

A Comparison of Ambient and Emissions VOC to NO_x Ratios at Two Monitors in Houston, Texas

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ABSTRACT

In an effort to validate the VOC inventory, VOC to NO_x ratios for 13 different VOC classes were calculated for emissions around the Deer Park and Clinton monitors located in the Houston, Texas area. These were compared to the corresponding ambient ratios using a “polar grid” approach, whereby ambient data from “wind bins” of ten degrees were compared to emissions data from the same direction. Emissions from both point sources, and non-point sources – including area, mobile, non-road, and ship combustion categories – were included in this comparison. “Wedges” consisting of several contiguous bins at each monitor were created to compare the accuracy of the non-point and point source inventories. The results of this comparison suggest that the non-point source VOC inventory is more accurate than the point source VOC inventory. Additional wedges were set up for specific VOC classes in certain directions where point source emissions were a strong influence. The results of this comparison suggest that the degree of underestimation of the VOC inventory, depending on the VOC class and particular wedges being studied, ranges from near zero (i.e. ambient:inventory unity) to about 9-fold, though numerous uncertainties in this study’s methodology make it difficult to assign any confidence to these figures; more work is needed to better understand this discrepancy.

INTRODUCTION

As a means of validating emission inventories, researchers have calculated ratios of different airborne compounds measured at one or more monitors, and compared these to the same ratios from nearby emissions sources.^{1,2,3} Typically the emissions used in these studies were spatially allocated in a two-dimensional cartesian grid; ratios were calculated for different combinations of grid cells, such as those cells surrounding the monitor, the single cell that includes the monitor, and cells located in the upwind quadrant. Although there are many uncertainties associated with such a comparison, the general idea behind this methodology is that if the emission inventory is approximately accurate, the ratio of its compound ratios to the same ratios measured at adjacent monitor(s) should be approximately 1:1.

In the heavily industrialized area in and near Houston, Texas, there are numerous plants that emit large amounts of VOCs and NO_x, among other pollutants. These sources are geographically distributed in an irregular manner along the Houston Ship Channel; the use of a standard emissions grid for comparison with ambient measurements may be too coarse to capture the significant emissions variability that can occur over a very short distance in the area. In a study that assessed the accuracy of emissions estimates of several very reactive VOCs in the area⁴, researchers grouped ambient data at the study monitors by wind direction, using a resolution of 10 degrees to demarcate bins. Ratios of the different reactive VOCs to NO_x measured at each bin were compared to those from upwind point emission sources.

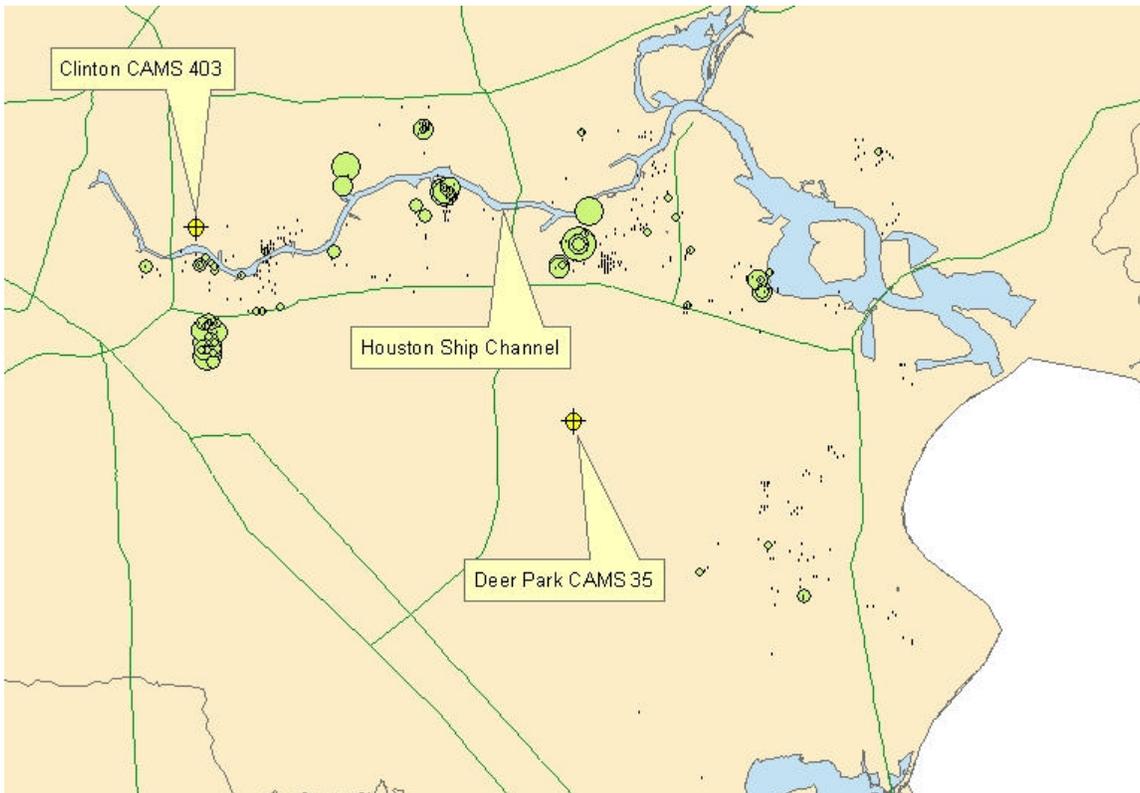
The present study follows this approach, and expands it significantly. Research done during the Texas Air Quality Study in 2000 (TexAQS 2000) has shown that emissions of two very reactive alkene species were strongly underestimated at isolated industrial facilities in the area, and that NO_x emissions estimates for the same facilities were relatively accurate.⁵ Given this, we use ambient and emissions VOC:NO_x ratios to estimate the relative accuracy of the VOC emission inventory, assessing a host of different VOC species by grouping them into 13 different VOC classes. Furthermore, we compare

ratios when air passed over areas concentrated in point source against ratios when the air traveled over areas devoid of point sources, in order to assess the relative accuracies of the non-point source and point source VOC inventories.

METHODS

Ambient data were collected at the Deer Park CAMS 35 and Clinton CAMS 403 monitors during the years 1999-2001 and analyzed for comparison with the emission inventory. (Figure 1 shows the location of these monitors.) Only data from the early morning hours, 0500 through 0659 local standard time (lst), were used for comparison, in order to minimize loss of hydrocarbons to reaction. Data collected included 40-minute average concentrations, sampled hourly, of approximately 55 different hydrocarbon species, measured using automated gas chromatographs. As well, hourly wind direction and wind speed averages, and hourly average NO_x concentrations, were collected.

Figure 1. Locations of Clinton and Deer Park monitors, with point source emissions of 1,3-butadiene shown as green circles.



Only validated NO_x, hydrocarbon, and meteorological data were included. Hours with NO_x measurements less than 10 ppbv, or wind speed less than 2 miles/hour, were excluded. In order to facilitate the comparison of ambient and emissions VOC to NO_x ratios for a broad range of VOCs, the majority of the hydrocarbon species were grouped into one of thirteen groups prior to calculating these ratios (see Table 1).

Table 1. VOC group names and constituents.

VOC Group Names and Constituents	
Group Name	Constituent(s)
butadiene	1,3-butadiene
butanes	isobutane, n-butane
butenes	t-2-butene, c-2-butene, 1-butene
c5cyclos	cyclopentane, cyclopentene
c6arom	benzene
c6cyclos	methylcyclopentane, cyclohexane
c7c10arom	m/p-xylene, o-xylene, m-ethyltoluene, p-ethyltoluene, o-ethyltoluene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, 1,2,3-trimethylbenzene, toluene, ethylbenzene, isopropylbenzene, n-propylbenzene, m-diethylbenzene, p-diethylbenzene, styrene
c7c11other	methylcyclohexane, 2,4-dimethylpentane, 2-methylhexane, 2,3-dimethylpentane, 3-methylhexane, n-heptane, 2,2,4-trimethylpentane, 2,3,4-trimethylpentane, 2-methylheptane, 3-methylheptane, n-octane, n-nonane, n-decane, n-undecane
ethene	ethene
hexanes	n-hexane, 2,2-dimethylbutane, 3-methylpentane, 2,3-dimethylbutane, 2-methylpentane
pentanes	isopentane, n-pentane
pentenes	2-methyl-2-butene, t-2-pentene, 3-methyl-1-butene, 1-pentene, c-2-pentene
propene	propene

For each monitor, the ambient data were split by wind direction into one of 36 “wind bins,” with each bin comprising 10 degrees. Bins were ordered in clockwise fashion starting at zero compass degrees. For example, bin one covered zero through nine compass degrees, and bin 36 covered 350-359 degrees. Ambient VOC:NO_x ratios for a bin were calculated by taking the median VOC:NO_x ratio of all the data points in a bin.

For each wind bin at each monitor, VOC:NO_x ratios for each of the thirteen VOC groups were calculated. These were compared to the same ratios calculated for the emissions sources located within each bin. The ratio of the two ratios was then calculated. Figure 2 shows the results of this for butadiene at Clinton. Emissions sources up to 14 km from a monitor were included in the calculations. The resulting study area formed a polar grid around each monitor. Figure 3 shows the study area around the two monitors, the polar grid around the Deer Park monitor, and propene point source emissions.

Figure 2. Ratio of median ambient butadiene:NOx to emissions butadiene:NOx at Clinton.

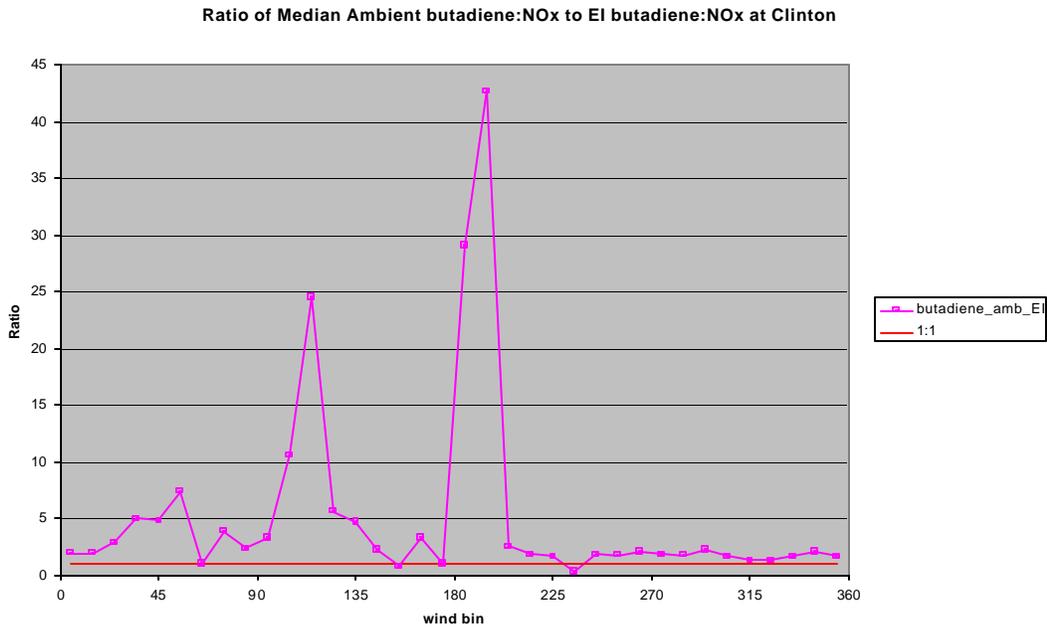
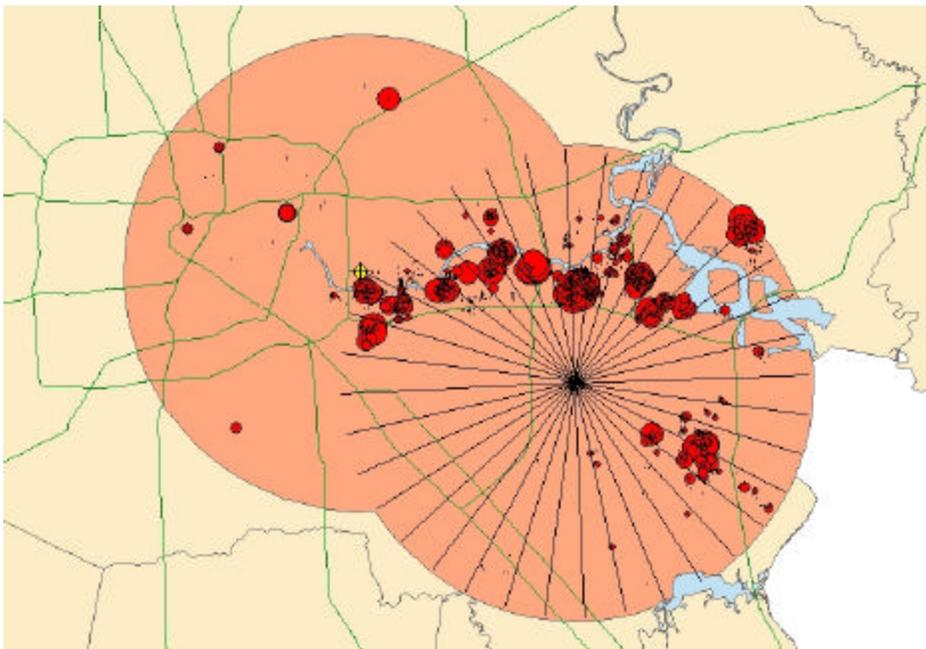


Figure 3. Clinton (yellow icon) and Deer Park monitors, with Deer Park grid, and propene point source emissions depicted as red circles.



Point source emissions were taken from version 15a of the TCEQ speciated emissions inventory. This inventory consists of speciated emissions by hour and by emission point, for the period 8/18/2000 – 9/6/2000. Emissions in this inventory are a combination of hourly emissions rates, for those plants that reported them, and daily emissions rates, for the remaining plants. Only point source emissions from 0500-0659 1st on 8/29/2000 were used. For those companies reporting daily emission rates, emissions for this two-hour period were calculated by dividing the daily emission rate by 12. Emissions from each

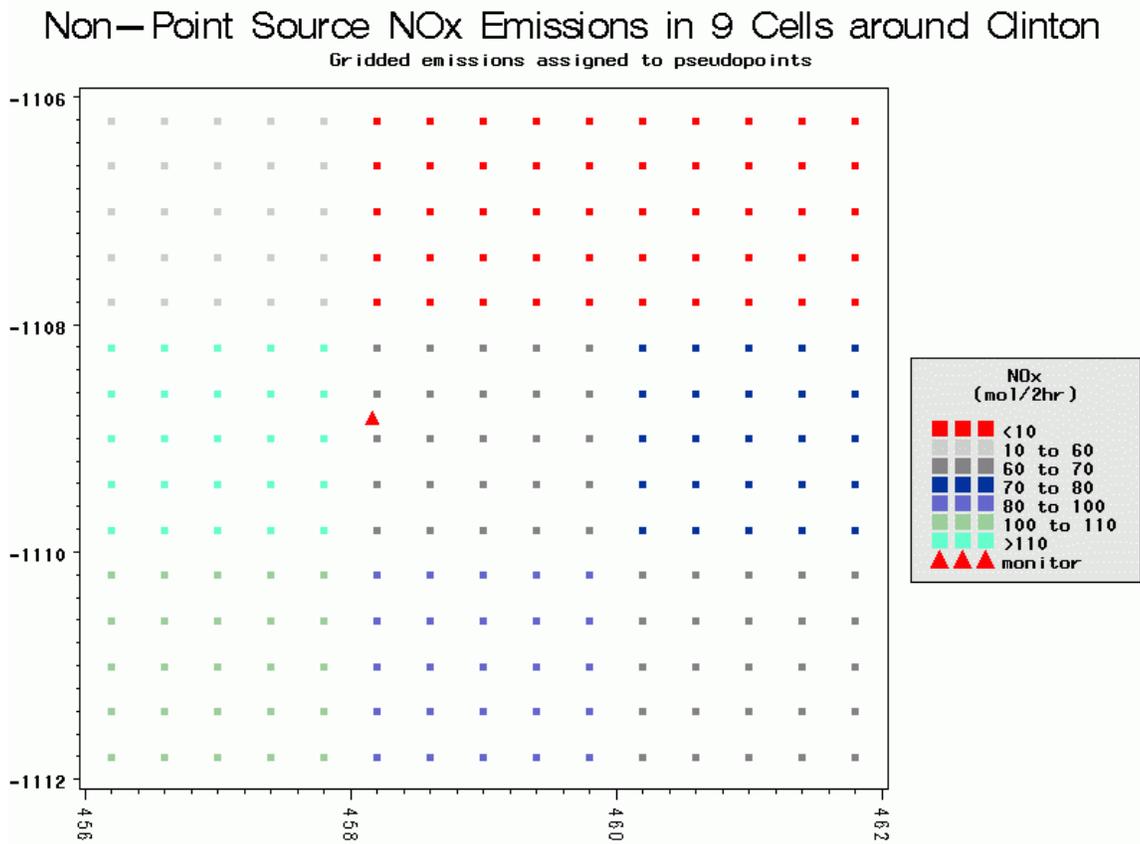
and every emission point in the study grid were weighted by the inverse distance between the emission point and the monitor before calculating VOC:NO_x ratios.

Emissions from area, mobile, non-road, and ship combustion categories were also included in this study. (Table 2 shows each of the component inventories used in this study.) The data for these categories originally existed in a 22 by 26 cell cartesian grid, consisting of 2x2 km grid cells, including the area covered in this study, but extending far beyond it. In order to convert these emissions to a polar grid format, the following procedure was followed. Emissions for each category in each cell were divided by 25, with the resulting fraction evenly allocated to 25 evenly spaced “pseudopoints” within the cell. Figure 4 depicts this for the nine cells closest to Clinton; the monitor itself is shown as a red triangle. As with the true point sources, pseudopoint emissions were weighted by the inverse distance between point and monitor before calculating ratios for the different VOC groups.

Table 2. Emission Inventory categories, sources, and dates of categories.

Category	Source	Inventory Date
Mobile	Mobile6	8/29/2000
Area	Base 1	8/31/1999
Non-road	Base 1	8/31/1999
Ships	Base 97	8/31/1999
Point – Low NEGU – Upsets	Upset1	8/29/2000
Point – Low NEGU – Routine Emissions	Special Inventory 4	8/29/2000
Point – Low EGU – Routine Emissions	Special Inventory 4	8/29/2000
Point – Elevated NEGU – Upsets	Upset1	8/29/2000
Point – Elevated NEGU – Routine Emissions	Special Inventory 4	8/29/2000
Point – Elevated EGU – Routine Emissions	Special Inventory 4	8/29/2000

Figure 4. Non-point source emissions in 9 cells closest to Clinton.



RESULTS

VOC emissions from point sources in the eight-county Houston ozone nonattainment area compose about 28% of the total anthropogenic VOC inventory⁶. However, these sources are concentrated in certain areas, and are sparse or nonexistent in other areas. Around both of the present study monitors are areas with concentrated point source emissions, and other areas dominated by non-point sources.

In this study, we attempted to evaluate the relative accuracy of the point versus non-point emissions inventories by creating “wedges,” which are groups of contiguous wind bins, where either point or non-point sources dominate. Table 3 shows three wedges – two at Clinton, one at Deer Park – that were set up for this purpose. The wedges were assigned names according to the monitor they are based at, and the bins they span.

The results in Table 3 show a comparison of the three wedges’ ambient to EI VOC:NOx ratios. Two of the wedges, Deer Park 215-255 and Clinton 255-285, are dominated by non-point source VOCs. These wedges were selected in part because each one has a “buffer” of at least one bin on either side of the wedge that, like the bins in the wedge itself, had little or no point source emissions. This buffer helped to minimize the effect of veering winds bringing point source emissions from outside the wedge into it.

The third wedge, Clinton 75-105, which points to the east, was dominated by point sources. As can be seen in Figure 5, there are substantial point source emissions also to the east of Clinton, but beyond the study boundary. Though these emissions may travel to the monitor, they were not accounted for in this study.

Table 3 shows that at each of these three wedges, the respective non-point source or point source dominance was true for each of the thirteen VOC classes. It also shows that the two non-point-dominated wedges' ratio of ratios were 2.1 and 3.4, considerably less than the value seen at the point-dominated wedge. This suggests that while the non-point VOC inventory is probably underestimated, the point VOC inventory may be underestimated even more.

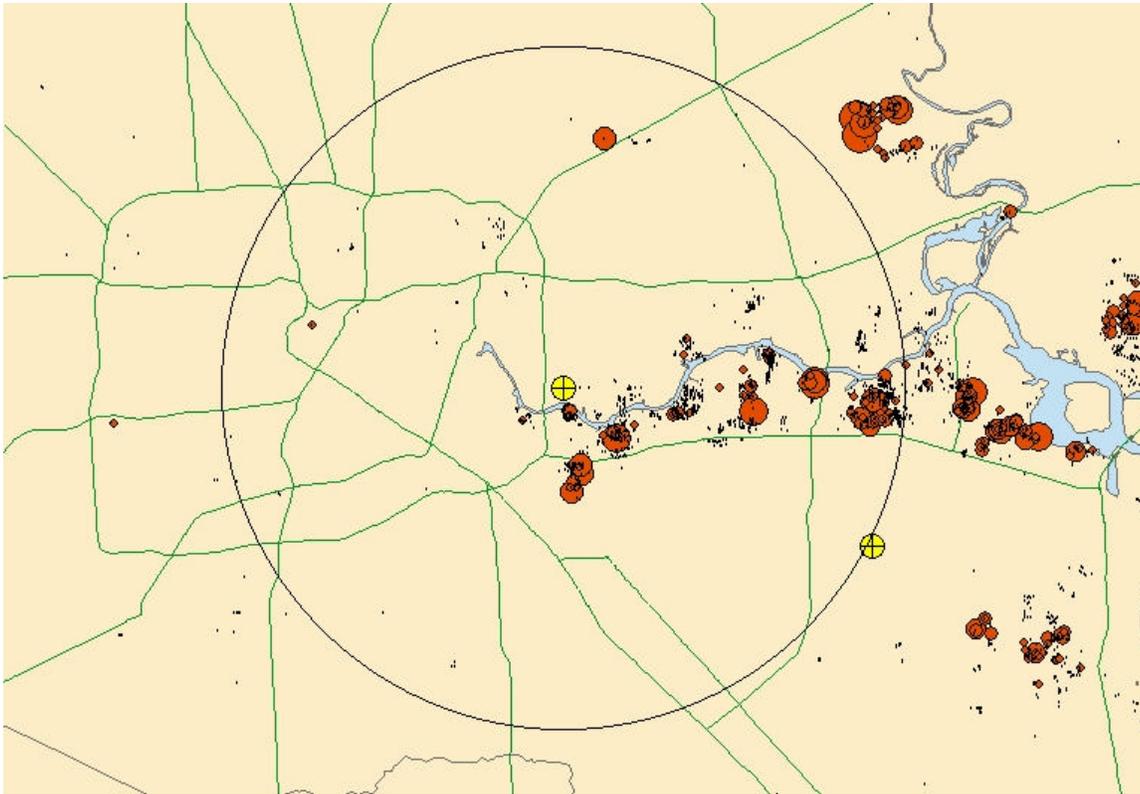
It can also be seen from Table 3 that nearly every VOC group's ambient to EI ratio, at either of the non-point dominated wedges, was less, and in most cases much less, than the same ratios at the point-dominated wedge. This further suggests that the non-point source VOC inventory may be more accurate than the point source VOC inventory.

One VOC group that defied this trend is butanes. Its ambient to EI ratio at the point dominated wedge was 8.2, as compared to 5.6 and 9.2 at the two non-point dominated wedges. This suggests that both the point and non-point butanes inventories are strongly underestimated.

Table 3. Point Source Dominated and Non-Point Source Dominated Wedges.

VOC Group	Point-Dominated		Non-Point-Dominated				mean nonpoint Ambient:EI Ratio
	Clinton 75-105		Clinton 255-285		Deer Park 215-255		
	Ambient:EI Ratio	pct point	Ambient:EI Ratio	pct nonpoint	Ambient:EI Ratio	pct nonpoint	
C5cyclos	6.4	92.2	1.2	100.0	2.8	100.0	2.0
C6arom	3.7	95.6	2.2	98.4	3.3	100.0	2.8
C6cyclos	6.5	94.9	2.3	99.6	5.7	100.0	4.0
C7C10arom	3.6	83.8	1.7	99.8	2.3	99.7	2.0
C7C11other	4.8	81.2	1.3	99.7	1.7	100.0	1.5
butadiene	3.8	91.9	1.9	100.0	2.0	100.0	2.0
butanes	8.2	97.4	5.6	98.4	9.2	100.0	7.4
butenes	7.7	95.3	1.8	96.8	1.8	100.0	1.8
ethene	9.0	90.9	2.0	99.7	3.9	100.0	3.0
hexanes	4.9	95.3	2.0	98.2	3.6	100.0	2.8
pentanes	5.8	95.7	1.5	99.3	2.4	100.0	1.9
pentenes	2.5	93.3	0.6	99.1	0.7	100.0	0.7
propene	7.6	97.6	3.6	95.1	4.7	100.0	4.2
mean	5.7		2.1		3.4		2.8

Figure 5. Clinton and Deer Park monitors (yellow icons), with Clinton grid boundary, and combined point source emissions of ethene, propene, butadiene, and butenes.



In addition to the wedges shown in Table 3, a series of additional wedges were created to compare ambient and inventory ratios in areas where maps indicated that point source emissions would strongly influence ambient concentrations. These wedges, along with their resulting ambient to EI ratios, and the percent of each wedge's emissions attributable to point sources, can be seen in Table 4 and Table 5. Note that the bins forming these wedges vary across the different VOC groups, because the plants they include may emit large amounts of one or more VOCs, without necessarily emitting large amounts of others.

It can be seen from Table 4 that at Clinton, 13 of the 15 additional wedges' ambient to EI ratios fell between 1.3 and 4.7. However, wedges of butenes and C5cyclos to the southeast of Clinton resulted in ratios of 10.1 and 6.8, respectively. Table 5 shows that at Deer Park, where 27 additional wedges were created, 23 of these had ratios between 0.7 and 7.2. However, two of the wedges pointing to the north (C5cyclos_355_5 and ethene_355_5) showed ratios of 13.6 and 11.9, respectively. As well, wedges pointing to the northeast (C6cyclos_25_55) and southeast (butenes_125_135) produced ratios (12.1 and 10.0, respectively) that also exceeded this range.

Table 4. Wedges around Clinton influenced by point sources.

Wedge	Ambient:EI Ratio	pct_point
cl_C5cyclos_125_195	6.8	45.5
cl_C6arom_135_185	1.4	89.4
cl_C6cyclos_115_215	2.8	84.0
cl_C7C10arom_125_145	2.6	76.7
cl_C7C10arom_155_185	3.3	65.8
cl_C7C11other_125_195	3.7	56.2
cl_butadiene_165_195	3.7	91.1
cl_butenes_135_195	10.1	72.6
cl_ethene_115_145	4.6	86.6
cl_ethene_155_195	1.3	85.8
cl_hexanes_125_195	2.5	85.5
cl_pentanes_115_185	4.3	82.9
cl_pentenes_125_185	4.7	42.7
cl_propene_135_155	1.7	84.2
cl_propene_165_195	3.5	89.8
mean	3.8	

Table 5. Wedges around Deer Park influenced by point source emissions.

Deer Park "Wedges" Influenced by Point Source Emissions		
Wedge	Ambient:EI Ratio	pct_point
dp_C5cyclos_355_5	13.6	81.2
dp_C6arom_105_125	5.7	85.5
dp_C6arom_355_25	5.7	95.2
dp_C6arom_45_55	2.1	95.8
dp_C6cyclos_125_145	3.8	85.5
dp_C6cyclos_25_55	12.1	86.7
dp_C6cyclos_355_15	7.2	89.3
dp_C7C10arom_355_5	3.7	85.2
dp_C7C11other_355_15	5.6	71.0
dp_C7C11other_45_55	3.0	73.3
dp_butadiene_355_5	6.1	95.3
dp_butadiene_55	0.7	95.2
dp_butenes_125_135	10.0	66.2
dp_butenes_355_5	7.1	93.9
dp_butenes_35_55	5.1	90.3
dp_ethene_105_125	3.5	94.4
dp_ethene_25_65	3.9	96.4
dp_ethene_355_5	11.9	91.7
dp_hexanes_345_5	4.0	90.9
dp_hexanes_35_55	3.0	94.4
dp_pentanes_345_5	4.4	90.0
dp_pentanes_45_55	4.7	92.2
dp_pentenes_345_5	1.3	86.7
dp_pentenes_45_55	2.4	81.1
dp_propene_115_125	2.1	98.5
dp_propene_25_55	6.2	98.9
dp_propene_355_5	5.8	98.2
mean	5.4	

DISCUSSION

If these ratios can be taken to measure the degree of inaccuracy of the VOC inventory in the area around these monitors, they suggest that for most VOC groups in most directions, estimates of inventory accuracy range from slightly overestimated (e.g. butadienes_55 at Deer Park with a ratio of ratios of 0.7) to underestimated by about 9 fold. They also suggest that the non-point source inventory is more accurate than the point source inventory.

The degree of VOC underestimation calculated here is less than that found in a different study conducted in the area. In a study which used aircraft to conduct plume transects around three isolated clusters of petrochemical manufacturing facilities in an adjacent county, researchers compared ethene:Nox and propene:Nox ratios calculated using emission inventory data versus those inferred from plume data, and found that the emission inventory figures for these two compounds were underestimated by about 20-70 fold. By comparison, the present study found, in the wedge that was consistently point-dominated for all VOCs (Clinton 75-105), that the ambient ethene:Nox was nine times greater than its inventory counterpart, and the ambient propene:Nox was 7.6 times greater than the same ratio using emissions data.

In a study which compared VOC to NOx ratios at and around seven monitors in the Ship Channel area⁴, researchers calculated discrepancies between ambient and inventory ratios for ethene, propene, 1,3-butadiene, and butenes (sum of 1-butene, t-2-butene, and c-2-butene). This study found discrepancies from 0.95 to 12 for ethene, 1.9 to 16 for propene, 0.46 to 19 for butenes, and 0.18 to 96 for butadiene. As this study used a wind bin approach similar to the present study, and it included only point source emissions, its results are worth comparing to those from the point source influenced bins in the present study. When the results from the point source dominated wedge in Table 3 are combined with those in Tables 4 and 5, ranges for the same 4 compounds can be determined. They are 1.3 to 11.9 for ethene, 1.7 to 7.6 for propene, 5.1 to 10.1 for butenes, and 0.7 to 6.1 for butadiene. Each of the ranges for the four compounds in the present study fall inside the ranges of the earlier one.

There are numerous uncertainties in the present study's methodology that must be addressed. One of its bases is that winds from a particular direction bring air exclusively from that direction – that the winds do not curve. But numerous trajectories performed in the Houston area have shown that curving winds are commonplace. No trajectory analysis was performed here. Figure 6 illustrates this curvature: the dominant bin for butadiene emissions at Clinton, bin 175, points slightly east of south, but the peak median ambient concentration of butadiene was observed at bin 195, 10-20 degrees to the west. As there are no other butadiene emissions sources remotely as large as the cluster in bin 175, there appears to be no other butadiene sources that could account for the strong measured peak seen in bin 195.

By including “buffers” of at least one bin on either side of the two non-point dominated wedges, we attempted to mitigate against the possibility of curving winds bringing point source emissions into these wedges, and thus biasing the results. We also attempted to eliminate hours with curving winds by excluding slow wind speed hours from analysis. Nevertheless, Figure 6 shows that winds can veer across two bins, and even with the exclusion of the slowest winds, it is possible that they may veer even more than that.

Another area of uncertainty is that emissions outside the 14 km radius were excluded from analysis. Aircraft flights performed during TexAQS 2000 tracked emissions plumes, and their reaction products, for at least four hours⁵ downwind of their sources; even at that distance, emissions, especially the less-reactive VOCs, were observed. This four hour path, under typical conditions, could easily translate to a distance of 30 or more kilometers, well beyond the study grid boundary. In all likelihood, both NOx and VOC emissions from outside the boundary were measured at these monitors; this makes it difficult to estimate any bias in results due to outside emissions.

This study did not account for loss of emissions due to reaction. While the exclusive use of early morning hours helps to minimize this, it does not eliminate it.

Another factor worth discussing is the effect of variable mixing heights and how this affects inclusion of emission sources. As this study only looked at data from the early morning hours (0500-0659 1st), this suggests at first glance that the mixing heights at those hours should be relatively low, and source inclusion should reflect that. Indeed, the first set of analyses conducted for this study used only sources with effective plume rises of 30 meters or less.

However, thanks to the use of year round data, mixing heights during the 2-hour study period probably varied greatly. In wintertime, sunrise is as late as 0713 hours, whereas in summer it's as early as 0521 hours. During TexAQS 2000, which occurred during late August and early September, sunrise ranged from 0552 to 0601 hours. Radar profiler data from that study showed mixing heights at 0700 ranged from 210 to 320 meters, and mixing heights at 0800 – about two hours after sunrise – ranged from 240 to 608 meters. Given that the present study's two-hour period included data, in summer, that was collected nearly two hours after sunrise, it was decided to use a value of 600 meters for effective plume rise as the cutoff for source inclusion, for the majority of analysis conducted in this study. This provided a conservative "upper bound" for analysis; unfortunately, for the results presented here, the corresponding "lower bound" (30 m effective plume rise cutoff) has not yet been performed, but should be performed in the near future, to address this cause of uncertainty.

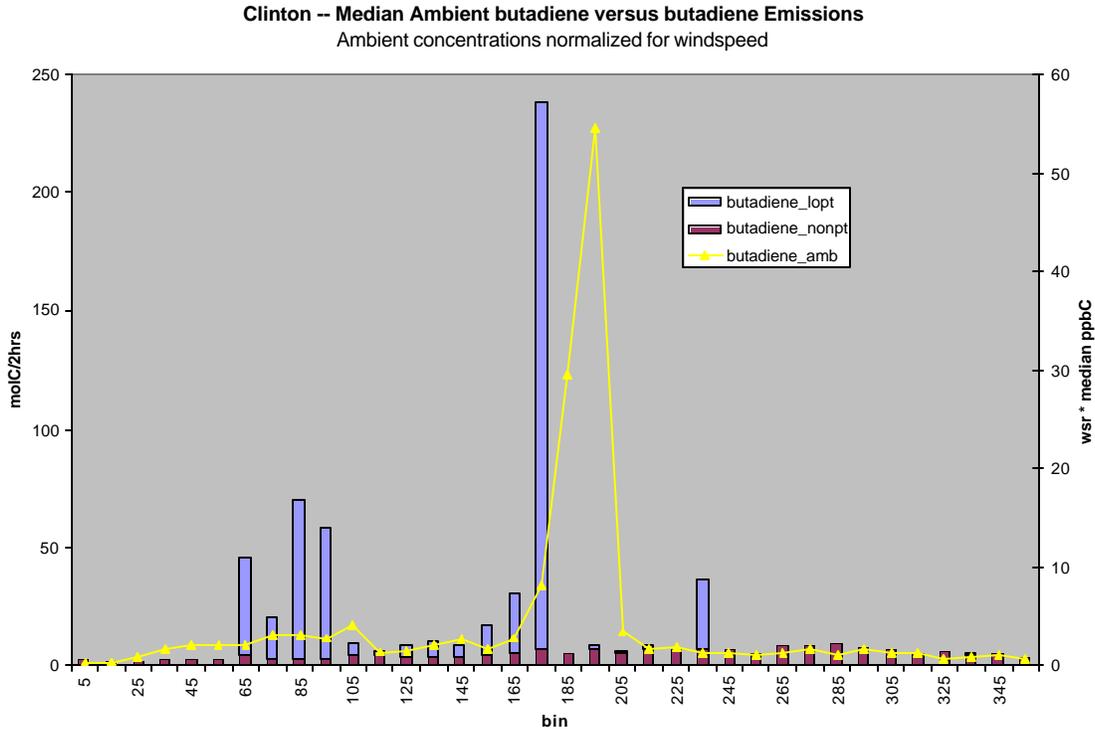
Another source of uncertainty is that the point source emissions inventory data used here comes from the companies themselves, and the VOCs that are reported are sometimes only partly speciated, or not at all. In such cases, TCEQ photochemical modeling staff have developed protocols for speciating the unspciated portion of VOCs⁷. As a result of applying these protocols, unspciated VOCs are sometimes speciated into groups, such as "isomers of hexane," rather than individual species. Of course, such groups are not included in the list of 55 or so PAMS species measured by the Auto-GCs at Clinton and Deer Park, yet the unspciated VOCs that they originate from probably include at least some species that are measured at the Auto-GCs, but are not reported in the inventory. To rectify this potential source of bias, for some of the VOC groups, some of these partly-speciated VOCs were included in the emissions. These include:

- 1) "isomers of pentane" (SAROAD 43122) in Pentanes;
- 2) "isomers of pentene" (SAROAD 43121, 43143) in Pentenes;
- 3) C7 paraffins (SAROAD 43142), C8 paraffins (SAROAD 43141), and C10 paraffins (SAROAD 43135) in C7C11other;
- 4) 8 additional codes in C7C10arom

Spot checks of emissions data for these VOC groups suggested that the effect of including these codes was small, but no systematic check was performed.

Finally, it is notable that butanes ratio discrepancies were large both in non-point and point dominated wedges. One factor which may influence this is that Houston-area motorists refuel with "summertime RFG (reformulated gasoline)" between May 1 and September 15, and the normal RFG at other times. Requirements for Reid Vapor Pressure (RVP) and other parameters differ between the two classes. Although refiners are not required to follow prescriptive formulas in developing their gasoline, in all likelihood, overall n-butane and isobutane content differs between the two seasons. As well, "Phase 1 RFG" was used through 1999, and Phase 2 RFG replaced it starting 1/1/2000. It is hoped that these two refinements can be included when this study is updated.

Figure 6. Median ambient butadiene concentration (normalized for wind speed), and butadiene emissions, by wind bin.



CONCLUSIONS

Ratios of ambient to emission inventory VOC:NO_x ratios, for 13 different VOC classes around the two monitors, suggest that for most VOCs, the emission inventory ranges from being approximately accurate to being underestimated up to a factor of 9, and in a few cases higher. When ratios were calculated for two non-point source dominated areas and compared with those from a point source dominated area, the results showed that the degree of inventory underestimation in the point dominated area, 5.6, was about twice the same figure for the non-point dominated areas, when their values were averaged, suggesting that the non-point VOC inventory is, on average, underestimated by about half the amount that the point source inventory is.

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