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Mapping of Air Pollution Levels in Tallinn

Measurement campaigns with diffusive samplers

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Summary

As a part of an Air Quality Management project in Tallinn, measurement campaigns in order to generate data with high spatial resolution have been performed. In Tallinn, databases containing high time resolution data from a few locations already exist, and these two measurement databases will together provide a good basis for future validation of dispersion model results.

Measurements of SO₂, NO₂ and VOC were carried out using diffusive samplers and were organised as a total of 12 monthly campaigns during the period from April 1996 to January 1998. The diffusive samplers give average air concentrations with continuous time coverage from a few days up to a month. The technique is based on the concept of molecular diffusion, where the gas molecules diffuse into a sampler where they are quantitatively collected. The samplers are easy to handle, and the method does not require electricity or calibration in the field.

In order to map concentration levels of the measured pollutants in different typical local environments within the urban area of Tallinn, a classification of sites was made according to exposure to road vehicle emissions and street ventilation conditions. Four different categories of sites, from busy street environment with poor ventilation to local background conditions representing a general pollution level were classified. In addition to these, measurements at background locations were also made. A total of between 20-80 sites of different classification were used simultaneously during the campaign measurements.

The measured monthly levels of SO₂ and NO₂ were, as expected, different at the different site categories. In the table below, the results from all 12 measurement campaigns are summarized.

| | NO ₂ , monthly averages, µg/m ³ | | | SO ₂ , monthly averages, µg/m ³ | | |
|----------------------------------|---|---------|-----|---|---------|-----|
| | Max | Average | Min | Max | Average | Min |
| Busy street, poor ventilation | 48 | 36 | 27 | 15 | 10 | 4 |
| Busy street, good ventilation | 50 | 29 | 10 | 21 | 8 | 1 |
| Residential area | 33 | 18 | 6 | 19 | 7 | 1 |
| Local background | 30 | 14 | 5 | 17 | 6 | 1 |
| Background | 16 | 8 | 1 | 12 | 4 | 1 |

There were larger differences in concentration levels between the site categories for NO₂ than for SO₂. The different concentration patterns for the two components are probably mainly due to the emission situation. NO₂ is mostly generated by traffic, hence the highest concentrations will be found close to roads with high traffic intensity. The sources of SO₂ can be assumed to be more widespread within the area studied, since a major part of the SO₂ emissions originate from fossil fuel combustion in large power plants as well as in local residential heating devices.

There was a seasonality of higher concentration levels in wintertime than during summer of SO₂ at all site categories, and for NO₂ at the less exposed sites. At the two most exposed site categories no evident seasonality in NO₂ levels was detected.

The average levels of VOC, all measured at busy street environments with good ventilation, ranged from 19-95 µg/m³ of total VOC (eight components) at the different sites during four campaign months. The corresponding levels of benzene were between 3-13 µg/m³.

A comparison of the measured levels of pollutants in Tallinn to Swedish conditions show that in Tallinn the SO₂-levels are higher while the levels of NO₂ are comparable to those in Stockholm and Gothenburg. Regarding VOC, although there are not any exactly comparable measurements, some of the sites in Tallinn seem to be more exposed to VOC while others presumably would be comparable.

Although no regular annual measurements have been performed during these campaign measurements in Tallinn, the calculated average values from all campaigns would give a good indication of longtime average levels. In comparison to EU annual limit values it seems that for SO₂ the measured levels in Tallinn would be well below the limit value. For NO₂ the measured average concentrations at the most exposed sites were approximately 10% below the EU annual limit value for protection of health, but exceed an upper assessment threshold requiring measurements. There are presently no limit values within the EU regarding VOC. The expert group on air quality has however recently suggested an annual average for benzene of 5 µg/m³. This level would presumably be exceeded on an annual basis at several of the measurement sites in Tallinn.

From the results achieved it is obvious that the use of diffusive samplers, in combination with a careful selection of measurement sites, has been a fruitful strategy in mapping geographical variations in levels of air pollutants in Tallinn, an area of complex emission pattern. An important support in air quality management has been generated during a relatively short time period and at a reasonable cost. All results in combination have given an excellent insight into the variations of air pollution levels in the region as a whole as well as in different typical environments.

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1 Introduction

The Swedish Environmental Research Institute (IVL) has mapped air pollution levels of sulphur dioxide (SO₂), nitrogen dioxide (NO₂) and volatile organic compounds (VOC) in Tallinn. This project is a part of an Air Quality Management Project in Tallinn, Estonia, financially supported by the Swedish International Development Co-operation Agency (Sida). The Estonian party is the Environment Department, Tallinn City Government and the Swedish party is Conexor Sensus AB.

The main purpose of the measurement campaign was to generate data with high spatial resolution. The current databases, based on traditional measurement techniques contain high time resolution data from a few locations. An additional purpose of the campaign was to introduce a cost-effective air pollutant measurement technique in Estonia, demonstrating its potential to rapidly create an adequate background material for decision-makers. The two measurement databases will together provide a good basis for future validation of dispersion model results.

The measurements were carried out using diffusive samplers and were organised as a total of 12 monthly campaigns during the period from April 1996 to January 1998. Monthly samples of SO₂ and NO₂ were collected during all campaigns, while four weekly samples of VOC were collected during each of four of the campaigns.

The siting of measurement stations was based on a classification into four types according to exposure to road vehicle emissions and street ventilation conditions. The measurements were made at urban as well as at background sites. The Environment Department and IVL designed the station network in co-operation.

All practical matters related to the sampling have been accomplished by the Environment Department, Tallinn City Government. All chemical analysis of samples has been made at IVL.

2 Background

2.1 *Monitoring techniques*

Traditionally, air quality monitoring has been carried out using point, or lately, remote sensing measurements, often with a high time resolution (hours, days). Due to economical aspects this can only be made at rather few sites in a region.

In order to achieve a high spatial resolution of the concentration distribution of different air pollutants, the use of diffusive samplers has become an interesting tool. The diffusive samplers give average air concentrations with continuous time coverage from a few days up to a month. The technique offers a cost-effective method for air quality monitoring and the samplers are easy to handle.

The use of diffusive samplers, in combination with a careful selection of measurement sites, has been shown to be a fruitful strategy in mapping geographical variations in levels of air pollutants in a region with a complex emission pattern.

2.2 Tallinn

Tallinn is the capital of Estonia, located by the sea (the Gulf of Finland, a part of the Baltic Sea) in the north-western part of the country at latitude 59°25', longitude 24°45'. In 1996 the resident population in Tallinn was approximately 430 000 persons, which since 1990 has decreased by about 10%.

2.2.1 Emission structural changes

In the 1990's there has been great changes in emission structure, when emissions from traffic have increased while emissions from stationary sources have decreased due to closing down of many industrial plants. Point source emissions have decreased by 40% for NO₂ and by 70% for SO₂ between 1990-1995 because of the decreasing production in heavy industry (Tallinna Keskonnaamet, 1996). In the centre of Tallinn there are no big point sources. The large heating plants in the Tallinn area use natural gas and heavy oil as fuel, and they are located 2 km (Ulemiste) and 4 km (Mustamäe) from the centre (Sucksdorff-Selämaa, 1997).

The main air pollution source of nitrogen oxides and VOC in Tallinn at present is traffic. According to Tallinn Vehicle Registration Centre there were 154 604 personal cars, 1860 buses and 21 196 heavy vehicles registered in Tallinn in January 1998, which makes approximately 400 vehicles per 1000 inhabitants. Due to an increasing number of new cars, the usage of unleaded gasoline is increasing. The market rate for leaded gasoline is approximately 5-7%.

2.3 EU limit values for air pollutants

The EU directive 96/62/EG (1996) includes the basic principles for an EU strategy on air quality. In the directive it is stated that measurements have to be made in urban areas of more than 250 000 inhabitants, in zones where the limit values are exceeded and in zones where concentration levels are lower than the limit but exceed a certain upper assessment threshold, given as a percentage of the respective limit values for the pollutants.

In order to achieve as good knowledge as possible of the air quality in different parts of the respective countries, measurements and modelling should be combined. When a first assessment is completed, a combination of models and measurements can be used in areas where concentration levels are lower than the upper assessment threshold. If concentration levels are higher, measurements are required. If the levels are considerably lower, that is below the lower assessment threshold, modelling is enough to determine the air quality.

For the specific pollutants, daughter directives are worked out. In a first step, so-called position papers have been completed for sulphur dioxide, nitrogen oxides, particles and lead. Recently work has begun to establish limit values and to write position papers for benzene and carbonmonoxide. In appendix 1 the EU limit values for SO₂ and NO₂ are presented.

3 System architecture - Practical topics

3.1 Measurement methods

The diffusive sampling technique is based on the theory of molecular diffusion of gases, hence the term "diffusive sampling". The gas molecules diffuse into the sampler, where they are quantitatively collected on an impregnated filter or an adsorbent material, giving a concentration value integrated over time. No electricity, pump or other surrounding equipment is needed. Further, the samplers are small, light, cheap, soundless, re-usable and do not require electricity or calibration in the field.

Inorganic gases are absorbed by chemical reaction on a filter, impregnated with a solution specific to each pollutant measured. The reaction product, which is washed out of the filter prior to analysis, is specific to the particular gas in question. Organic gases, which do not react sufficiently fast with other chemicals, are instead trapped on an adsorbent material and subsequently desorbed thermally during the analysis.

The form of diffusive sampler used in this project for inorganic pollutants (SO₂ and NO₂) has been developed at IVL for measurements of ambient air pollutants. The sampler is 25 mm in diameter and 12 mm thick, see figure 1, mainly consisting of an impregnated filter and a diffusion barrier that keeps the sampling rate constant.

During transport and storage the sampler is placed in a plastic container. The sampling starts as soon as the container is opened and stops when the container is closed. The analyses are performed using ion chromatography for SO₂ and flow injection analysis for NO₂.

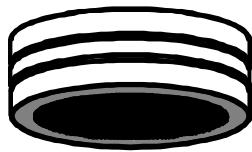


Figure 1 Diffusive sampler for SO₂ and NO₂.

Volatile organic compounds are collected using diffusive samplers consisting of a tube of stainless steel, packed with an adsorbent material. The tubes are conditioned and analysed prior to sampling. During storage and transport the tubes are capped in both ends. When starting the sampling, one of the caps is removed and replaced by a diffusion cap. To avoid the migration of water droplets from outside the cap into the front gauze, a special, brimmed diffusion cap has been developed.

During sampling, the diffusion tube is oriented vertically with the diffusion cap down (see figure 2). A field blank consisting of a capped sampling tube is mounted in parallel with the diffusion tube. Re-capping the diffusion tube ends the sampling. The analyses are performed using a gas chromatograph with a flame ionisation detector (FID).

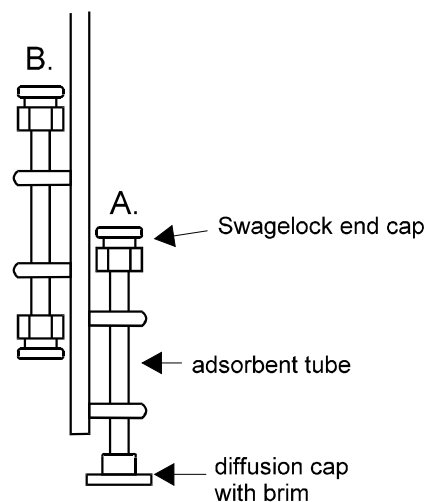


Figure 2 Diffusive sampler for VOC.

3.2 Distribution and classification of sites

The locations of sites were chosen in order to achieve results representative for the conditions in various sub-areas of Tallinn.

The measurement sites were classified into four types according to exposure to road vehicle emissions and street ventilation conditions, and the measurements were made at urban as well as at background sites. The classification of sites enables comparison of similar environments in a whole region, and of the variations in air pollution levels in different typical local environments.

The four classes of sites were:

- A = busy street environment, poor ventilation
- B = busy street environment, good ventilation
- C = residential area
- D = general pollution level, local background

A total of around 90 measurement sites have been used. The locations of the stations were selected by the Environmental Department in Tallinn in co-operation with IVL. In Figure 3 the station network in Tallinn is presented.

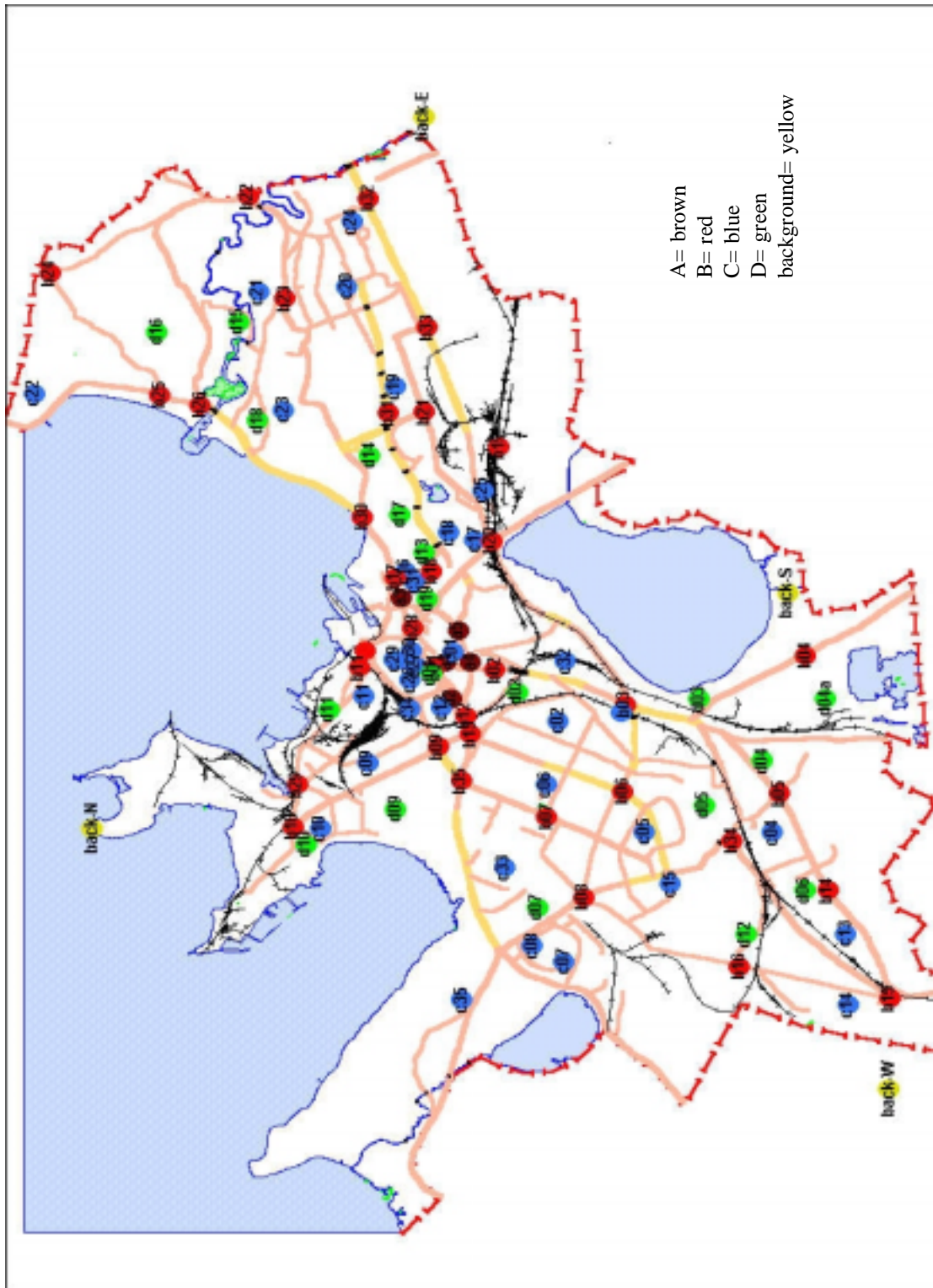


Figure 3 Map of Tallinn with the station network where the classification of sites is included.

3.3 Measurement programme

The measurements have been performed during 12 campaign periods. The campaigns have been run as “full” campaigns (3 times at all sites) or at a reduced level (including 20% of the stations), see table 1. The sampling periods were monthly for SO₂ and NO₂, while for VOC four weekly samples were used for monthly averages. During most of the campaigns sampling was also performed in background areas, in addition to those given in table 1. This was made in order to be able to evaluate the difference between conditions within the city to those in the very outskirts of the urban area. Sampling was made at two to four background locations in different geographical directions from Tallinn.

Table 1 Measurement campaigns

| Year, month | Number of sites (number of doubled samplers) | | |
|-------------|---|-----------------|-----|
| | SO ₂ | NO ₂ | VOC |
| 9604 | 78 (10) | 78 (10) | 9 |
| 9607 | 20 | 20 | |
| 9608 | 89 | 89 | 10 |
| 9610 | 27 (3) | 27 (3) | |
| 9612 | 26 | 26 | |
| 9701 | 86 (8) | 86 (8) | 10 |
| 9702 | 19 | 19 | |
| 9704 | 22 (1) | 22 (1) | |
| 9707 | 22 | 22 | |
| 9708 | 22 | 22 | |
| 9710 | 23 | 23 | 10 |
| 9801 | 18 | 18 | |

3.4 Meteorological conditions

A brief description of the meteorological conditions during the campaign periods is given in table 2, where average temperature, wind speed and dominant wind direction(s) are presented.

Table 2 *Meteorological conditions during the campaign months.*

| Month | Mean temperature, °C | Mean wind speed, m/s | Dominant wind direction |
|-------|----------------------|----------------------|-------------------------|
| 9604 | 3.9 | 2.6 | N,SW |
| 9607 | 15 | 3.0 | W,SW |
| 9608 | 18 | 2.2 | S |
| 9610 | 7.5 | 3.0 | SW,SSW |
| 9612 | -2.7 | 3.4 | SW,SSW |
| 9701 | -1.9 | 3.7 | SW |
| 9702 | -1.4 | 4.4 | SSW,SW |
| 9704 | 2.6 | 3.5 | SSW,SW,W |
| 9707 | 18 | 2.2 | NE,NNW |
| 9708 | 19 | 2.2 | NE,SW,NNW |
| 9710 | 4.6 | 2.6 | NW, SW |
| 9801 | -0.4 | 3.3 | S, SE |

4 Results

The measurements were performed as campaigns, with three as “full” campaigns and nine as “reduced” campaigns (see chapter 3.3). The number of samples in each of the classes in the two different types of campaigns are presented in table 3, which is valid for both NO₂ and SO₂. VOC was measured at 9 or 10 sites during the three “full” campaigns and during one of the “reduced” campaigns.

Table 3 *Number of sampling sites in each of the classified environments during the campaigns.*

| Classes | Number of sampling sites, “full” campaigns | Number of sampling sites, “reduced” campaigns |
|---------|---|--|
| A | 5 | 1-2 |
| B | 30-34 | 8-10 |
| C | 24-29 | 3-9 |
| D | 15-18 | 3-6 |

Background measurements of NO₂ and SO₂ were made in the outskirts of Tallinn at two or four different geographical directions during most of the campaigns.

4.1 Levels of NO₂

4.1.1 Measurement results

The results from the different classified environments and from background measurements are presented as a total for all 12 campaigns in table 4, and for each campaign event in Appendix 2. It is evident from the differences in the results that the selection of sites, mainly regarding the traffic situation, has been successful. As was expected, there is a falling average concentration in the order A, B, C, D and background sites. The minimum monthly mean in the A sites, 27 µg/m³, is way higher than in the other local environments. The difference between the background measurements and the “local background”, represented by the D sites is obvious, showing a clear influence also on D-sites from urban traffic.

Table 4 *Results from all campaigns, monthly averages of NO₂, µg/m³.*

| Class | Max | Average | Min | Number of results |
|------------|-----|---------|-----|-------------------|
| A | 48 | 36 | 27 | 32 |
| B | 50 | 29 | 10 | 173 |
| C | 33 | 18 | 6 | 136 |
| D | 30 | 14 | 5 | 95 |
| Background | 16 | 8 | 1 | 26 |

In figure 4 the levels of NO₂ are presented as the average, maximum and minimum value for each month and for the four classified environments respectively. The darker bars represent the “full” measurement campaigns.

Residential areas and areas of "local background" (C and D sites) generally exhibited lower exposure levels of NO₂ than more traffic exposed areas (A and B). This is applicable for all measurement periods.

The concentration levels were quite uniform throughout the whole measurement period, with mean values between 30 to 40 µg/m³ at the most exposed (A) sites down to 10-15 µg/m³ at the class D sites. A seasonality is however visible at the C and D sites with generally somewhat lower levels measured during summer months than in wintertime. The lowest mean values were measured in July 1996, at all types of sites. For the more traffic exposed A and B sites the highest mean values of NO₂ were measured in April 1996 and in August of 1997 respectively. These results do not follow the expected variations in pollution levels, which are of higher levels during wintertime than in summer. At the less traffic exposed C and D sites, the highest average values observed were, as for the A sites, in April 1996 but also in January 1997. As discussed further in chapter 4.2.1, the highest SO₂ levels were also measured in April 1996. In July of 1996, when the lowest mean values were observed, the predominant wind direction was from the west, which was the only campaign with this dominant wind direction. The occasions with high average levels of NO₂ were dominated by northerly, south westerly or north-easterly winds. Besides the dominant wind direction, also the average temperature and wind speed influences the levels of pollutants in air. The average calculated results might also be influenced by the number of sampling sites, as the "reduced" campaigns only used approximately 20% of the total number of stations (see tables 1 and 3).

The geographical variations in concentration levels are illustrated in figure 5 for the three "full" measurement campaigns, for AB and CD sites respectively. All monitoring sites are marked as black triangles. The differences in concentration load between the exposure classes are clearly shown, as well as the difference between the periods studied. The central parts of the town, where the traffic load can be expected to be high, exhibited the highest monthly means of NO₂.

4.1.2 Comparison to EU limit values

The EU limit values are not directly applicable to the measurements performed, but the average values calculated from all campaigns can be compared to the EU annual limit values (see Appendix 1). For NO₂ the annual limit value for protection of human health is 40 µg/m³ and the upper and lower assessment thresholds are 32 and 26 µg/m³ respectively. The limit value was not exceeded in Tallinn according to the measurements, but the upper assessment threshold was exceeded at the A points (36 µg/m³ as an average), and the lower assessment threshold also at the B points (29 µg/m³) (table 4). The average NO₂-levels at C, D and background measurement sites were all below the lower assessment threshold for protection of human health.

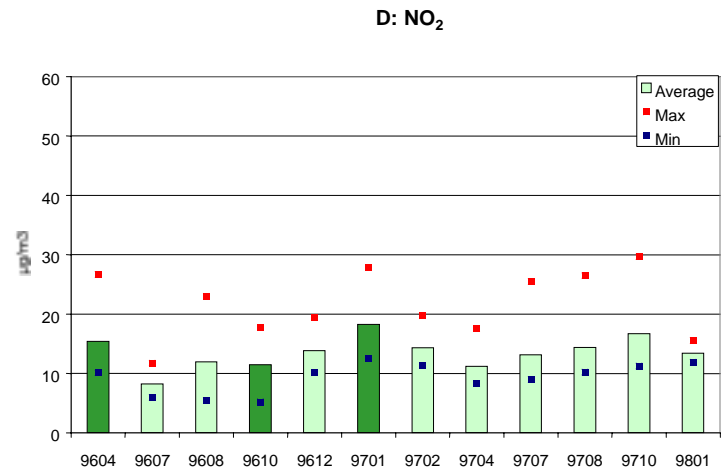
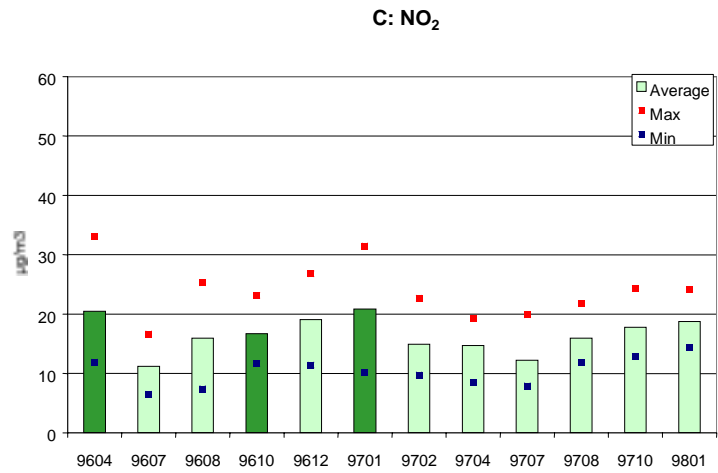
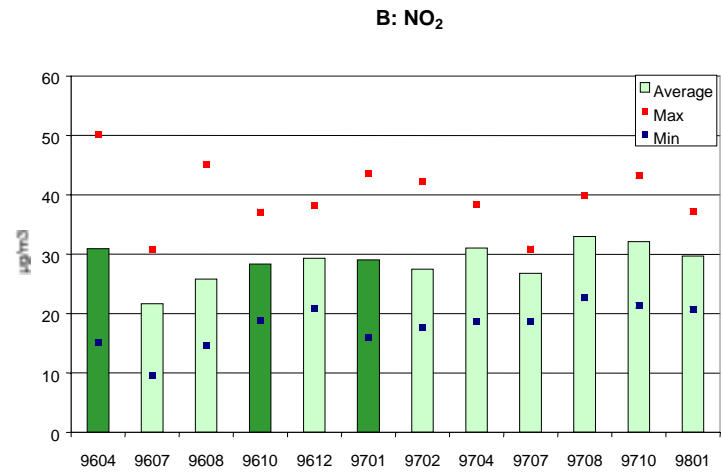
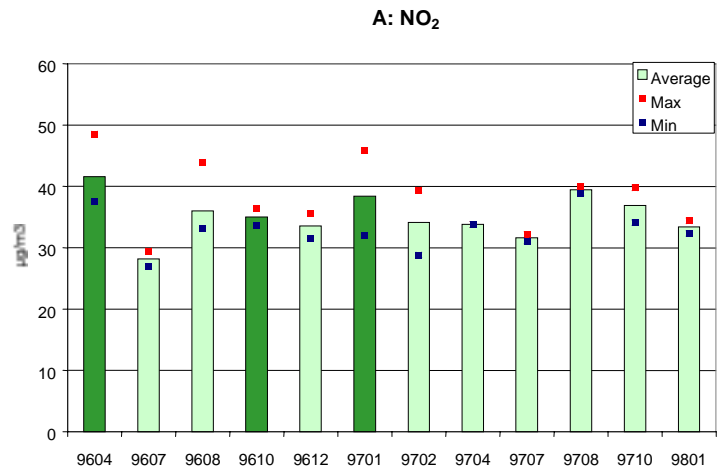


Figure 4 Average, maximum and minimum levels of NO₂ (µg/m³).

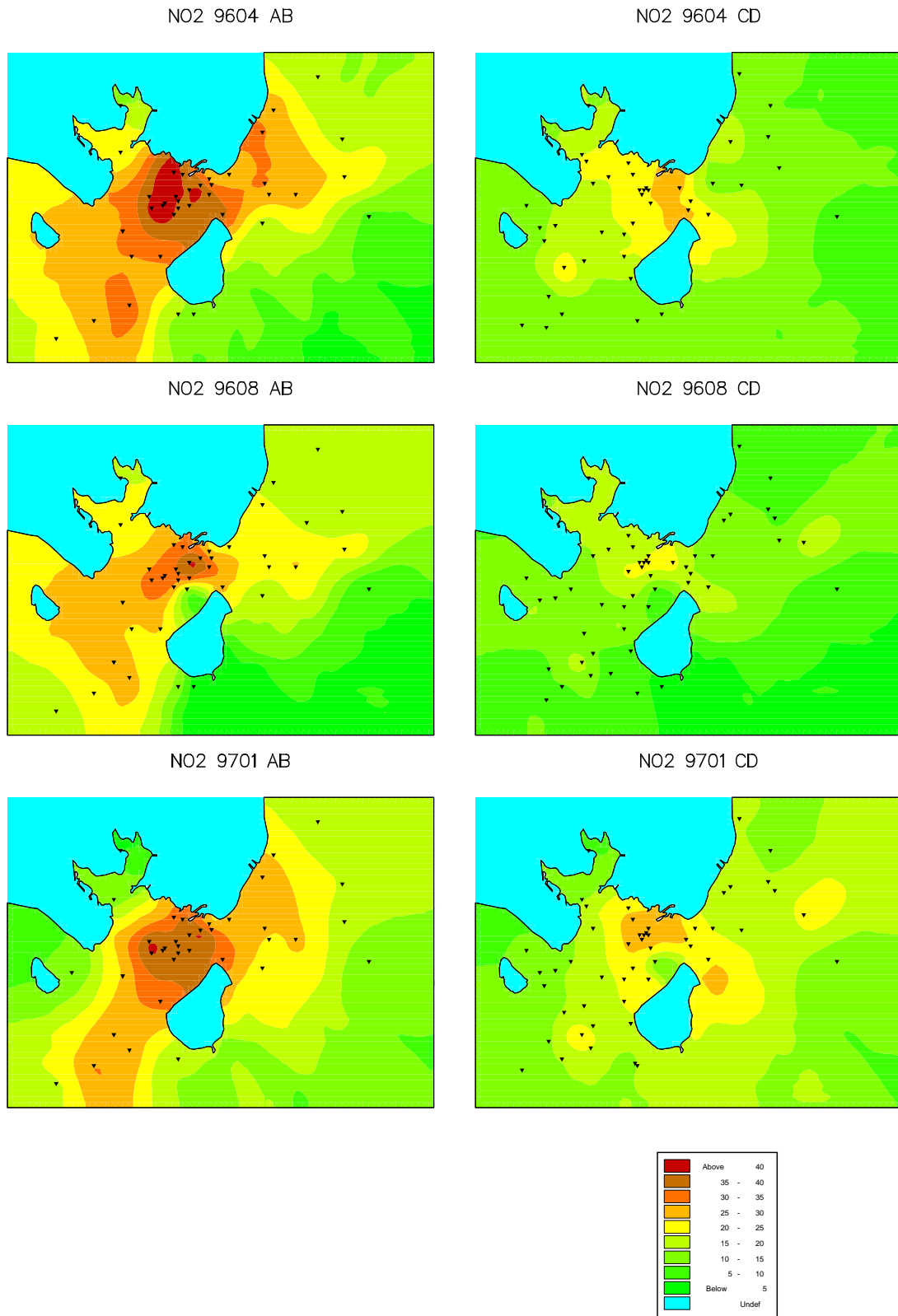


Figure 5 Levels of NO₂ ($\mu\text{g}/\text{m}^3$) in AB and CD site categories respectively.

4.2 Levels of SO₂

4.2.1 Measurement results

The results from the different classified environments and from background measurements are presented as a total for all 12 campaigns in table 5, and for each campaign event in Appendix 2. There is a falling average concentration in the order A, B, C, D and background sites, but the differences are not as pronounced as for NO₂. The maximum measured monthly means do not follow the classification of sites, but the highest minimum concentration was observed at an A site, while the minimum concentration in all other classes were comparable.

Table 5 Results from all campaigns, SO₂, µg/m³.

| Class | Max | Average | Min | Number of results |
|------------|----------|---------|-----|-------------------|
| A | 15 | 10 | 4 | 32 |
| B | 21 | 8 | 1 | 173 |
| C | 19 (39)* | 7 | 1 | 134 |
| D | 17 | 6 | 1 | 94 |
| Background | 12 | 4 | 1 | 26 |

* The result 39 µg/m³/month, very much higher than any of the other observations, was observed at one C site. It is uncertain if this is a valid result.

The average, maximum and minimum levels of SO₂ for each month and the four site classes are illustrated in figure 6. As for NO₂, the darker bars represent the “full” campaigns.

Also for SO₂ the highest average air concentrations were measured at the most traffic exposed sites, although the differences in average concentrations between A and D sites were considerably smaller than for NO₂. On the other hand greater relative variations in average concentration levels between different measurement periods were found.

Measured levels of SO₂ generally show a seasonal trend with higher levels in winter months and lower during summer. The lowest levels of SO₂ (2-3 µg/m³) were thus observed during July 1996. In July 1996, with predominantly westerly winds, the lowest average levels of NO₂ were also observed. The highest average SO₂ concentrations, above 10 µg/m³, were, however, measured in April 1996 at all classes of sites. In that month NO₂ levels were also amongst the highest. There is no obvious explanation to this. Generally higher levels of SO₂ would be expected during the colder part of the year. Examining the meteorological conditions, as presented in table 2, chapter 3.4, April of 1996 is the only month where the predominant wind direction was from the north. During this month the background result north of Tallinn was almost twice as high as any other background result, 12 µg/m³. The elevated levels of SO₂ and among the highest of NO₂ might be due to shipping in the Gulf of Finland or long-distance transport of air from other source areas north of Tallinn.

In figure 7 the geographical patterns of SO₂ levels at AB and CD sites respectively for the periods with the largest number of samplers (April and August 1996, January 1997) are shown. In the figures the variation in concentrations between these campaigns can be seen, and it is also evident that the differences between the site categories were relatively small. In some areas higher SO₂ concentrations were observed at CD sites than at sites in classes A and B.

There are large areas in Tallinn with private houses using wood, turf-briquettes and coal for heating, while other areas are dominated by apartment buildings using central heating. These areas were all covered as C-sites in the measurements. Selecting results from C-sites in areas with private houses separately from areas with apartment buildings, results from the “full” campaigns (April 1996, August 1996 and January 1997) show that the total average of SO₂ from private house areas, 7.7 µg/m³ were a little higher than in areas with apartment buildings, 6.7 µg/m³. In April and January only, when heating is on, the levels of SO₂ in the private house areas were clearly higher. In August, when heating is not necessary, the levels of SO₂ were a little lower in the private house areas than in the apartment building areas.

4.2.2 *Comparison to EU limit values*

The measurement results, as an average for all campaigns, are well below the EU annual limit value of 20 µg/m³ (Appendix 1). This limit value is set on the basis of protection of ecosystems, and should not be applied in the close vicinity of sources. The highest average value calculated on all campaign events, as well as only on winter months, was 10 µg/m³ at the A-sites, that is at 50% of the limit value. Since the limit value is not intended to be used close to sources, a comparison between the assessment threshold values and results from D-sites or background values could be made. In this comparison the average values from the measurements, 6 and 4 µg/m³ at D-sites and background sites respectively (table 5), are below the lower assessment threshold of 8 µg/m³.

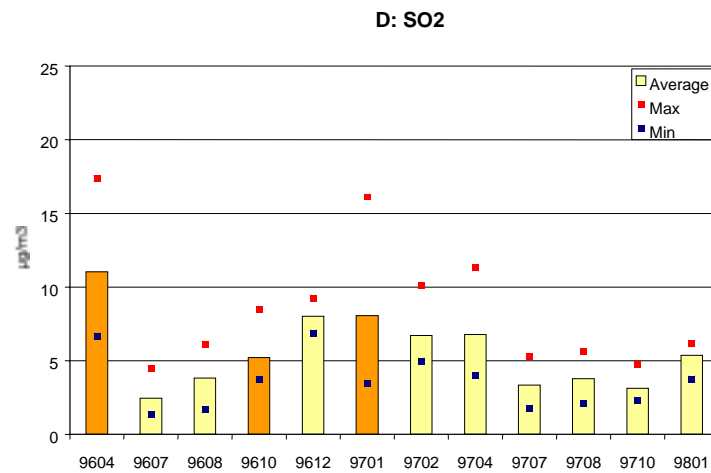
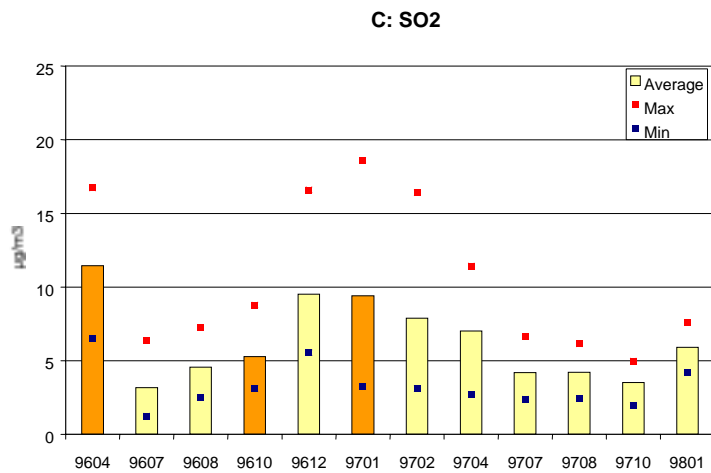
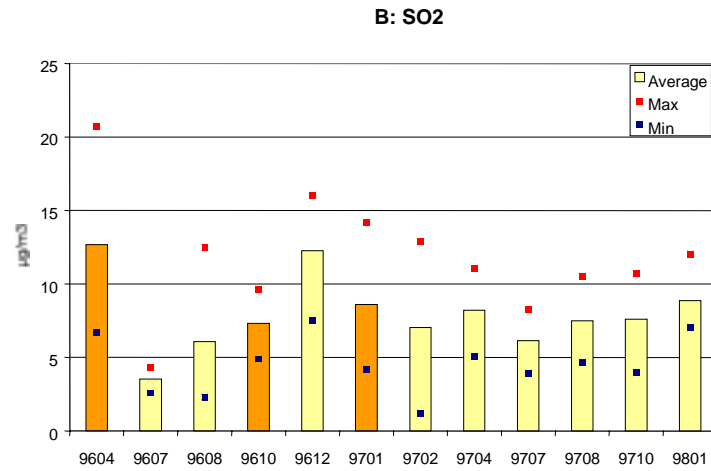
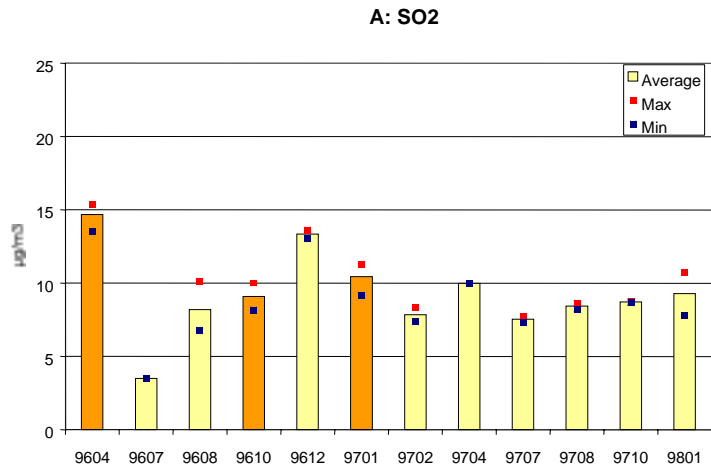


Figure 6 Average, maximum and minimum levels of SO_2 ($\mu g/m^3$).

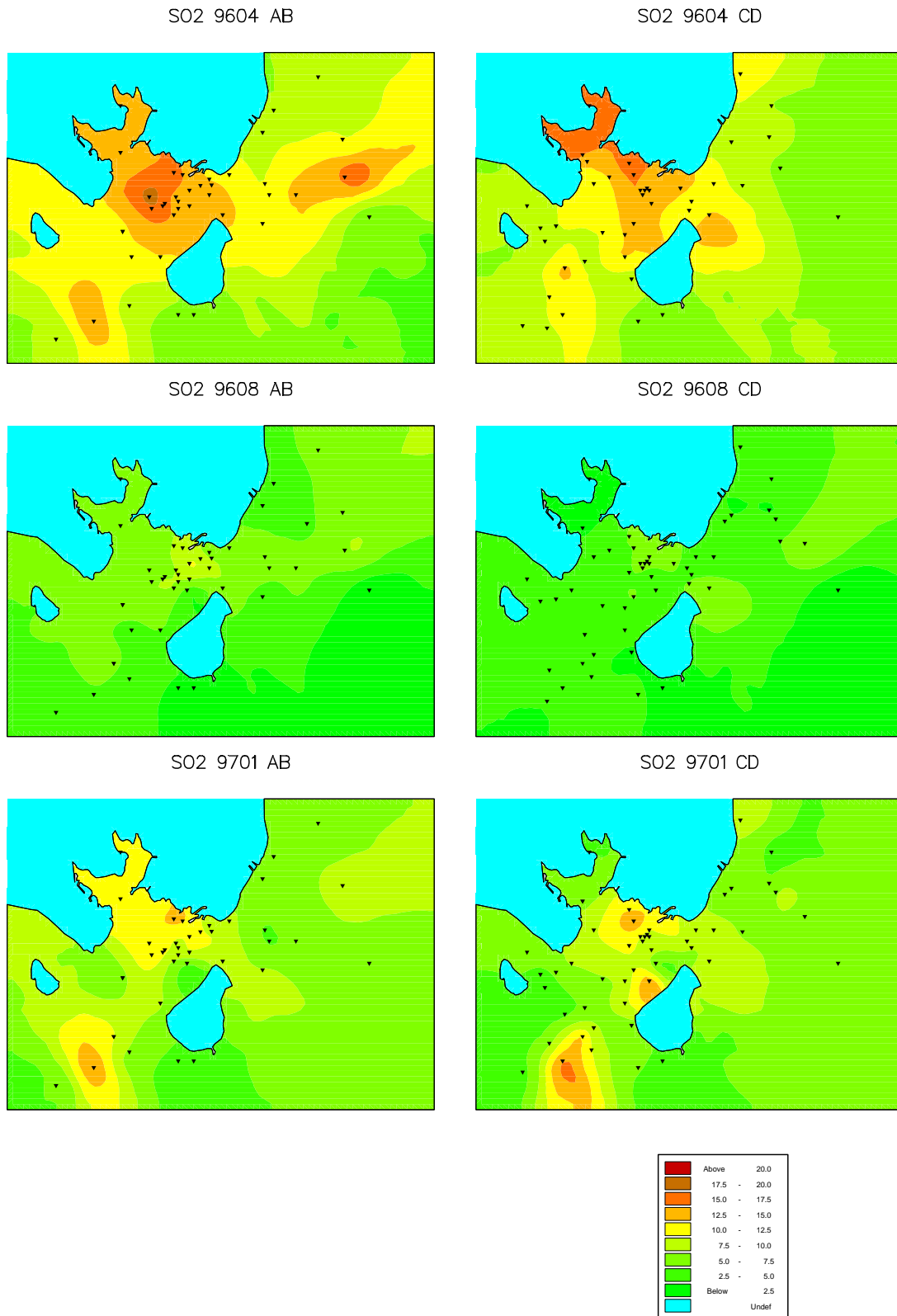


Figure 7 Levels of SO₂ (µg/m³) in AB and CD site categories respectively.

4.3 Levels of VOC

4.3.1 Measurement results

Eight different components of VOC were analysed; benzene, toluene, octane, butylacetate, ethylbenzene, m+p-xylene, o-xylene and nonane.

The sites V4 - V9 were located in areas classified as B-sites, while V1, V2 and V3 can be characterized as source sites. A brief description of the sites is found in table 6. V10 was a "moving" point, located at different places during the different campaigns .

Table 6 Description of sites

| Station number | Station name | Site type | Comments |
|----------------|------------------------------|------------------------|--|
| V1 | Trammi depoo | Source site | located around a furniture factory in the city centre |
| V2 | Tehnikakooli hoov | - " - | - " - |
| V3 | Magasini tänav | - " - | - " - |
| V4 | Endla ristmik | High traffic crossroad | |
| V5 | Kadaka ringtee | - " - | Gasoline station located at the north side of the crossroad |
| V6 | Järvevana/Pärnu mnt. Ristmik | - " - | Gasoline station located at the south side of the crossroad |
| V7 | Peterburi mnt. Ringtee | - " - | 3 gasoline stations around the crossroad |
| V8 | Lasnamäe kanalis | 8 lane highway | relatively low traffic intensity |
| V9 | Viru väljak | City centre | Station situated in a high traffic intensity area |
| V10 | Different sites | Different | 9604, roof level 9701, living area, close to oil heated power plant 9710, living area, near gasoline loading harbour |

The results from the VOC measurements are presented in figure 8, as site columns of monthly averages for the four months respectively. The third measurement period was performed during two weeks in January and two weeks in February 1997, here referred to as the January campaign.

The highest levels of VOC as a sum were observed in August 1996, and the lowest for most of the sites, in January 1997. The comparatively low levels in January/February were probably caused by a somewhat higher wind speed during that measurement period (see table 2). There is a general negative correlation between measured VOC-levels and wind speed, as reported from measurements in urban areas in Sweden (Svanberg et al., 1997 and 1998).

Generally there was a difference in pollution load regarding VOC, where site V4 consistently showed the highest levels, with V9 having the second highest levels. The source sites (V1 - V3), together with V8, an 8-lane highway with relatively low traffic intensity, and V10 at different locations, exhibited the lowest concentrations measured. Thus, the sites located at high traffic crossroads (V4-7), and at Virju väljak (V9) with high traffic intensity in the city centre, were the most polluted sites with regard to VOC.

In order to trace sources of the VOC measured the relation between individual VOC can be calculated. For the measurement sites in Tallinn the ratio of toluene:benzene and of xylenes:benzene have been calculated. In table 7 the ratios are presented, together with the average concentration of benzene and of total VOC at the different sites. The ratios of individual VOC depend both on the composition of the source, but also on the general pollution level of VOC, of the distance from the source etc. Comparisons among the sites in Tallinn do show that two of the source sites, V1 and V2, have a somewhat higher ratio for toluene:benzene than the other sites, and among the highest for xylenes:benzene. This might indicate influence from the furniture factory located close to these sites. The site V4, with the highest total load of VOC, is the only one with a higher xylene:benzene ratio. It has been observed in Swedish measurements that there is a correlation between higher total concentration and higher ratio of xylenes or toluene to benzene (Svanberg et al 1998).

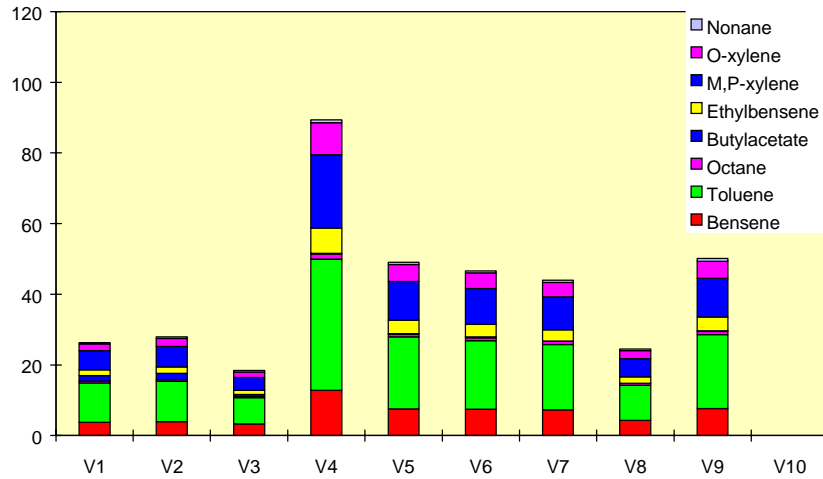
Table 7 *Ratio of toluene to benzene, and sum of xylenes to bensen, as well as average concentrations of benzene and total VOC at the different sites during four months measurements.*

| | Ratio | | Average concentrations, $\mu\text{g}/\text{m}^3$ | |
|----|-----------------|----------------|--|-----------|
| | toluene/benzene | xylene/benzene | benzene | Total VOC |
| V1 | 3.5 | 2.3 | 3.6 | 31 |
| V2 | 3.2 | 2.3 | 3.5 | 28 |
| V3 | 2.4 | 1.7 | 3.1 | 19 |
| V4 | 3.1 | 2.5 | 12.9 | 95 |
| V5 | 3.1 | 2.2 | 7.1 | 50 |
| V6 | 2.9 | 2.1 | 6.8 | 48 |
| V7 | 2.8 | 2.1 | 6.9 | 46 |
| V8 | 2.2 | 1.6 | 3.4 | 20 |
| V9 | 3.0 | 2.2 | 7.7 | 55 |

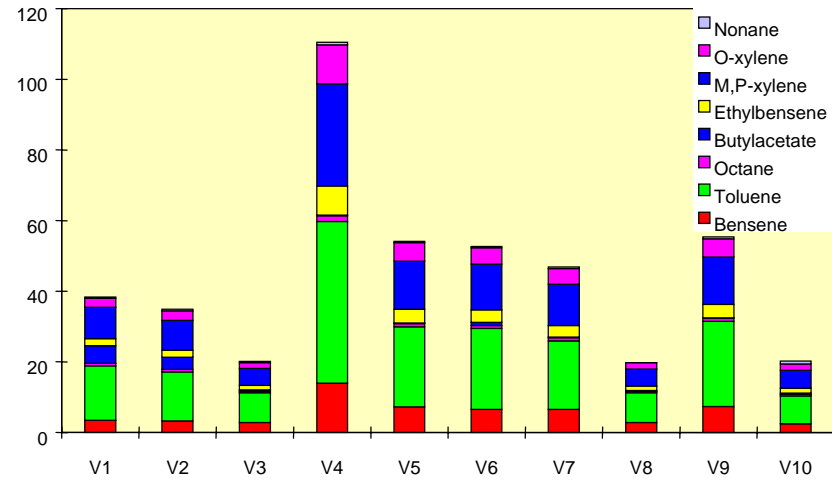
4.3.2 *Comparison to limit values and to VOC levels in other cities*

There are presently no EU limit values regarding VOC. Within the EU commission discussions are held regarding a limit value for benzene. The suggested limit value, probably an annual average for benzene of $5 \mu\text{g}/\text{m}^3$ to be met everywhere in 2010, will be the second daughter directive to the EU directive EU 96/62/EC. The average levels of benzene measured in Tallinn, based on four months measurements, exceed this level at some of the sites. This is however also the case in other European countries. In Germany for example annual averages for benzene in a number of cities (urban background, expected to be less polluted than the measurement sites in Tallinn) vary between ca $2\text{-}6 \mu\text{g}/\text{m}^3$. In Göteborg, Sweden, measurements in a street with heavy traffic and poor ventilation conditions (approximately an A-site, more exposed than the Tallinn sites), weekly averages of benzene varied in the interval $6\text{-}25 \mu\text{g}/\text{m}^3$ during the winter season 1997/98 (Svanberg et al 1998). In Tallinn the weekly results varied between approximately $2\text{-}23 \mu\text{g}/\text{m}^3$ of benzene.

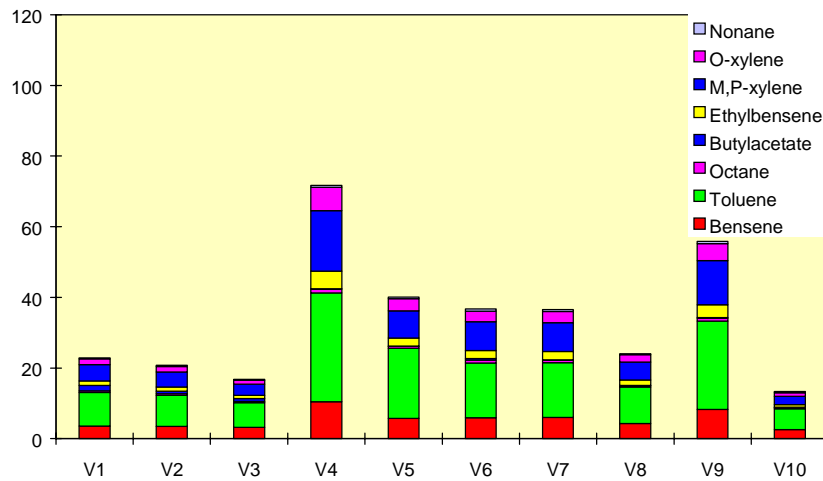
VOC ($\mu\text{g}/\text{m}^3$), April 1996



VOC ($\mu\text{g}/\text{m}^3$), August 1996



VOC ($\mu\text{g}/\text{m}^3$), January 1997



VOC ($\mu\text{g}/\text{m}^3$), October 1997

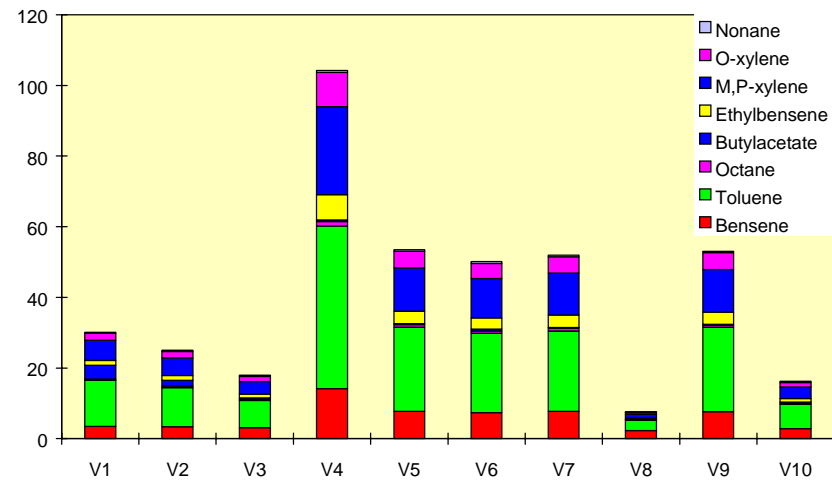


Figure 8 Average concentrations of VOC ($\mu\text{g}/\text{m}^3$).
Average of 3 weeks at site V8, 9604, Average of 2 weeks at site V8 and V10, 9608; V10, 9701

4.4 Comparison to Swedish conditions

Results from the Swedish Urban measurement network (Svanberg et al 1997, 1998) have been used for comparison of measurement results from Tallinn. The two largest cities in Sweden, Stockholm, ca 1 million inhabitants, and Gothenburg, approximately 500 000 inhabitants, were used. The measurements within the urban network are made at only one sampling point in the centre of the city, which is located not to be directly influenced by traffic, for example in a square without vehicles. The closest classification would be as a B-site. The measurements are not directly comparable, neither as to number of sampling sites, nor as to measurement techniques and sampling intervals. Furthermore, the measurements within the Swedish Urban network are performed only during the winter months, October-March. The results do however give an indication of the pollution levels in the three cities.

In table 8 the results from the last two winter periods in Stockholm and Gothenburg are presented and compared to the results from the winter period measurement campaigns in Tallinn.

The most obvious difference, as expected, is in the incidences of higher SO₂ concentrations in Tallinn compared to in the Swedish cities. For NO₂ the levels are of the same order of magnitude.

Table 8 Highest and lowest monthly average concentration ($\mu\text{g}/\text{m}^3$) in Stockholm and Gothenburg during the two winter seasons, 1996-97 and 1997-98 as compared to winter results from B sites in Tallinn.

| | Tallinn (B-sites) 34 sites | Stockholm 1 site | Gothenburg 1 site |
|---|-------------------------------|---------------------|----------------------|
| NO₂ ($\mu\text{g}/\text{m}^3$) | | | |
| Highest monthly mean | 44 | 32 | 40 |
| Lowest monthly mean | 16 | 24 | 23 |
| SO₂ ($\mu\text{g}/\text{m}^3$) | | | |
| Highest monthly mean | 16 | 3 | 4 |
| Lowest monthly mean | 1 | 1 | 1 |

5 Discussion and recommendations

The measurement campaigns using diffusive samplers performed in Tallinn have given a lot of interesting information.

The relations between the results from the different site categories seem to be reasonable. As expected, air concentrations of NO₂ were considerably higher at sites directly exposed to road traffic than at sites in near-by residential areas and in areas of "local background". For SO₂ the difference in average levels between the four classes were not as pronounced as for NO₂, but even for SO₂ residential areas and areas of "local background" generally exhibited lower exposure levels than areas more exposed to traffic. However, at some individual measurement sites there were deviations from the general pattern.

The different concentration patterns for the two components are probably mainly due to the emission situation. NO₂ is mostly generated by traffic, hence the highest concentrations will be found close to roads with high traffic intensity. The sources of SO₂ can be assumed to be more widespread within the area studied, since a major part of the SO₂ emissions originates from fossil fuel combustion in large power plants as well as in local residential heating devices.

The span in concentration between sites within one type category during one campaign period was less for A sites than for the others, probably at least partly due to the lower number of A sites.

Generally the SO₂ levels were higher during winter months, while for NO₂ the concentration levels were more even throughout the year. There was, however, a tendency of lower levels during summer months in the C and D points. The high concentrations of SO₂ and NO₂ observed in April 1996 might be due to meteorological conditions, where the prevailing wind direction was from the north, with addition of southwesterly winds. The background measurement site north of Tallinn showed the highest SO₂ background value measured during that month. The lowest SO₂ and NO₂ levels were measured in July 1996, when the wind direction was from the west and southwest. However, to be able to evaluate temporal variations in concentrations more thoroughly, longer time series and more meteorological data are needed.

In contrary to what was expected, the values of VOC were lower during the only wintertime campaign (January-February campaign 1997) than during the three other campaigns. One probable explanation to this is the higher average wind speed in January/February 1997 than during the other three VOC measurement periods. A general negative correlation between VOC-levels and wind speed has been observed elsewhere.

A brief comparison to concentration levels of NO₂ and SO₂ in the two largest Swedish cities, Stockholm and Gothenburg, show higher levels of SO₂ in Tallinn while the levels of NO₂ are in the same order of magnitude.

From the results achieved so far it is obvious that the use of diffusive samplers, in combination with a careful selection of measurement sites, is a fruitful strategy in mapping geographical variations in levels of air pollutants in Tallinn, an area of complex emissions. An important support in air quality management has been generated during a relatively short time period and at a reasonable cost. All results in combination have given an excellent insight into the variations of air pollution levels in the region as a whole as well as in different typical environments.

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Appendix 1, EU limit values

EU limit values for SO₂ and NO₂ not to be exceeded after 31dec. 2005.

| SO ₂ | Time period | Limit value | Lower assessment threshold | Upper assessment threshold |
|---|--|---|---|--|
| 1. Hourly value for protection of human health | 1 hour | 350 The value must not be exceeded more than 24 times/year | - | - |
| 2. 24-hour value for protection of human health | 24 hours | 125 The value must not be exceeded more than 3 times/year | 50 40% of 24 hour limit value | 75 60% of 24 hour limit value |
| 3. Annual value for protection of ecosystems. Not to be applied close to sources. | 1 year and during winter season (1 oct-31 march) | 20 | 8 40% of limit value for winter season | 12 60% of limit value for winter season |
| NO ₂ | Time period | Limit value | Lower assessment threshold | Upper assessment threshold |
| 1. Hourly value for protection of human health | 1 hour | 200 The value must not be exceeded more than 18 times/year | 100 50% of hourly limit value | 140 70% of hourly limit value |
| 2. Annual value for protection of human health | 1 year | 40 | 26 65% of limit value | 32 80% of limit value |
| 3. Annual value for protection of vegetation. Not to be applied close to sources. | 1 year | 30 As the sum of nitrogen oxides, NO ₂ +NO | 19.5 65% of limit value | 24 80% of limit value |

Appendix 2, Measurement results

NO₂ (µg/m³). Average, max, min and number of results for each campaign period.

| A | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
|----------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| Average | 41.6 | 28.2 | 36.0 | 35.0 | 33.6 | 38.4 | 34.1 | 33.8 | 31.6 | 39.5 | 36.9 | 33.4 |
| Max | 48.5 | 29.5 | 43.9 | 36.4 | 35.6 | 45.8 | 39.4 | 33.8 | 32.1 | 40.0 | 39.8 | 34.4 |
| Min | 37.5 | 26.9 | 33.1 | 33.6 | 31.5 | 32.0 | 28.8 | 33.8 | 31.1 | 38.9 | 34.1 | 32.3 |
| No. of results | 5 | 2 | 5 | 2 | 2 | 5 | 2 | 1 | 2 | 2 | 2 | 2 |
| B | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
| Average | 30.9 | 21.7 | 25.8 | 28.4 | 29.3 | 29.0 | 27.5 | 31.0 | 26.8 | 33.0 | 32.1 | 29.7 |
| Max | 50.1 | 30.8 | 45.1 | 37.1 | 38.2 | 43.6 | 42.2 | 38.4 | 30.9 | 39.9 | 43.2 | 37.2 |
| Min | 15.1 | 9.6 | 14.7 | 18.8 | 20.9 | 15.9 | 17.6 | 18.7 | 18.7 | 22.7 | 21.4 | 20.8 |
| No. of results | 31 | 8 | 34 | 9 | 8 | 30 | 9 | 10 | 8 | 8 | 9 | 9 |
| C | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
| Average | 20.5 | 11.2 | 16.0 | 16.7 | 19.0 | 20.9 | 14.9 | 14.7 | 12.2 | 16.0 | 17.8 | 18.7 |
| Max | 33.1 | 16.6 | 25.2 | 23.2 | 26.8 | 31.5 | 22.7 | 19.3 | 19.9 | 21.7 | 24.3 | 24.2 |
| Min | 11.9 | 6.5 | 7.4 | 11.6 | 11.4 | 10.2 | 9.7 | 8.5 | 7.9 | 11.8 | 12.9 | 14.4 |
| No. of results | 24 | 6 | 28 | 9 | 9 | 29 | 4 | 6 | 6 | 6 | 5 | 4 |
| D | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
| Average | 15.4 | 8.2 | 12.0 | 11.5 | 13.8 | 18.3 | 14.3 | 11.2 | 13.1 | 14.4 | 16.7 | 13.4 |
| Max | 26.7 | 11.8 | 23.0 | 17.8 | 19.4 | 27.9 | 19.7 | 17.5 | 25.5 | 26.5 | 29.7 | 15.5 |
| Min | 10.1 | 6.0 | 5.5 | 5.2 | 10.1 | 12.6 | 11.3 | 8.3 | 9.0 | 10.3 | 11.3 | 11.9 |
| No. of results | 15 | 4 | 18 | 5 | 5 | 18 | 4 | 5 | 6 | 6 | 6 | 3 |

SO₂ (µg/m³). Average, max, min and number of results for each campaign period.

| A | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
|----------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| Average | 14.7 | 3.5 | 8.2 | 9.1 | 13.3 | 10.4 | 7.9 | 10.0 | 7.5 | 8.4 | 8.7 | 9.3 |
| Max | 15.4 | 3.5 | 10.2 | 10.0 | 13.6 | 11.3 | 8.3 | 10.0 | 7.7 | 8.6 | 8.8 | 10.8 |
| Min | 13.6 | 3.5 | 6.8 | 8.2 | 13.1 | 9.2 | 7.4 | 10.0 | 7.4 | 8.2 | 8.7 | 7.8 |
| No. of results | 5 | 2 | 5 | 2 | 2 | 5 | 2 | 1 | 2 | 2 | 2 | 2 |
| B | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
| Average | 12.7 | 3.5 | 6.1 | 7.3 | 12.3 | 8.6 | 7.0 | 8.2 | 6.1 | 7.5 | 7.6 | 8.9 |
| Max | 20.7 | 4.3 | 12.5 | 9.6 | 16.0 | 14.2 | 12.9 | 11.0 | 8.3 | 10.5 | 10.7 | 12.0 |
| Min | 6.7 | 2.6 | 2.3 | 4.9 | 7.5 | 4.2 | 1.2 | 5.1 | 3.9 | 4.6 | 4.0 | 7.1 |
| No. of results | 31 | 8 | 34 | 9 | 8 | 30 | 9 | 10 | 8 | 8 | 9 | 9 |
| C | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
| Average | 11.5 | 3.2 | 4.6 | 5.3 | 9.5 | 9.4 | 7.9 | 7.0 | 4.2 | 4.2 | 3.5 | 5.9 |
| Max | 16.7 | 6.4 | 7.3 | 8.8 | 16.6 | 18.6 | 16.4 | 11.4 | 6.7 | 6.2 | 5.0 | 7.6 |
| Min | 6.5 | 1.2 | 2.5 | 3.1 | 5.5 | 3.3 | 3.1 | 2.7 | 2.4 | 2.4 | 2.0 | 4.2 |
| No. of results | 24 | 6 | 27 | 9 | 9 | 29 | 4 | 6 | 6 | 6 | 5 | 3 |
| D | 9604 | 9607 | 9608 | 9610 | 9612 | 9701 | 9702 | 9704 | 9707 | 9708 | 9710 | 9801 |
| Average | 11.0 | 2.5 | 3.8 | 5.2 | 8.0 | 8.1 | 6.7 | 6.8 | 3.3 | 3.8 | 3.1 | 5.4 |
| Max | 17.4 | 4.5 | 6.1 | 8.5 | 9.3 | 16.1 | 10.1 | 11.3 | 5.3 | 5.7 | 4.7 | 6.2 |
| Min | 6.7 | 1.4 | 1.7 | 3.7 | 6.9 | 3.5 | 5.0 | 4.0 | 1.8 | 2.1 | 2.3 | 3.8 |
| No. of results | 15 | 4 | 18 | 4 | 5 | 18 | 4 | 5 | 6 | 6 | 6 | 3 |

Monthly averages of VOC ($\mu\text{g}/\text{m}^3$).

| April 1996 | | | | | | | | |
|---------------------|----------------|----------------|---------------|---------------------|---------------------|-------------------|-----------------|---------------|
| Site | Benzene | Toluene | Octane | Butylacetate | Ethylbenzene | m,p-xylene | o-xylene | Nonane |
| V1 | 3.8 | 11.2 | 0.6 | 1.5 | 1.6 | 5.4 | 1.9 | 0.4 |
| V2 | 4.0 | 11.4 | 0.5 | 1.8 | 1.8 | 5.8 | 2.2 | 0.5 |
| V3 | 3.3 | 7.4 | 0.4 | 0.5 | 1.2 | 3.6 | 1.5 | 0.4 |
| V4 | 12.9 | 37.1 | 1.4 | 0.2 | 7.1 | 20.8 | 9.0 | 0.8 |
| V5 | 7.6 | 20.3 | 0.8 | 0.2 | 3.8 | 11.0 | 4.8 | 0.6 |
| V6 | 7.4 | 19.4 | 0.9 | 0.2 | 3.5 | 10.2 | 4.4 | 0.6 |
| V7 | 7.3 | 18.5 | 0.8 | 0.1 | 3.2 | 9.3 | 4.1 | 0.6 |
| V8 | 4.3 | 10.0 | 0.5 | 0.1 | 1.8 | 5.1 | 2.2 | 0.4 |
| V9 | 7.7 | 20.9 | 0.9 | 0.2 | 3.8 | 11.0 | 4.8 | 0.8 |
| V10 | | | | | | | | |
| August 1996 | | | | | | | | |
| Site | Benzene | Toluene | Octane | Butylacetate | Ethylbenzene | m,p-xylene | o-xylene | Nonane |
| V1 | 3.5 | 15.4 | 0.8 | 4.9 | 2.0 | 9.0 | 2.5 | 0.4 |
| V2 | 3.3 | 14.0 | 0.7 | 3.4 | 2.0 | 8.5 | 2.7 | 0.4 |
| V3 | 2.8 | 8.4 | 0.4 | 0.4 | 1.3 | 4.7 | 1.8 | 0.3 |
| V4 | 14.0 | 45.7 | 1.7 | 0.2 | 8.2 | 29.0 | 11.1 | 0.7 |
| V5 | 7.3 | 22.6 | 1.0 | 0.2 | 3.9 | 13.6 | 5.2 | 0.4 |
| V6 | 6.6 | 22.9 | 0.8 | 1.0 | 3.5 | 12.9 | 4.7 | 0.3 |
| V7 | 6.6 | 19.4 | 0.9 | 0.2 | 3.3 | 11.7 | 4.4 | 0.4 |
| V8 | 2.8 | 8.5 | 0.4 | 0.1 | 1.4 | 4.7 | 1.8 | 0.1 |
| V9 | 7.4 | 24.2 | 0.8 | 0.2 | 3.8 | 13.4 | 5.2 | 0.6 |
| V10 | 2.5 | 7.9 | 0.4 | 0.4 | 1.4 | 5.0 | 1.9 | 0.8 |
| January 1997 | | | | | | | | |
| Site | Benzene | Toluene | Octane | Butylacetate | Ethylbenzene | m,p-xylene | o-xylene | Nonane |
| V1 | 3.6 | 9.6 | 0.4 | 1.5 | 1.3 | 4.7 | 1.6 | 0.3 |
| V2 | 3.5 | 8.7 | 0.4 | 0.7 | 1.2 | 4.3 | 1.5 | 0.4 |
| V3 | 3.2 | 7.0 | 0.4 | 0.8 | 0.9 | 3.1 | 1.1 | 0.3 |
| V4 | 10.5 | 30.8 | 1.0 | 0.2 | 4.9 | 17.1 | 6.6 | 0.6 |
| V5 | 5.7 | 19.8 | 0.6 | 0.1 | 2.3 | 7.7 | 3.5 | 0.4 |
| V6 | 5.9 | 15.5 | 0.8 | 0.4 | 2.3 | 8.1 | 3.0 | 0.6 |
| V7 | 6.0 | 15.5 | 0.7 | 0.1 | 2.4 | 8.1 | 3.2 | 0.6 |
| V8 | 4.3 | 10.3 | 0.4 | 0.1 | 1.5 | 5.1 | 2.0 | 0.3 |
| V9 | 8.3 | 24.9 | 0.8 | 0.2 | 3.6 | 12.6 | 4.8 | 0.6 |
| V10 | 2.6 | 5.9 | 0.4 | 0.1 | 0.8 | 2.4 | 1.0 | 0.3 |

October 1997

| Site | Benzene | Toluene | Octane | Butylacetate | Ethylbenzene | m,p-xylene | o-xylene | Nonane |
|-------------|----------------|----------------|---------------|---------------------|---------------------|-------------------|-----------------|---------------|
| V1 | 3.5 | 13.0 | 0.4 | 3.8 | 1.4 | 5.7 | 2.0 | 0.3 |
| V2 | 3.4 | 11.0 | 0.4 | 1.7 | 1.3 | 5.0 | 1.8 | 0.4 |
| V3 | 3.2 | 7.6 | 0.3 | 0.5 | 1.0 | 3.6 | 1.4 | 0.4 |
| V4 | 14.2 | 46.0 | 1.2 | 0.4 | 7.2 | 24.9 | 9.7 | 0.6 |
| V5 | 7.7 | 23.8 | 0.8 | 0.3 | 3.5 | 12.2 | 4.7 | 0.4 |
| V6 | 7.4 | 22.5 | 0.6 | 0.5 | 3.2 | 11.1 | 4.3 | 0.5 |
| V7 | 7.7 | 22.8 | 0.8 | 0.2 | 3.4 | 11.9 | 4.6 | 0.5 |
| V8 | 2.3 | 2.9 | 0.2 | 0.1 | 0.3 | 1.1 | 0.4 | 0.3 |
| V9 | 7.6 | 23.9 | 0.6 | 0.2 | 3.4 | 12.1 | 4.8 | 0.4 |
| V10 | 2.9 | 6.8 | 0.4 | 0.3 | 1.0 | 3.2 | 1.3 | 0.4 |
