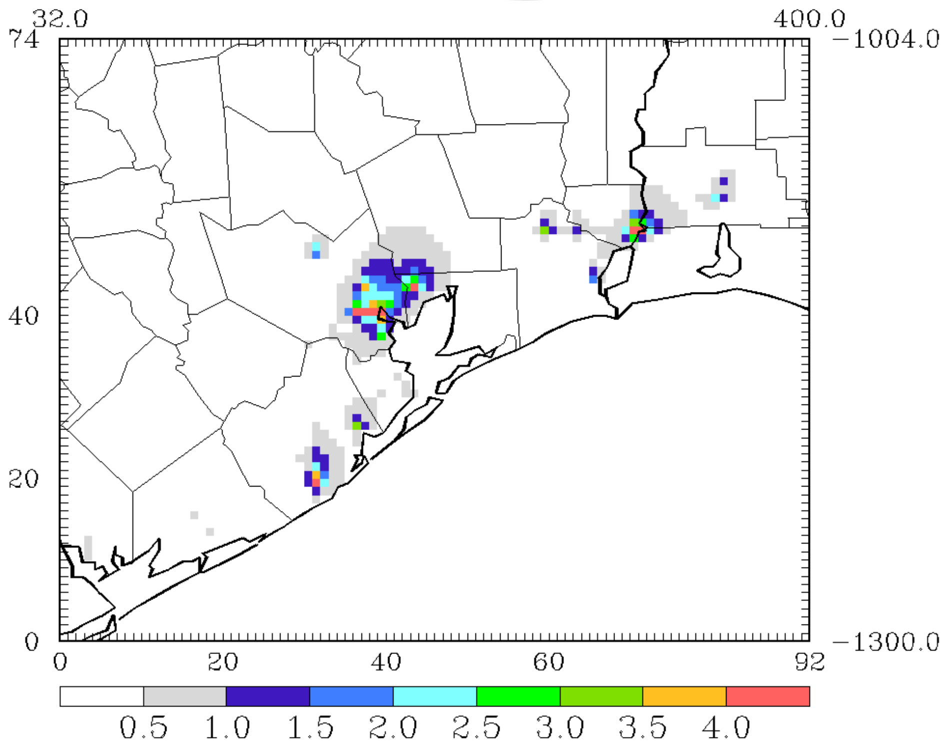


Percentage Contribution
Peak: 81.2%



Base Concentration
Peak: 123.0 ppb
99th Percentile: 7.10 ppb
Median: 2.80 ppb

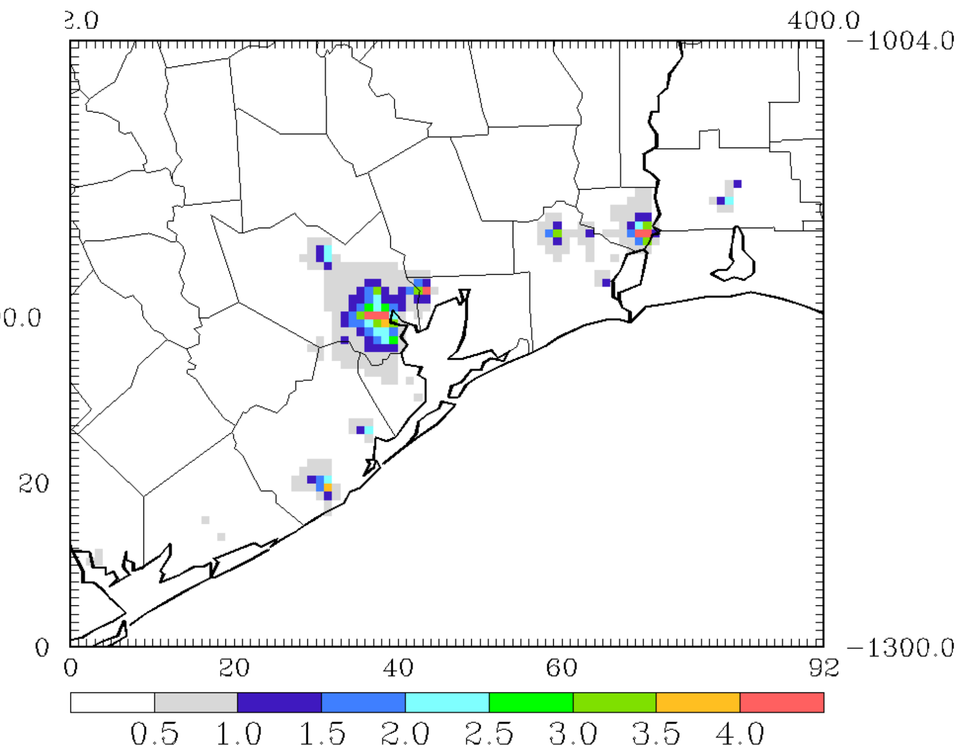
Modeling Results (Aug. Conc)



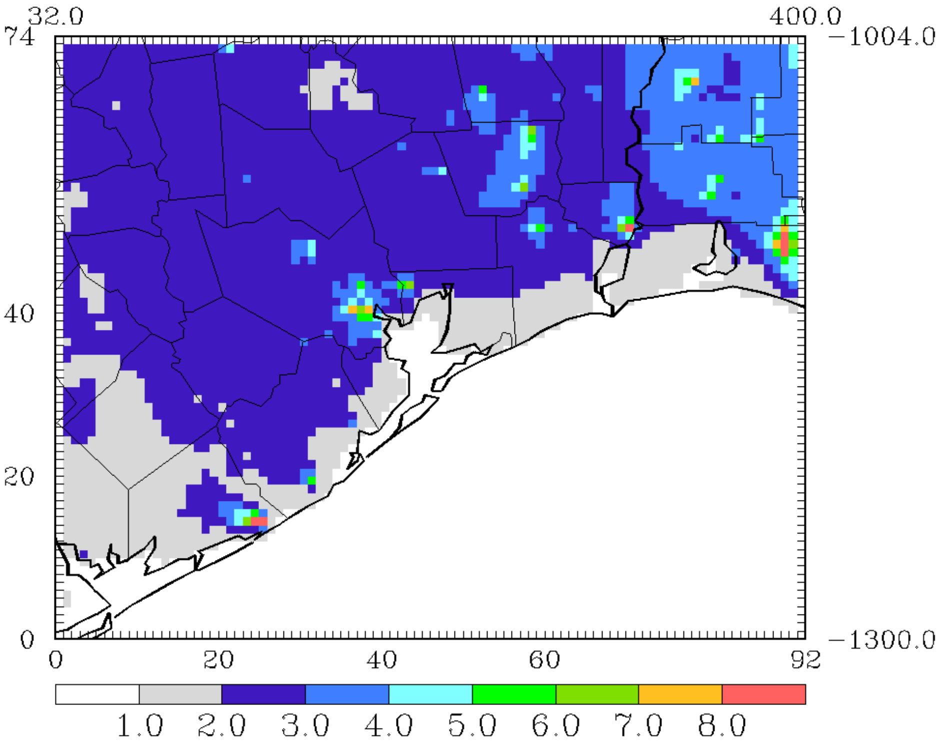
Zero-Out
Peak: 11.2 ppb
99th Percentile: 1.46 ppb
Median: 0.05 ppb

Industrial Contribution (ppb)

RTRAC
Peak: 11.2 ppb

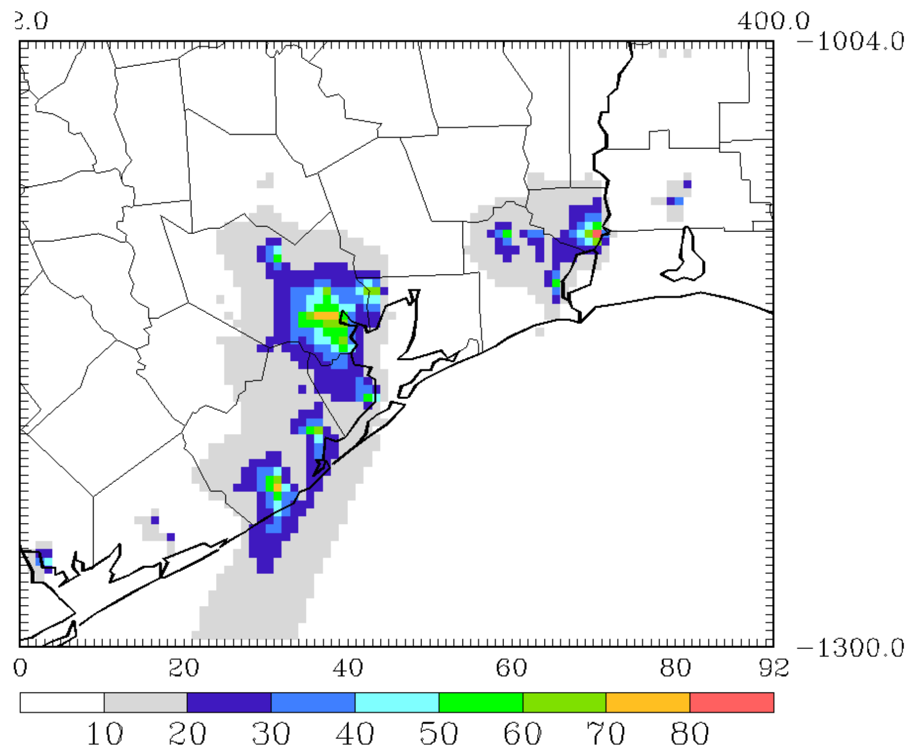


Modeling Results (September

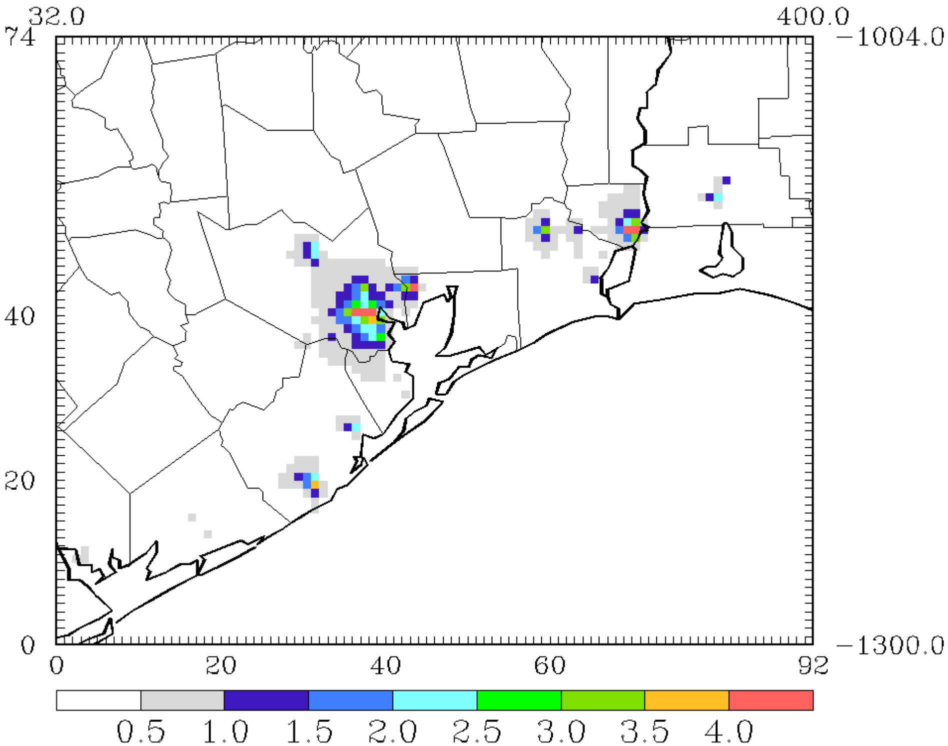


Base Concentration
Peak: 25.3 ppb

Percentage Contribution
Peak: 83.5%



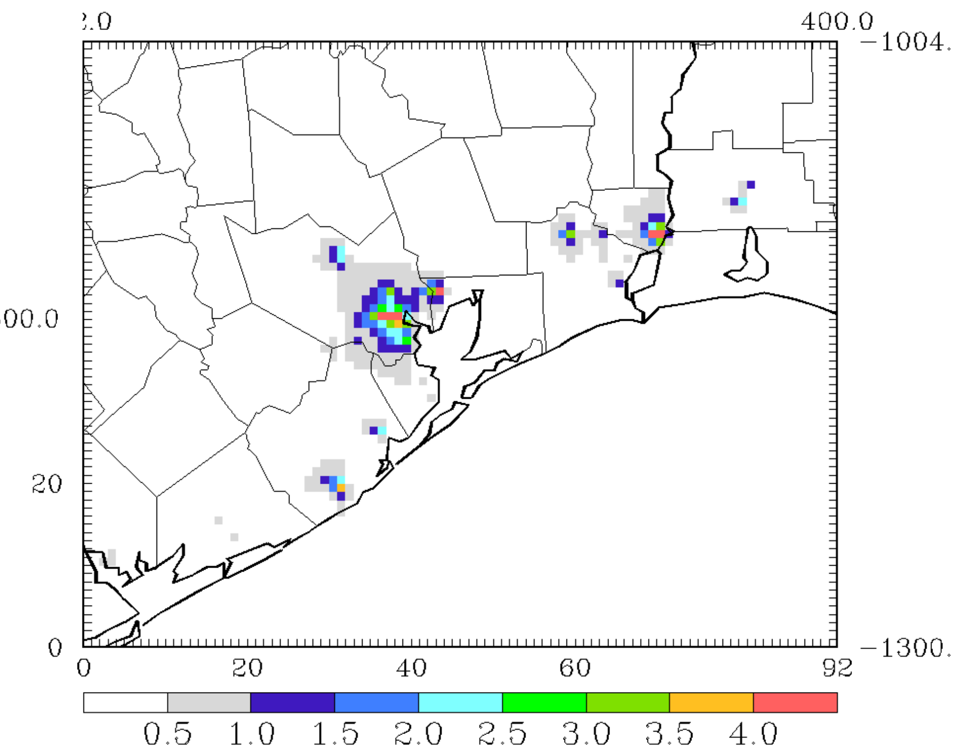
Modeling Results (Sep. Conc.)



Zero-Out
Peak: 10.4 ppb
99th Percentile: 1.24 ppb
Median: 0.06 ppb

Industrial Contribution (ppb)

RTRAC
Peak: 10.4 ppb



Observations

- Industrial ethylene impacts > 0.5 ppb are fairly localized in industrial areas
- Zero-out and RTRAC results agree very closely
- Significant month to month variation in concentrations
  - Likely both meteorologically and emissions driven
- Impact distributions very long tailed
- Industrial contribution exceeds 20% fairly close to industrial source regions