ATTACHMENT D

to

City's Petition to EPA Challenging Emission Factors

July 9, 2008

Fugitive VOC-emissions measured at Oil Refineries

in the Province of Västra Götaland in South West Sweden

- a success story

development and results 1986 – 2001

commissioned by The County Administration of Västra Götaland

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PRODUCTION | THE COUNTY ADMINISTRATION OF VÄSTRA GÖTALAND **TEXT** | LENNART FRISCH, AGENDA ENVIRO AB
LAYOUT | CILLA ODENMAN **PUBLICATION** | 2003:56 **ISSN** | 1403-168X
PRINT | GÖTEBORGS LÄNSTRYCKERI AB

PREFACE

This report describes the environmental trends that have been on the agenda of the Swedish oil refineries in recent years, specifically focusing on emissions of Volatile Organic Compounds (VOC). In the case of oil refineries this is more or less also synonymous with hydrocarbons and in most cases VOC is synonymous with NMVOC (Non-methane VOC). If methane is included this is clearly stated in the report.

The issue of VOC-emissions has been high on the agenda for the Swedish oil refineries since the mid 1980's, when the first major discussions started on how to carry out measurements at the sites. Later the issue also has been raised for, among others, oil harbours and other main tank storage areas. The total crude oil throughput of the Swedish oil refining sites is about 20 million ton per year.

Today we have more than 15 years of measurement experience with the laser based DIAL-system (Differential Absorption Lidar). The system has been shown to be a very powerful tool in the measurement, as well in the combat, of the true VOC-emission. Other systems have also been tested (DOAS, HAWK) but have been shown to be non-reliable in performance.

This report is written by Lennart Frisch, MD at the environmental consulting bureau Agenda Enviro AB, and is commissioned by the County Administration of Västra Götaland (former the Provincial Government of Göteborg and Bohus) and the Swedish Environmental Protection Agency.

The Author is fully responsible for the content in the report.

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Between 1981 – 1986 he was process engineer and head of computer systems at the Shell Refinery in Göteborg, later environmental officer at the regional authorities of the Province of Göteborg and Bohus, and since 1996 an environmental consultant for mainly industrial clients but also for the Swedish environmental ministry, the Swedish EPA as well as regional and local environmental authorities. He has amongst others been Swedish representative at the EU-commission network IMPEL (Implementation and enforcement of environmental law) and the Article 19 committee of EMAS at the EU-commission. He has been a multi-annual member of the Swedish EPA advisory board on implementation and enforcement of environmental law and of the Swedish EPA scientific committee on air quality and emissions to air.

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1. SHORT HISTORICAL BACKGROUND

In Sweden there are three fuel producing oil refineries. On top of that there are also oil refining facilities for other products like bitumen and lube oil.

Out of the total of five oil refineries in Sweden four lie in the Province of Västra Götaland and out of these, three are situated in the town of Göteborg (Gothenburg), the Capital of the Province and the second biggest town of Sweden. The fourth refinery in the province - with the highest capacity - is situated in the municipality of Lysekil some 100 km north of Göteborg. The fifth oil refinery is mainly producing lube oil and is situated in Nynäshamn, some 100 km south of Stockholm. Crude oil and the products received when processing it are also handled at a number of Oil Harbours along the Swedish coast. The largest facilities for this are the Gothenburg Port and the oil harbour at the Scanraff oil refinery in Lysekil.

The first refinery in the area, the Koppartrans refinery, later bought by Shell, was on stream in 1953. Originally this plant was planned and designed for China, but with the changing political realities at that time, the facilities were redirected to Sweden and Göteborg. Prior to that the Nynäs oil refinery in Nynäshamn had already opened in 1928, at that time also being a fuel producing refinery. The second Nynäs-refinery, was opened in Göteborg in 1956 aiming at a production of mainly bitumen.

In the mid 1960's the Shell refinery was revamped doubling its capacity and in 1967 BP got its own refinery on stream (later sold to OK Petroleum and later renamed Preem). Until the beginning of the 1970's there were no refineries in the province having other than low skimming facilities. In 1972 Shell installed a thermal cracker unit and 1975 the Scanraff facilities in Lysekil came on stream with about the same production outline as the Shell refinery, but with significant higher capacity (7 Mton/a). In 1984 the Scanraff refinery was extended with a catalytic cracker unit. Scanraff is also today within the Preem Group.

At the beginning of the 1970's there were plans for major extensions of the Shell and BP refineries (up to 13 and 15 Mton/a respectively). These plans were however subsequently turned down because of the energy crisis in 1973/74 as well as the startup of Scanraff. There were also plans for a second refinery, "Statsraff" close to Scanraff. These plans were also never fulfilled.

Environmental issues were not really on the refinery agenda in the beginning, although equipment for the removal and recovery of sulphur in process streams - such as Claus-units - were installed all over during the 1960's and 1970's. The function of these units in the BP and Shell case though left some doubt, leaving BP to slaughter the old Claus-units, installing a new (smaller) one in the early 1980's and Shell revamping its units also in the 1980's.

The turning time in environmental thinking at the refineries came during the second half of the 1980's with some court cases on sulphur emissions. This lead to a subsequent change of policy at the oil refineries towards an environmental image. After substantially reducing overall emissions in the late 1980's advanced facilities for sulphur removal - tail gas treating units - were installed at both the Shell and Scanraff refineries in 1993/4 and soon after that also at Preem. Scanraff also reduced the use of oil as internal fuel early on so that the entire refinery - with the exception of the FCC-unit using coke – was normally fired on gas only.

In the beginning of the 1990's low NOx-burners were introduced at the refineries, starting with some mixed experience. Clearly though that introduction, as well as the increased knowledge in the control rooms of the impact of firing conditions to the creation of NOx-emissions, also reduced NOx -emissions substantially although it has been difficult to describe exactly how much as historically NOx never was measured. Later also SCR-units, beginning with the FCC at Scanraff, were installed, today also being used for boilers at the Shell and Preem refineries.

Emissions of volatile organic compounds (VOC) historically were only roughly calculated either as a figure based on throughput, or on the number of certain process-units in the plant multiplied by certain theoretical emission data. Historically emissions from storage facilities, such as tanks were only very rarely thought of being of any magnitude to count with. Because of hard pressure from the Provincial Government in the second half of the 1980's sophisticated measurement devices were taken out of the laboratories to be used for field measurements.

Measured – true – VOC-emissions showed to be substantially higher than what could be thought of based on the old calculations, especially for the storage facilities. Based on the first measurements with the laser technique in 1988 and 1989, later measurements in 1992, 1995, 1996 and 1999 have shown tremendous reductions of VOC-emissions during these years. The reduced emissions clearly follow fruit-bearing actions taken by the companies to reduce emissions.

Starting in 1996, VOC-measurements with the DIAL-technique were also carried out in the oil harbour of the Göteborg Port, giving the same principal results as at the oil refineries. In 1999 also the first DIAL-measurement was carried out at the Nynäshamn oil refining site of Nynäs, also here giving the same principal results as the early measurements at the other oil refineries some 5-15 years earlier.

The only emissions not in accordance with the diminishing trend are the emissions of carbon dioxide. As a basic rule, further refining of the crude oil needs more energy than if no such refinement took place. This was already the case for the deeper conversion introduced by the Shell refinery in the early 1970's and with the introduction of the FCC at Scanraff in 1984 and of course with an increased production in itself. In recent years CO2-emissions have increased further based on the demand from society on the oil refineries to produce new, less environmentally disturbing products. This of course is a contradictory situation, which politically has been shown not to be all too easy to handle.

2. SHORT-CUT INFORMATION ON THE SITES FOR OFFICIAL DIAL-MEASUREMENTS

Below the main features are described for the different refineries as well as for the Oil harbour at the Göteborg Port.

2.1 Preem Raffinaderi AB, Göteborg

Being a BP Refinery until 1991, since its start on stream in 1966, the refinery has had a low-skimming profile until the mid 1990's. In 1994 an isomerization unit was set on stream as the first new major process change since the startup. In 1996 facilities for the desulphurization of gasoil as well as for the production of "Environmental diesel" were installed. At the same time new big tail-gas treating units for process-sulphur came on stream.

The licensed throughput is 6 Mton/a although a practical limit could be assumed at somewhat more than 5 Mton/a. The normal annual throughput has been around 4 Mton/a, with the exception of some years in the beginning of the 1980's, when then throughput dropped below 3 Mton/a as a result of a major fire.

The number of people employed is about 250. The refinery is situated in the municipality of Göteborg on the Hising Island.

Measurements with DIAL (Spectrasyne) have been executed in 1988, 1989, 1992, 1995/96 and 1999.

2.2 Skandinaviska Raffinaderi AB, Scanraff, Lysekil

This refinery was planned in the 1960's and got its licensing in the early 1970's. In the early years the licensed throughput was 7 Mton/a. After a period having the limit on 8.3 Mton/a it is now set at 10 Mton/a.

In 1984 the refinery was extended with a FCC unit, originally with a licensed capacity of 1.3 Mton/a. In 1992 this was raised to 1.5 Mton/a, with 1.75 Mton/a from 1995 and onwards.

The owners have differed throughout the years, now being owned by Preem, the same owner as for the Preem refinery.

The refinery is situated in the municipality of Lysekil, without any other industry of its size in the neighborhood and being the industrial facility of highest importance in the area. The refinery employs some 550 people.

Measurements with DIAL (Spectrasyne) have been executed in 1992, 1995 and 1999.

2.3 Shell Raffinaderi AB, Göteborg

The equipment for the refinery was originally built in the USA with destination for mainland China just after the 2nd World War. Due to the political changes in China at that time an alternative destination was thought of. Starting under the name of Koppartrans with a shared ownership by two Swedish companies, Kopparberg and Transatlantic a small fuel producing refinery with two minor crude oil units was set up in the 1950's. The maximum capacity at this time was about 2 Mton/a.

After being bought by Shell, new facilities were installed in the mid 1960's more than doubling the throughput. Licensed throughput is 5 Mton/a, but the practical maximum could be set at around 4 Mton/a. The refinery is situated in the municipality of Göteborg on the Hising Island.

The number of people employed is a little less than 200.

Measurements with DIAL (Shell Research) have been executed in 1996 and 1999.

2.4 Nynäs AB, Göteborg

Nynäs AB is a refinery in Göteborg (Gothenburg) situated at the Hising Island producing mainly bitumen and related products. The licensed throughput is 450 000 ton/a. The facilities were built in 1956 and subsequently put on stream in 1957 slowly increasing the throughput from some 100 - 200 kton/a in the early years to around 400 kton in recent years. On a monthly basis the throughput is about 50 kton, but as the plant normally has a winter shut down the possible level of some 600 000 ton/a is never reached at the present situation. As the winter shut down is based on the needs of the domestic market, changes could though be brought about in the future if the market picture is being altered.

The Nynäs refinery is normally referred to as the "small bitumen plant" in the Province as the facilities for the fuel producing refineries are much bigger.

The number of people employed is about 50 at the refinery.

Measurements with DIAL (Spectrasyne) have been executed in 1995 and 1999.

2.5 Nynäs AB, Nynäshamn

The refinery is situated in Nynäshamn some 100 km south of Stockholm and has the longest history of the Swedish refineries. The refinery was started in 1928 and was a fuel producing refinery until 1983. At that time Nynäs, as a company, left the Swedish fuel market and the refinery was revamped in order to produce bitumen products and naphtenic special oils including lube oils using a very heavy crude oil. The license for the refinery is limited to 1,8 Mton of crude oil intake, and the refinery is equipped with, amongst others, one vacuum distillation and three hydrations units, the latter being one hydrofinisher and two hydrotreaters.

The desulphurization capacity has been increased during the last years and new equipment has been installed for the removal of sulphur. The refinery uses external fuel.

Measurements with DIAL (Spectrasyne) have been executed in 1999.

2.6 Gothenburg Port, Oil Harbour, Göteborg

The Gothenburg Port has a long history dating back to the time when Göteborg was founded in the 17th century. Since about 1850 the Gothenburg Port has held the position of being the largest Swedish – as well as Nordic – port, now being about number 10 in Europe. Annually some 34 Mton of goods is handled in the port of which the Oil harbour is handling close to 20 Mton of crude oil and oil products.

The Oil harbour is situated on the northern shore of the Göta Älv river and thus nowadays lies more or less as a part of the Göteborg city, although a bit west of the centre. Within the oil harbour site also a number of handling and distribution companies have their facilities including tank storage and off-loading of products to trucks and railway.

Measurements with DIAL (Spectrasyne and Shell Research) have been executed in 1996 and 1999 respectively.

3. INITIAL MEASUREMENTS

In the early and mid 1980's the problem with the – at that time – unknown real emissions of Volatile Organic Compounds (VOC) from the oil refineries, lead to a number of discussions between representatives from the environmental enforcing authority, the Provincial Government of Göteborg and Bohus, and representatives from the refineries.

The discussions finally lead to a decision on January 19th 1988 by the Provincial government, that one of the oil refineries had to start doing measurements. The oil refinery chosen, at that time the BP refinery in Göteborg (nowadays Preem Raffinaderi AB) , was considered to have the best location for a first trial of measurements. The reason for this was that the refinery at the time was a simple low-skimming facility, with the geographical positioning of the process area and the tank farms well separated. Also the infringement of emissions from other sources in the area could easily be taken care of as the distance to other emitting sources – also taking in account the prevailing wind direction – was considered to be more than sufficient. The decision was coupled with a fine of SEK 2 million – at the time some USD 300 000 - in case measurements and reporting were not carried out as decided by the authorities.

By coincidence BP at the time had already developed an in-house laser based DIAL-system (Differential Absorption Lidar) which had already been used inside of the BP group under the flag of BP Research.

The first measurements were carried out at the BP refinery (later Preem) in May 1988, in June 1989 and also in February 1992, before it was considered that it was without any doubt possible and feasible to use the system in an appropriate way to determine the true VOC-emissions also for the other Swedish oil refineries.

At the time of the initial measurements at the BP refinery, theoretical (API- and Radian-based) calculations had been used to get some rough idea of the VOC emission level. The emissions calculated showed that some 700 ton VOC/a could be estimated to be emitted. This was virtually turned upside down when the figures of the real emissions – based on the DIAL-measurements – were released during the autumn of 1988. The emission level at the refinery turned out to be about 10 000 tons/a instead, and in this figure the product tank farm was not included. With that included (it was first measured in the measurements in 1989), the real emission level in 1988 for the BP refinery could be estimated at some 14 000 tons/a, ie. 20 times higher than what the calculations showed.

The presentation of the measured figures to the public – in Sweden all these data are open to the public domain – resulted in a heated discussion in the papers and in subsequent meetings between the representatives of the environmental authorities of the Province and the management of the then BP refinery. These discussions resulted in a number of decisions, which showed to be of great value in the coming combat of the VOC-emissions, namely:

- the management of the BP Refinery confirmed that the measured values, although high, were reliable
- the management of the BP Refinery confirmed that they felt obliged to undertake actions in order to reduce emissions. As a matter of fact the measurements showed amongst others one single leak corresponding to some 4 000 ton VOC/a in itself. This leak was subsequently tightened up by the end of the measurements

- the management of the BP Refinery declared that they were willing to undertake a new measurement in about one years time in order to both confirm the results of the undertaken measurements and to receive a proof of the impact due to measures planned to be undertaken in the meantime before that measurement.

With this declaration by the BP Refinery a good basis, between the environmental authorities at the Provincial Government and the refinery itself, was laid for a mutual cooperation climate on these issues.

It was agreed that the coming measurements should also consider the possible impact of such ambient factors as wind speed and outdoor temperature as well as the impact of a shining sun. The results that were achieved showed that the impact of wind speed for some installations could be other than negligible, namely the tank storage area, but that a knowledge of normal average wind speed could be of good value in assuming normal average emisisons. Outdoor temperature as well as the impact of the sun rays on the other hand was shown to be of a negligible impact, this being specifically – and amongst others – proved by the measurements at the now Preem refinery during one of the coldest February-periods in 1992.

4. MEASURES UNDERTAKEN TO REDUCE EMISSIONS

Information on the current situation on implemented measures has been gathered for the two main fuel producing oil refineries and for the small bitumen-producing plant. Generally it can be noted that the different actions started with a major implementation phase as a result of the 1988 measurement results, which – as noted above - were staggering high.

On top of what is presented here it is also obvious that the refineries now pay much more attention to problems of VOC's and to the emissions of these pollutants compared to the situation only a decade ago. Today it is a normal part of life in the crude oil processing to think of solutions to keep down the VOC emissions, especially in case new facilities are designed, constructed and taken on stream.

The following measures to reduce emissions could specifically be noted:

4.1 Preem Raffinaderi AB, Göteborg

4.1.1 Tanks and other storage

- For oil pumps the sumps are covered and the trays tilted. At the place of the crude oil tanks the clear water pumps are vented directly to air.
- Inner floating roofs are equipped with primary seals (4 tanks)
- Blanket gas is used for three different tanks containing naphta and equipped with fixed copula roofs. In recent years the blanket gas has been changed from hydrogen-rich reformer off gas to nitrogen.
- Secondary seals on the outer floating roofs of the crude oil tanks.
- All product tanks with outer floating roofs have been equipped with a secondary sealing (excl. tanks with kerosene) as well as with equipment to reduce evaporation around the piping for level control.
- External fixed cupola roofs with internal floating roof equipped with primary and secondary seal on gasoline components, gasoline and slops tanks. The cupola roofs also play a role to avoid rain water entering the tanks.
- Drainage of tanks being better surveyed during operations. The drainage is led from the crude oil tanks to other tanks, ie. led back to a sludge tank and not directly to the WWT.
- A new type of roof drainage system is installed on the crude oil tanks. The size of the drainage devices have been decreased, allowing also a decrease of the area where VOC is exposed to the atmosphere.
- Changed roof drainage systems on all other tanks to abolish old piping which was due to leak VOC.
- Caverns are kept with a low filling degree and designed with a common gas phase to keep the pressure low and thereby diminishing the risk of evaporating or venting VOC's at all filling levels.

4.1.2 Process area

- Piston rod seals have been exchanged for products of the latest technique, mainly related to the material of the seal on the piston compressors.

- All control valves on the refinery re equipped with live-loading packing. All valves for manual operation on the new parts of the process area are from 1996/97 also equipped with live loading packing as well as also some other valves which – due to other reasons - have been up for exchange in recent years. All valves in service with light hydrocarbons are equipped with live-loading packing.
- Safety relief valves are led to the flare due to basic design by the plant in 1967.
- Pumps: In 1994 the first LPG-pump was equipped with magnetic drive. Now all LPG-pumps have been equipped with tandem seals (not pressurized). Pumps working with a magnetic drive amongst others are being used in service where H_2S is present in more than negligible concentration levels.
- All flanges serving light hydrocarbon streams are equipped with expanding graphite seals.
- For new process equipment the number of flanges are reduced by design.
- Flanges to purge or drain ends are either equipped with caps, blinded or plugged.
- Streams of product samples sent to on-line instruments to control specifications are returned to the processes are led to the flare.
- Most of the sampling stream to places for manual caught analyses are returned to the process or to the flare.
- A flue gas compressor installed in 2002.
- In line mixing of products is the general means of establishing final products.
- A leak detection and repair programme has been in full implementation for about 10 years.

4.1.3 Waste water treatment

- A settling tank of 10 000 m^3 has been installed before the WWT to reduce the hydrocarbon content to the API also enabling an uncovering of the API. Measurement tests will be undertaken to see if the uncovering is a possible option or not.
- The well to gather incoming water to the WWT is covered.
- The PPI-separator is kept covered by water, by which no further coverage is necessary.

4.2 Skandinaviska Raffinaderi AB, Scanraff, Lysekil

4.2.1 Tanks and other storage

- A balancing line in between tanks in light hydrocarbon service to improve pressure balancing and to reduce the risk of venting through safety relief valves.
- Secondary seals are being used on all tanks with floating roofs which are in service for products with a higher evaporating pressure than kerosene, in total 14 storage tanks.
- A new liquefied secondary seal installed on one of the crude oil tanks, following very high measured emissions by the DIAL trial in 1999.
- Vent gas from caverns is led to the flare instead of to the atmosphere.

4.2.2 Process area

- For four centrifugal compressors the vent gas is led to the fuel gas/flue gas system.
- For 13 piston compressors the leakage from the piston rod is led to the fuel gas/flue gas system.
- All pumps used for hydrocarbons with a density below 0,65 (at 200°C) are revamped and have new axis seals of tandem type.
- About 250 control valves in service with naphta and lighter hydrocarbons are equipped with improved packing material (graphite) which in some cases also is combined with a system based on springs.
- Valves run manually are all equipped with a new type of glandered packing (graphite rings in combination with a plait of carbon fibre)
- For flanges a spiraled graphite packing is used.
- Streams for on-line samples to GC's are led to the flue gas system.
- Streams for samples of LPG are equipped in such way that purge gas is led either back to the flare or returned back to the product.
- A leak detection and repair programme has been in full implementation for about 10 years.

4.2.3 Waste water treatment

A number of changes have in recent years been undertaken on the WWT in order to both reduce the amount of oil led to the plant and to reduce the amount of open space where oil can evaporate. This has been done by the following actions:

- Settling tanks with inner floating roofs prior to the waste water treatment to reduce the oil led to the WWT.
- Installing skimmers in a pre treatment basin to the API-separators system.
- Removal of oil at different underground culvert systems leading to the WWT.
- The waste water stemming from the product quay is led to the settling tanks instead of directly to the WWT.
- Total coverage of the API-separators.

4.2.3.1 Actions in 2002

During 2002 the WWT was rebuilt to enable the refinery to fulfil new emission limits set out in the license for the plant, specifically concerning the amount of suspended material and nitrogen in the effluent water. These changes were also used in order to improve the balance of the emissions to air at the WWT. Existing API-separators and flotation units were exchanged for new flotation units. The basin for pumping of waste water to the settling tanks, as well as to the flotation units, was completely covered and the gas recovered sucked off and led to the biological cleaning stage of the WWT. Also the biological cleaning stage was renewed. Existing equipment for supplying air were replaced with systems entering the air to the bottom of the basin. The new system for the cleaning of nitrogen in the water also should lead to a situation where the air supply is turned off from time to time in both of the basins where the air is supplied. This is presumed to also reduce the VOC-emission to air.

4.3 Nynäs AB, Göteborg

The refinery has in the late 1990's, after some staggering measurement results on the VOC-emissions in 1995, been introducing a complete system for vapour recovery for nearly all tanks on the refinery. The system is also continuously extended.

As the initial measurements were carried out in 1995 there were not any major emissions expected from the site, as nearly only heavy products were being produced and fed through the system. On the contrary very high emission levels were encountered due to the raised temperature in the bitumen tanks, held at around some 200 °C. This was contrary to all the old techniques for calculating emissions, where emissions from storing such heavy products by these calculation methods as a definition were set to zero. The measurements proved this completely wrong.

In the mid 1990's the refinery subsequently decided to introduce a complete system for vapour recovery at the tanks of the refinery. The system is divided in two parts, one where all tanks with non-oxidized products are put together, and one where the oxidized products are taken care of. There is also a connection in between the both systems to level the pressure out. The system uses nitrogen as blanket gas.

By later measurements it has been shown that the carried out actions substantially have decreased the emissions and on top of that an improved reliability in the processing has been achieved as the tanks, mainly those with oxidized products, now do not get choked at all, allowing for far fewer shut-downs of tanks and for far fewer cleaning operations than before.

Roughly the reduction in emissions from the tanks being put together in the vapour recovery system was reduced by half from 1995 until 1999, due to the system described above.

5. DESIGNING A MEASUREMENT SURVEY

Measurements of fugitive VOC emissions need both sufficient time to be carried out, and to be sufficient in area coverage. They also need to take into account variations in the meteorological circumstances during the measurement survey as well as its relation to the meteorological normal conditions.

It will not be possible to defend continuous measurements on the site by DIAL or any – if so – equivalent measurement technique at today's cost. The costs for such an exercise will be too high. On the other hand too short measurement periods will not give sufficient data, and will make the data received doubtable in both accuracy and relevance.

The methods for a good survey, in that the aim is to really sort out and define the real emission levels, vary from site to site depending on differences in both localisation and possible interference from other sources, topography and meteorology as well as fluctuations in the normal running of the facilities at the site. Never the less, below are proposed some basic rules to run a successful measurement exercise, based on the Swedish experience.

VOC's to be included

5.1 Define at an early stage which VOC's are to be included! For a petrochemical plant it might sometimes be possible to distinguish this to a few and well defined number of specified VOC's due to the production of well distinguished hydrocarbons. This does on the other hand not mean that in case of an ethylene-cracker you only can go for ethylene. You need also to measure ethane, propane, propylene, butane and aromatics and maybe also some other well defined VOC's to get the major part of the emission.

For oil refineries on a general basis there is a vast spread on which VOC's really are emitted. This means that a measurement should be covering the widest scope possible. With current existing equipment it is possible to measure alkanes and alkenes in the span $C_2 - C_{22}$. In case a too narrow span is used the figures measured will be too low compared to the real situation. As normally the share above C_{15} is low, it is sufficient to measure $C_2 - C_{15}$.

Aromatics should also be included and it is possible to measure at least up to some $C_{10} - C_{11}$ with today's techniques. On a GC that would correspond to about C_{15} when talking about the retention times of the straight hydrocarbons. The normal way is to use the DIAL-equipment for measuring one typical aromatic substance, normally toluene or benzene, and the other aromatics present are measured by sorption tube equipment in order to get a sufficiently proper value on their presence related to that aromatic substance measured directly by the DIAL.

Other VOC's to be taken care of are the cyclic ones with a cycle less than C_6, which could be included in the alkanes/alkenes-measurement set, although at a maximum they look to account for some 5-7% of the total, which on the other hand cannot be said to be negligible.

In case certain interest is lying within the field of methane as a green-house gas, this could of course also be measured by the DIAL, and should be done so in case it cannot be defined as a less important parameter for the plant. When describing emission figures it should however preferably be done separately for methane as the environmental impact of different types of VOC vary quite substantially. A proposed division would be the following:

SUBSTANCES	MEASURED (KG/H)	ANNUALIZED (TON/A)	REMARKS
Alkanes $C_2 - C_8$			
Alkanes $C_9 - C_{15}$			
Σ Alkanes			
Alkenes $C_2 - C_8$			
Alkenes $C_9 - C_{15}$			
Σ Alkenes			
Aromatics Benzene			
Aromatics Toluene			
Aromatics $C_8 - C_{11}$			
Σ Aromatics			
Cyclic hydrocarbons			
Σ NMVOC			
Methane			
Σ VOC			

This division should preferably be done for – primarily – the site as a whole but also for each of the main subsections defined for the site, normally at least the crude oil tank storage, the process area, the waste water treatment plant and the product tank storage.

The DIAL-system gives by its measurements in the normal operation mode, at a refinery or the like, levels of a sum of those VOC's that are detected for a certain wavelength. To get a picture of which VOC's are present it means that the DIAL has to be added to tube sorption measurements or other methods to get the full picture and the distribution. For this it is very important that the equipment used is able to detect all VOC's fully in the whole of the above described span, i.e. not only up to a level of $C_8 - C_{10}$ when talking about straight hydrocarbons, but up to about C_{15} instead, and subsequently also such a range for aromatics.

Below the measurement strategy is described mainly in terms of the use of the DIAL – or another equivalent system – as this gives the basic variation in emissions on a mass flux basis. To get the real figures in mass flux it is nearly equally important that the sorption tubes – or equivalent equipment – are used more or less in parallel to get the full picture. Without this, or in case a too narrow range is used for the VOC's, the received data will not give the full picture.

5.2 Meteorological measurements

The meteorology, as wind speed and direction, should be continuously measured at at least three heights during the whole time of each measurement activity. Normally the levels should be something like 5-8 m, around 10 m and 15 – 25 m above ground level, to get an accurate picture of the wind profile.

Continuous reliable information about the wind profile is necessary for getting an accurate measurement of the emissions from the facilities and continuous data on the wind direction is also basic information for defining the plume during the measurement as a whole, as it is the flux perpendicular to the plane that counts.

Another basic requirement for the measurement of the meteorological conditions is that the free air wind is given and that the met stations are placed in the **scan plane** so that the effect of possible partial wind shadows are accounted for.

5.3 Measurement strategy

5.3.1 The whole site

The running of an oil refinery, or the like, in itself contains a lot of parameters which in different ways can be varied and thus differently affect the operations and thereby also the emission levels. This is true for both the storage areas and the process area, although the general influence of day-to-day variations for the storage area, on a general level, is definitely greater than for the process area in the case where we do not talk of sudden leaks in the process or shut-down operations.

To receive reliable data, measurements therefore have to be undertaken in such a way that variations in normal operations are taken care of and, as much as possible, also are included in and analyzed during the measurement campaign. This means that surveys need to be undertaken over such a length of time that variations can be taken care of, and under such operative conditions within the site so that it during the measurements is possible to gather all necessary and relevant data for the later analyses and determination of emission mass flux levels. This has to be emphasized even more when measuring the tank storage area and the different off-loading operations. It is always recommended that, initially, an overall measurement of the whole site is carried out. This is also possible to undertake if the site is not all to big in size (up to some 1,2 * 1,2 km). In case the site is very large it is still of importance to get an initial overall picture of the emissions situation. This then has to be done by splitting the area up in sections with sizes which are possible to cover by the measurement device.

Each initial overall measurement should not be less than half a day, preferably one whole day. The time to be spent is also depending on the number of repeat visits aimed for and is of course also wind direction dependant. When the wind curtails the initial measurements, this could also be made up for later by the following measurements during the total measurement survey.

5.3.2 Division into Sub-sections

Having a broad picture of the overall emissions situation the next step is to focus on the defined sub-sections of the main area. For an oil refinery this would generally mean a division in at least the following areas:

- the storage facilities for raw material (crude oil etc.)
- the process area
- the waste water treatment area
- the product storage and
- other specific areas which might be of certain interest or by other means large emitters like ship, truck or railway loading

For other types of sites non-relevant parts of these could of course be omitted, i.e. for an oil harbour the "process area" generally is a non existent part.

The geographical and topographical parameters of course also have a general impact on this choice and generally there is no problem in a further division into sub-sections other than what is indicated above. Typically it could on top of this also be of interest to study just one or a few storage tanks or parts of the process area, mainly perhaps where there are new installations or parts of the plant which are suspected to have higher emissions than others. See also below chapter 5.3.3. In case the geographical area for the site is small (in relative terms) the number of specific sub-sections could be reduced, but then only when taking into account that the possibilities for interpreting the results are not hampered.

Measurement quantity is a tricky issue. As a general rule you can never have too much data but this has to be balanced against economics. The idea is to get sufficient data to cover the day to day or hour to hour normal operations and peaks. Overlaid on this will be the more abnormal peaks due to a whole range of accidental or maintenance activities etc. It is rare that you don't detect one or two of these 'abnormals' during the course of a survey, which however is positive as they anyway have to be taken care of in a correct manner to address a real annual emission level. An exclusion is debatable because, although the specific incident may be very unusual, there is infinite potential for other unusual incidents. Something unusual will be happening with a relative high frequency at complex sites, and it needs to be recognized that unusual incidents will add to the emission total of "normal operations".

As a guideline for a survey, a minimum of two to three days should be devoted to each sub-section. This time should be split up into at least four separate visits of 3 - 4 hours each at random choice of time during the total survey, but in conjunction with situations when the right conditions for measuring are met. Where a sub-section is very large it may be necessary to sub-divide it into even smaller parts with consequently less time spent at each spot. If several sub-divisions are necessary then the total time devoted to the whole main area preferably should be proportionately increased.

Specifying the number of scans to be carried out, i.e. single shot measurement of about 10 – 15 minutes, is often counter productive because scans can be shortened and coarsened, so a measurement time utilization is better to specify. This should consist of the system utilization time per day or for the whole survey. What is required is the actual measurement time of the system excluding between-scan setting and relocation time, although provision needs to be made for these. A good system should give over 4 hours a day of integrated measurement time, which is then also the basis for the timings described in this report.

As the emissions performance of the different sub-sections at an oil refinery are differing quite substantially the following advice would be given based on which type of sub-section we are focusing at:

6.3.2.1 Tank storage

Especially in case of outer floating roofs, emissions are expected to vary with wind speed and liquid level in the tanks. This proposes specific measurement activities to cover the impact of these parameters. Measurements should be carried out in such a way that they in the analyzing can be split up in different single and/or groups of tanks in order to enable a reception of data which can be used to implement measures to reduce emissions.

Typically one division is normally for crude oil tanks and product tanks respectively. Here it is very important to point out that the old traditional calculation methods more or less say that emissions are virtually zero for products heavier than kerosene. This is a huge mistake in these calculation methods, as the real measurements will show substantial emissions and this especially if such products are heated up above normal ambient temperature. Another mistake by the old calculations is the misinterpreting of the huge influence individual variations in between tanks, due to construction, history and maintenance, although they at a first glance look very similar.

The individual conditions of a tank, especially when looking at the larger tanks with outer floating roofs, has a sometimes tremendous impact on the real emissions. Sometimes emissions can be about fifty times higher or more compared to what is predicted by the old calculation methods, even if the liquid used is of kerosene type and lighter. Each measurement activity should be divided up into a sufficient number of scans so that enough information is gathered to enable an annualization as well as to have a good picture of the individual tanks with the highest emissions as well as a general picture of the variability of emissions due to wind speed and the filling height of the tanks. The latter especially is important for tanks with outer floating roofs.

5.3.2.2 Process area

Emissions do nearly not at all vary with meteorological conditions, but could be varying due to sudden leaks, changes in leak pattern and – in some cases – throughput as well as due to major changes in operational conditions. The relatively constant expected processing conditions could indicate that in some cases – when equal emission levels are measured from one time to another – the number of measurements to cover this subsection during the survey could be reduced to as low as two measurements of the above indicated length if the sub-divisions is not too large.

Measurements need on the other hand normally to be divided up into different parts of a site as the processing at many sites geographically is split up into different and well divided sub-divisions. If so, for each of these further sub-sections measurements have to be carried out as specified above. In a normal situation we talk of some two to three sub-sections.

5.3.2.3 Waste water treatment plant

Measurements of VOC-emissions from the WWT should be carried out in an analogy with those done for the tank storage. The variation of emissions with wind speed normally is far less compared to that of storage tanks. In the case where the WWT consists of a large open surface, emissions to air will normally both be high ("cleaning the water by letting the pollutants evaporate to air") and to some extent also affected by meteorological conditions. If settling tanks and other – intermediate storage facilities – are used in combination with the traditional WWT, their emissions should be measured separately.

The content of hydrocarbons in the effluent water and the mix of different types of hydrocarbons – and thereby also the corresponding mix in the emissions to air - will vary more than what is the case for the other areas. This indicates that the use of measurement device facilities to speciate the hydrocarbons need to be frequently used for the WWT plant.

5.3.2.4 Other facilities

Measurements around loading facilities should be carried out in such a way as to making it possible to arrive at some statistical sound level when looking at the normal operation, the working hours and other general performance parameters for the trucks as well as the railcar or ships being used. It is expected that there will be good possibilities to arrive at such measurement data when talking of trucks, as such operations are quite frequent, and railcars where they are frequently used. The aim has to be to arrive at typical emission levels for the specific operations and then sum that up to annual values depending on the number of such operations which are carried out as a whole, also taking into account typical daily start-up and shut-down situations.

Typically loading operations to truck often to a high degree take place outside of normal operation hours – quite frequently during early morning hours – which means that measurements need to cover this period also in an appropriate way.

5.3.3 Dividing sub-sections

When having a good picture of a certain sub-section, or even prior to that, it is recommended to also focus measurements on different already detected or expected hot spots. These could be defined due to many reasons of which some could be:

- Newly constructed plant at the site
- Plants with old equipment
- Plants where certain measures have been carried out to reduce emissions
- Plants where the strategy to reduce emissions would differ from other parts at a site
- Specific tank operations (such as major tanks with outer floating roofs)
- Parts of a sub-section or sub-division where specifically high emissions are expected or already have been initially measured

The latter is quite often due to "surprising" bad operations, typically in facilities like splitters, distillation towers or due to poor maintenance of storage tanks.

For each of these single spots, irrespective of its size, there should be counted up to 2-3 hours of effective measurements in order to get accurate data. In case spots with very high emissions are detected - and which are possible to tighten within short notice

- this should of course be done. The measurements however still have to be included in the reporting to show the actual measures that have been undertaken on the plant in order to display the real situation.

5.3.4 Conclusions

As described above the amount of time and the necessity to, for a single site, split measurements up is different from site to site. Still it is possible, based on the experience at the Swedish refineries, to foresee what an average measurement survey would look like in time and methodology when following the guidelines described in the chapters above.

It has also to be noted that the risk of arriving at disputable data due to not working according to the points outlined above cannot be ruled out as there always tend to be a discussion about the final contents, possibly leading to the need for new repeated measurements, thus making the whole story more expensive than it had been if it had been done in the right way from the beginning. Good planning and contact with the measuring team by personnel at the site therefore is an essential part of any survey.

As noted above a measurement day at the site should normally mean at least 4 hours of real collecting of data, which means concentration and meteorological data, the rest of the time allowing for accurate placing of the measurement devices (normally in a truck), tuning of instruments and adhering to the right wind directions. Data should also be analyzed daily, to in the best way configure the measurements for the coming days. Summing up the time needed for a measurement survey would thus – as a rule of thumb – look like the following number of days at the site for a measurement team:

- Measurement of the whole site: 2 days
- Subsections:
  - Crude Oil Tank storage: 2-3 day
  - Process Area: 2-6 days (for 1-3 sub-sections)
  - Waste water treatment: 2-3 days
  - Product Tank Storage: 2-3 days
  - Loading operations etc.: 2-3 days
- Other certain hot spots: 2-4 days

This makes out a total of 14 – 24 days which for small sites could be reduced to about one third, but for large sites even more time could be needed.

Preliminary reporting should already be made by the measurement team to the site at the end of the survey, but there should of course also be daily discussions with the responsible personnel at the site on the ongoing findings and the proposed coming measurements.

A final written report should normally be presented within one month from the last day of measurements.

6. MEASUREMENT RESULTS

As mentioned above DIAL measurements have been undertaken at 6 different Swedish sites since 1988. There have been 15 measurement surveys using two different systems, the Spectrasyne system with 12 surveys and Shell Research with the remaining three. Out of the 15 measurements, 10 are on fuel producing oil refineries, three on bitumen/ lube oil producing oil refineries and two on an oil harbour.

The easiest way to present the measurement results of all these surveys in a report like this would be to – for each single measurement survey – just present the data that were reported at the time of each measurement. As the systems and the methodology have been continuously improved throughout the years, this would however not give a really fair description of the results when comparing them with each other.

The presentation of, and the abilities to assess, the achieved measured data has been continuously improved. Initially only hydrocarbons in the range of C_3 – C_8 were measured as well as one of the aromatics, normally toluene or benzene. The aromatics-content has been shown to vary quite substantially with different areas of the refineries and there has also turned out to be a non-negligible content of hydrocarbons in the volatile part being heavier than C_8, which was the initial upper limit of chain length to be measured. Now up to C_{22} is measured. On the light side in recent years C_2 is now also included, initially only reaching down to C_3.

To make a true presentation of the real emission values the information of the old measurement surveys has to be processed together with the once recorded and presented figures to arrive at comparable and even – with today's knowledge - more exact data and to assess trends. It would be of very limited value, with current knowledge of what is being emitted and with the improved techniques of recording met data and tube sorption analyses, to go back to old methods for measurement and reporting. The methods for displaying results in this report are thus discussed below.

6.1 State of the art methodology

Today techniques exist to measure NMVOC's from C_2 up to about C_{22} in the non-aromatic range. Hydrocarbons in the heavy range above C_{15} only make out a small portion of the emission and could thus be exempted as they can be difficult to analyze.

Aromatics are possible to measure up to the same retention times as straight C_{15}. It is therefore no reason for not measuring these, as aromatics of this size seem far from negligible for the total mass flux.

For storage facilities it is also important that wind measurements are correct, and that it is possible to normalize emissions to air from the tank storage facilities to what is defined as the normal meteorological situation, mainly talking in terms of wind speed. There is an impact of wind-speed on the emission levels at the storage facilities. Specifically this impact is high when the wind-speed is very high and the impact has been shown to also have a slight exponential profile. On the other hand, very high wind speed is normally not the predominant situation but the impact of such situations should at least be addressed, in case there temporarily is a high windspeed when measuring. At the initial Swedish measurements, data was not specifically related to any normalized wind speed, but emphasis has been put on the issue in recent years establishing wind-normalized emission data. Normalization should then be done to a situation typical to the specific spot within a site where measurements are carried out, which means that for one refinery site there could be different average wind speed levels depending on the place of measurement.

The means of reducing this possible impact is of course by, during one measurement period, doing a number of repeated measurements at each of the different measurement spots. By experience the wind normalization at tanks could as a rule of thumb mean up to about +- 10-20% of the measured emission level, but of course less in comparison with the total measured emissions for the site as a whole.

As the DIAL-measuring system works in a real life situation, i.e. measuring the VOC which are passing through the measurement-plane, there are of course also emitted VOC's that do not get across that plane (the lower the wind, the higher the degree), which means that the DIAL-measured emissions always can be expected to be on the low side due to this. Trials carried out to get a better understanding of this phenomenon show that the maximum "lost" emission due to this normally would be about 10% of the real emission. In the figures below this "loss" is not taken into account, but is here anyway mentioned as it indicates that emission-levels in fact could be even higher than what is being measured and presented below.

6.2 Presented data

The data presented below consists of both comparable data for a number of years at those sites where the highest number of measurements have been carried out, the Preem and Scanraff refineries, as well as data showing typical changes in emission levels due to variations in the conditions of some of the equipment at the Scanraff refinery.

All presented figures are exclusive of methane, as methane has a completely different environmental impact than does the other VOC's – although they also within themselves show big differences in environmental impact – as methane is more of a green house gas than anything else. A rough guess is however that methane could add some 10-20% on top of the total emission as a rough approximation, and then of course varying with site, equipment, service and with time. Some specific measurements have also been carried out on the Swedish refineries to indicate typical levels of methane emissions for crude oil tanks and the waste water treatment. Methane in these exercises have amounted to 12-33% of the total NMVOC-emissions for the crude oil storage and being as high as 50-80% of the NMVOC-emissions from the WWT.

Measurements on the Swedish sites have been carried out with two different systems, the Spectrasyne (former BP Research) system which can measure both in the infrared (alkanes etc.) and the ultraviolet (aromatics) and the Shell Research system which only can measure in the infrared (alkanes etc.).

Combined with these systems meteorological measurements have been carried out as well as tube sorption measurements. The range of hydrocarbons covered vary with the system used. Generally it could be noted that emissions of VOC's could be expected up to at least the C_{15}-level (pentadecane). The Spectrasyne measurements have been carried out to meet this requirement, whereas Shell Research only reaches the level of some C_8– C_{10}. Spectrasyne, in contrary to Shell Research, also includes C_2 in the total and has also made some spot measurements on methane, although this is not included in normal VOC-figure, i.e. VOC-figures should in this report be looked at as NMVOC if nothing else is stated.

The conclusion is that it already from the beginning are different results to be expected from the two systems as the measurements are not done in an equivalent way. It is quite obvious that the Shell Research measurement device will record too low values compared to the real life situation as hydrocarbons heavier than C_8-C_{10} are not detected, and also not C_2. As the Shell Research DIAL does not include the ultraviolet it does not measure aromatics either which also makes the presented figures a bit more doubtable. Being aware of this, it does seem to be possible to in some way interpret the Shell Research measurements, although it will not be possible to in an accurate way say how much too low the presented figures are compared to the true total emissions as C_2 and hydrocarbons above C_8/C_{10} are left out and no aromatics are measured with the Shell Research DIAL. As noted above however in contrary though the Spectrasyne measurements fulfil the needed requirements.

Presenting single measurement surveys at a site only gives a rough indication of the emissions level at the site – still however much more reliable than any theoretical calculation – and is therefore not the best way of describing the emissions situation as the level of emissions may vary from time to time due to the condition of the site, both when talking about the tanks (i.e. conditions of seals and tanks as a whole) and the process area (sudden leaks etc.).

A better way of describing the emissions situation is to describe it for those sites where at least three measurement surveys have been carried out and where the measurement results can be compared with each other. The measurements should then preferably also be compared bearing in mind the current state of the art of measurements, which means that DIAL-measurements for aromatics, and hydrocarbons in the range of C_2 – C_{15} also should be taken into account.

To make this possible the choice in this report has been to present the data of measurement surveys for the Preem and Scanraff refineries during the period of 1992 – 1999, bearing in mind the current state of the art of the measurements. This means that the older and historically reported measured data is, where it has been shown to be needed, recalculated to the current standards to be comparable and to show trends. Data for the tank storage area has also been normalized to the wind speed on average being accurate for the area of the specific refinery. For Scanraff data is also presented for a few practical situations, showing the impact and value of real life emissions and the uselessness of old calculated data.

In the tables data are transferred from measured kg/h to tons/a by presuming an average of 8 600 hours/a of emissions a year. By this periods of maintenance and shut downs are taken into account. Of course minor individual differences do exist, but their impact can anyway be assumed to be below the total level of accuracy of the presented figures.

6.3 Preem Raffinaderi AB, Göteborg

AREA	1988		1989		1992		1995		1999	
	kg/h	tons/a	kg/h	tons/a	kg/h	tons/a	kg/h	tons/a	kg/h	tons/a
Crude Oil Tanks (Wind Normalized)	410	3500	350	3000	180	1590	90	790	80	700
Process Area	1640	14100	530	4600	115	1000	130	1150	170	1490
Waste Water Treatment	56	480	55	470	9	80	17	140	37	320
Product Tanks (Wind Normalized)	750	6400	750	6400	310	2700	270	2330	170	1470
Total	**2860**	**24500**	**1680**	**14500**	**620**	**5360**	**510**	**4410**	**460**	**3980**

Note: In 1988 emissions from Product Tanks were not measured, as the emissions were presumed to be low. As to make figures comparable, figures for these emissions have 1989 have been put in the table to also represent 1988.

6.4 Skandinaviska Raffinaderi AB, Scanraff, Lysekil

AREA	1992		1995		1999	
	kg/h	tons/a	kg/h	tons/a	kg/h	tons/a
South Tanks incl Crude oil (Wind normalised)	310	2700	90	770	350	3020
Process Area	380	3270	260	2270	230	2000
Waste Water Treatment	160	1350	80	690	55	480
Main Tanks (Wind Normalized)	660	5660	320	2760	430	3670
Total	**1500**	**12900**	**760**	**6500**	**1060**	**9160**

6.4.1 Example of tank storage conditions – tanks with outer floating roofs

A set of comparative measurements and analyses have been undertaken at a sertain set of tanks at the Scanraff refinery. The results are shown below in a table. As a background to the table the following should be noted: At the Scanraff refinery in 1992 high levels of emissions were recorded from tanks with Vacuum gasoil, because light hydrocarbons had slipped to the tank with the product, i.e. due to poor upstream operation. High emissions from crude oil tank Tk-1401 were recorded due to high liquid level being kept in the tank. In 1992 only two gasoline component tanks were equipped with secondary seals.

At the 1995 measurement survey all storage tanks with outer floating roofs had recently been equipped with secondary seals and thus maintenance work had been done quite close before the measurements, so emissions were measured to be very low.

In 1999 the emissions from the gasoline as well as the gasoline component tanks (with outer floating roofs) had increased due to presumed poor performance of the seals introduced. High emissions were experienced from the crude oil tanks. The inspection that followed due to the high recorded measurement results proved a number of leakages along the roof sealing of the crude oil tanks. Major maintenance works were carried out on two of the crude oil tanks and on the third the seals were changed to new ones. Unfortunately however no follow-up measurements were carried out to see the results of the latter installation.

Below the measured emissions at the tanks are presented as comparison-figures with the results related as factors compared to the old rigid calculation methods, ie. the true emission value is presented as a factor compared to what emission level is accounted for when only relying on calculations. In the table measured data from 1992 and 1995 is in this case not compensated to the 1999 state of the art knowledge, but it anyway quite clearly shows the dependence of good maintenance work and the need for frequent control on emission levels. Recalculating all data to 1999 standards would even clearer have shown the trends and the value of measured emission data compared to only calculated data, the latter not varying at all from time to time.

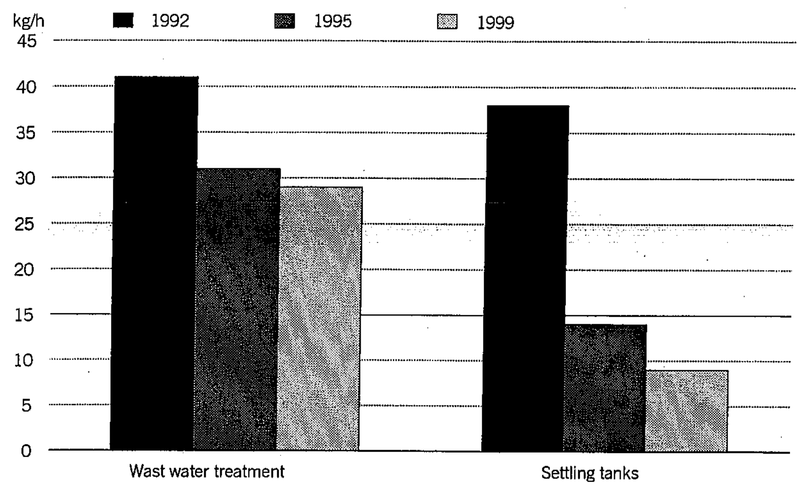
TANKS WITH OUTER FLOATING ROOFS	DIAL MEASUREMENT (YEAR)		
	1992 (factor)	1995 (factor)	1999 (factor)
Gasoline tanks	2,7	2,0	2,4
Gasoline component tanks	1,9	1,7	2,2
Crude oil tanks (Tk-1401 and Tk-1402)	26	2,6	13
Crude oil tank (Tk-1406)	-----	1,1	52

6.4.2 Rearranging the waste water treatment unit

Also for the waste water treatment unit a follow-up study has been carried out for the Scanraff the existing refinery. In 1992 at Scanraff settling tanks were used for ballast water only and they were also equipped with fixed roofs only. The API-separator was only partially covered.

At the 1995 measurement survey one of the settling tanks was equipped with an inner floating roof and was also put into a somewhat different service, being used as a settlingtank for both ballast and all other waste water produced at the site.

In 1999 both settling tanks were used for all waste water produced at the site and had also become equipped with inner floating roofs. The API-separators were now completely covered.



In the table data from 1992 and 1995 is not compensated to 1999 state of the art conditions, but anyway quite clearly show the dependence of good maintenance work and need for frequent control on emission levels. Recalculating all data to 1999 standards would have shown the decreased emissions even more clearly.

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