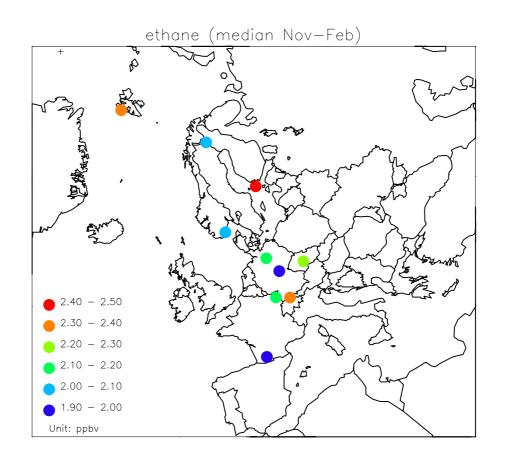
# **VOC** measurements 1999

**Sverre Solberg, Christian Dye, Michiel Roemer and Norbert Schmidbauer** 



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# EMEP Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

# **VOC** measurements 1999

Sverre Solberg<sup>1</sup>, Christian Dye<sup>1</sup>, Michiel Roemer<sup>2</sup> and Norbert Schmidbauer<sup>1</sup>

Norwegian Institute for Air Research
P.O. Box 100, N-2027 Kjeller, Norway

2) TNO – MEP
P.O. Box 342, NL-7300 AH Apeldoorn,
The Netherlands

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# Summary

This report presents measurements of VOC performed during 1999 at EMEP-VOC monitoring sites. In line with the recommendations made the national laboratories are gradually taking over the chemical analyses of the samples. In 1999 the official measurement data for hydrocarbons have all been analysed by the national laboratories. The official data for the carbonyls are analysed by the national laboratory in France for the French data, whereas EMEP-CCC at Norwegian Institute for Air Research (NILU) is responsible for the carbonyl analyses at all the other sites.

The results of parallel sampling and monitoring of VOC at a few sites in 1999 are presented, indicating in general a fairly good agreement for light hydrocarbons (at DE2, Waldhof) whereas larger differences was found for carbonyls. The agreement for formaldehyde was good, however. Although the number of monitoring sites is much too small to provide a regional picture of the VOC concentration distribution in Europe, the 1999 results indicate high concentrations of the natural gas components ethane and propane in northeast and higher concentrations of traffic related compounds, ethene and acetylene, in southeast.

Compliance monitoring is one reason behind the EMEP VOC monitoring programme. Due to the large spread in data, both with respect to day-to-day variation and interannual differences, and the limited number of sites, estimation of long-term trends are difficult. Except for NO1, Birkenes, the length of the VOC monitoring data are too short to draw conclusions on the concentration trends. The results from Birkenes do indicate a downward tendency of most compounds from the late 1980-ies to the late 1990-ies. There are, however, marked differences between the individual species, possibly reflecting different emission reductions for different source types in Europe.

# **VOC measurements 1999**

#### 1. Introduction

The Geneva Protocol concerning the Control of Emissions of Volatile Organic Compounds or their Transboundary Fluxes was adopted in November 1991. It entered into force on 29 September 1997. Three options for emission reduction targets are specified by the Protocol:

- (i) 30% reduction in emissions of VOC by 1999 using a year between 1984 and 1990 as a basis;
- (ii) The same reduction as for (i) within a Tropospheric Ozone Management Area (TOMA) and ensuring that by 1999 total national emissions do not exceed 1988 levels;
- (iii) Finally, where emissions in 1988 did not exceed certain specified levels, Parties may opt for a stabilization at that level of emission by 1999.

The EMEP VOC monitoring programme was initiated at the EMEP Workshop on Measurements of Hydrocarbons/VOC in Lindau, 1989 (EMEP, 1990). A three-fold objective of the measurement programme was defined at the workshop:

- Establishing the current ambient concentrations
- Compliance monitoring ("Do the emission control programme lead to a reduction of atmospheric concentrations?")
- Support to the transboundary oxidant modelling (prognostic and diagnostic)

The Workshop recommended that as a first step it would be sufficient with VOC monitoring at 10-15 rural sampling sites and taking two samples per week at each station centred at 12 noon GMT. Collection in stainless steel canisters and analyses by high resolution gas chromatography was recommended for the detection of light hydrocarbons, whereas impregnated adsorbent tubes sampling combined with high performance liquid chromatography (HPLC) was recommended for the detection of carbonyls. A list of required and desirable compounds was defined and is shown in Table 1.

Certain additional remarks at the Workshop were underlined in the proceedings report (EMEP, 1990). The need for more information on VOC concentrations close to the emission sources for modelling purposes was raised. Harmonisation with national urban measurement programmes was recommended as well as the assembling of VOC emission inventories. Furthermore, the importance of concurrent measurements of oxides of nitrogen was strongly emphasised.

At the Lindau Workshop it was also recommended that during the starting period the analyses of the VOC samples should be made by the CCC and that other laboratories should be included later on.

Table 1: List of volatile organic compounds that are "required" or "desirable" to measure within the EMEP programme as defined at the EMEP Workshop in Lindau, 1989 (EMEP, 1990).

	required	desirable		
Alkanes	ethane	hexane		
	propane	branched hexanes		
	i-butane	heptane		
	n-butane	branched heptanes		
	i-pentane	octane		
	n-pentane			
Alkenes	ethene	butenes		
	propene	pentenes		
	isoprene			
Alkynes	acetylene			
Aromatics	benzene	styrene		
	toluene	propylbenzenes		
	o-xylene	ethyltoluenes		
	m,p-xylene			
	ethylbenzene			
	trimethylbenzenes			
Aldehydes	formaldehyde	propionaldehyde		
	acetaldehyde			
Ketones	Acetone	methylethylketone		
		methylvinylketone		

The measurements of VOC within EMEP started with the collection of grab samples of light hydrocarbons in the middle of 1992, whereas measurements of carbonyls started in 1993. In the beginning five stations were included in the monitoring programme, Rucava (LV010), Košetice (CZ003), Langenbrügge (DE002), Tänikon (CH032) and Donon (FR008). Since then the number and selection of VOC measurement sites have changed several times.

The first laboratory intercomparison of light hydrocarbons in EMEP was organised already in 1993 (Romero, 1995). The variation or relative deviation among the laboratories were in a range  $\pm 25\%$  from the median. The exercise showed that the majority of the participating laboratories had the required analytical technique to correctly analyse a wide range of NMHC within an accuracy of  $\pm 10$ –15%. Furthermore, the results showed no substantial differences whether the air samples were analysed immediately after collection or after a period up to 2 months (for  $C_2$ – $C_5$  hydrocarbons).

The measurements are reported annually, and officially made public by the Steering Body of EMEP. There was no EMEP VOC report in 2000 as it was decided to postpone the reporting one year to bring it in line with the other data reporting in EMEP. Previous results from the EMEP VOC programme have been presented in annual reports (Solberg et al., 1993; Solberg et al., 1994; Solberg et al., 1996a; Solberg et al., 1997; Solberg et al., 1998; Solberg et al., 1999). An EMEP expert meeting on VOC measurements was organised in Berlin, 1994

(EMEP, 1995), and an evaluation of the measurement programme was made in 1995 (Solberg et al., 1995). Highlights and findings from the EMEP VOC programme have also been presented in a number of scientific papers (Solberg et al., 1996b; Hov et al., 1997; Solberg et al., 2001).

#### 2. Status of the measurement programme in 1999

#### 2.1 Status of station network

The location of the monitoring sites for VOC presented in this report is shown in Figure 1. An overview of the EMEP VOC measurement programme and the accompanying measurements presented in this report is given in Table 2.

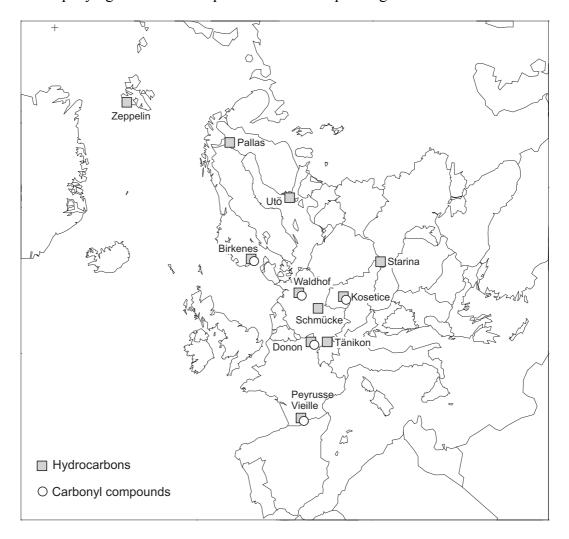


Figure 1: Monitoring sites for VOC in 1999.

As indicated by Table 2, data for 11 measurement sites for VOC have been reported to CCC and 4 of these with carbonyls. Carbonyls were also sampled at the Zeppelin Mountain station in 1999, but the data were not available for publication. VOC measurements were started at Peyrusse Vieille in France in July 1999.

Table 2: Status of the VOC monitoring programme in 1999. The columns give the station names, site code, and the sampling frequencies for hydrocarbons (HC) and carbonyl compounds (Carb). The laboratory responsible for the chemical analyses is also given. Additional laboratories taking part in parallel measurements are indicated in parenthesis.

Station	Code	HC <sup>1)</sup>	Lab. <sup>2)</sup>	Carb <sup>1)</sup>	Lab. <sup>2)</sup>	Comments
Zeppelin	NO042	Reg.	NILU			Only the first half year
Pallas	FI096	Reg.	FMI	n.m.	-	
Utö	FI009	Reg.	FMI	n.m.	-	(Carbonyl sampling started in April 2000)
Birkenes	NO001	Reg.	NILU	Reg.	NILU	
Waldhof	DE002	Reg.	UBA (NILU)	Reg.	NILU (UBA)	Parallel analyses of hydrocarbons first half year. Parallel analyses of carbonyls during all of 1999.
Schmücke	DE008	Reg.	UBA	n.m.	-	
Košetice	CS003	Reg.	СНМІ	Reg.	NILU	
Starina	SK006	Reg.	SHMI	n.m.	-	
Donon	FR008	Reg.	EMD	Reg.	EMD (NILU)	Parallel analysis of carbonyls January-April
Peyrusse Vieille	FR013	Reg.	EMD	n.m.	-	Sampling of HC started in July. Sampling of carbonyls started in 2000.
Tänikon	CH003	Con.	EMPA	n.m.	-	

- 1) Reg. = regularly, Con. = continuous, n.m. = not measured.
- 2) NILU = Norwegian Institute for Air Research
  - FMI = Finnish Meteorological Institute
  - UBA = Umweltbundesamt
  - CHMI = Czech Hydrometeorological Institute SHMI = Hydrometeorological Institute in Slovakia
  - EMD = Ecole des Mines de Douai (France)
  - EMPA = Swiss Federal Lab. for Materials Testing and Research

Extensive parallel sampling is carried out when the responsibility for chemical analyses are transferred to local laboratories. Long-term parallel sampling and/or analyses have been carried out at Starina, Košetice, Waldhof and Donon when the national laboratories took over the responsibility of the measurements. Thus, in 1999 there were no parallel measurements at Košetice or Starina, while at Donon and Waldhof the parallel analyses were carried out during the first part of the year only.

As in previous years, EMPA kindly shared their results from the continuous hydrocarbon monitoring at Tänikon with EMEP. A detailed comparison between the continuous monitoring and grab sampling at Tänikon was given by Solberg et al. (1996b) and Solberg et al. (1997).

Table 3 gives the sampling frequencies and the data coverage for the sites reported in this report. The term 'raw data' refers to the total number of samples reported to the CCC, and the fraction of rejected data is relative to this number. Note that 'rejected data' in this context refers to samples which are classified as outliers and rejected by inspection of the CCC. Outliers may arise due to either local pollution episodes close to the monitoring site, contamination of the samples

or errors in the chemical analyses. Normally the responsible laboratory removes samples which are wrong due to technical problems, thus there is always a screening (and rejection of samples) prior to the outlier detection carried out by the CCC.

Table 3: The number of samples of hydrocarbons (HC) and carbonyls (Carb) in 1999 available to NILU/CCC, relative to a recommendation of two samples/week (raw data coverage), as well as the fraction of data rejected by CCC due to local contamination or analytical error. The percentage of concurrent sampling of hydrocarbons and carbonyls (i.e. on the same days) is also given.

Station	-	data erage	Data rejected due to local contamination or analytical error		Net data coverage		Concurrence (HC and Carb)	
	(	%)		(%)		%)	(%)	
	НС	Carb	HC	Carb	HC	Carb	-	
Zeppelin a)	54	0	2	-	52	-	-	
Pallas	92	-	0	-	92	-	-	
Utö	98	-	1	-	97	-	-	
Birkenes	132	80	2	3	130	77	89	
Waldhof	84	106	0	0	84	106	82	
Schmücke	98	-	1	-	97	-	-	
Košetice	97	97	1	1	96	96	96	
Starina b)	92	-	(100)	-	(0)	-	-	
Peyrusse Vieille <sup>c)</sup>	37	0	1	0	36	0	-	
Donon	100	96	0	0	100	96	100	
Tänikon <sup>d)</sup>	con	-	0	-	con	-	-	

- a) Only measurements during the first half year on Zeppelin
- b) Preliminary data for Starina. Needs further evaluation
- c) VOC measurements at Peyrusse Vieille started in July
- d) Continuous monitoring at Tänikon

The net data coverage of Table 3 is the total number of samples reported from the responsible laboratories to the CCC subtracted the number of samples rejected by CCC. 'Concurrence', given in the last column of Table 3, denotes the fraction of hydrocarbon and carbonyl samples which were collected at the same days relative to the maximum possible number (based on the raw data). According to EMEP's recommendations, the samples should be taken twice a week, and the hydrocarbons and carbonyls should be sampled on the same days. The data in Table 3 are given relative to this recommendation, i.e. 104 samples/year. In practice, however, the sampling frequency will vary at the sites due to the removal of outliers occurring as a result of e.g. local pollution episodes or technical problems.

The table shows that in general the VOC measurements are satisfactory both with regard to sampling frequency, outliers and the concurrent sampling of hydrocarbons and carbonyls. Compared to previous years there has been a general improvement in these parameters. A 90% data completeness of daily values is given as data quality objective according to the EMEP manual (EMEP/CCC, 1995) and that is fulfilled at most VOC sites. The sampling at Zeppelin Mountain

is however, less frequent than recommended as it was carried out only the first half year. The hydrocarbon data reported for Starina was considered preliminary and awaits a further evaluation of sampling and analytical procedures.

The number of VOC monitoring sites is small. For hydrocarbons the number of sites is at the low end of the original recommendations of 10-15 set up in Lindau 1989 (EMEP, 1990). Carbonyls were only measured at four sites in 1999 which is much less than the recommendations. An effort to increase the number of carbonyl monitoring is therefore particularly needed. Carbonyl sampling has been started at Utö in 2000 with assistance from CCC/NILU as the responsible laboratory and a similar procedure could be applied at other sites.

## 2.2 Analytical procedures and quality control

The procedures for sampling and chemical analyses were similar in 1999 to previous years, and are not discussed in this report. A detailed description of the procedures used by NILU is given in the EMEP manual (EMEP/CCC, 1995). The technical procedures for the sampling and analysis of hydrocarbons by FMI at the two Finnish stations, as well as a site description and data interpretation, are given by Laurila and Hakola (1996). As noted previously, a NIST certified standard was used for the calibration of the samples in 1999 as opposed to the description in the paper by Laurila and Hakola (1996). A detailed presentation of the sampling and analyses performed by the laboratories at EMD (France), CHMI (Czech Republic) and SHMI (Slovakia) has been given previously (Solberg et al., 1998) and is not repeated in the present report. A comparison between grab sampling in canisters and the continuous monitoring of hydrocarbons at Tänikon has been reported previously (Solberg et al., 1996b).

At the new French VOC site, FR0013 Peyrusse Vieille, the hydrocarbons were measured by an automatic method using canisters with AVOCS and with sampling on Mondays and Thursdays. The conditions of sampling were the following:

• Starting time: 12 UT

• Duration: 4 h

• Air flow rate: 30 mL/min

• Purge time: 15 min

A standard operating procedure has been written by the responsible laboratory (Ecole Mines de Douai) and regular tests are realized for avoiding chemical contaminations.

For the EMEP VOC measurements in general, the quality control of the VOC measurements includes QA procedures at all stages from sampling to chemical analyses and integration. The QA procedures are described in the EMEP manual (EMEP/CCC, 1995) and are the laboratories' responsibility to follow up. In addition, data received from the individual laboratories are inspected before classified as valid or invalid by the EMEP/CCC. The routines for this is by visual inspection of plots and by several outlier tests as described in more detail in previous VOC reports (Solberg et al., 1997). First of all, seasonal or monthly average levels are compared with data from previous years and used to identify

serious shifts in the general concentration levels. Then, provided that sufficient data exists, each concentration value for the year reported is checked against the centred running mean and standard deviation using the data for several previous years, and assuming a log-normal distribution. If the new value is found to be more than  $4\sigma$  from the running mean of the previous years' data, it is flagged as an outlier. Whether the data value then is rejected or kept varies from sample to sample. Samples are rejected if contamination or other problems are likely. Rejection of sample values are done in agreement with the laboratory providing the data. Additionally, the data for several years together are also checked by a Rosner's test (Gilbert, 1987), suitable for detecting multiple outliers. Cross-correlation plots are also used to detect outliers in individual components as previous experience indicates well-marked correlations between pairs of hydrocarbons. The number of data rejected (classified as invalid) by these methods is indicated by Table 3.

#### 3. Results from parallel analysis

#### 3.1 Parallel analysis of hydrocarbons at Waldhof by NILU and UBA

In the first half year of 1999 (until early June) the hydrocarbon canisters sampled at Waldhof were first analysed by UBA's laboratory and then shipped to CCC/NILU for a second analysis. The resulting parallel analyses are shown in Figure 2.

For most components and except for a few outliers, these time series indicate satisfactory results. Except for the data for 7 January the results indicate a close relationship between the two time series for ethene, propane, propene, acetylene, n-butane and i-butane. Also for n-pentane, i-pentane, n-hexane, isoprene, benzene and toluene the relationship indicated by these time series are good except for a few more outliers. The results indicate a systematic difference in ethane during the first three months, with UBA's data being lower than NILU's, and good agreement after that. For the butenes, ethylbenzene and the xylenes the discrepancies are larger, indicating analytical difficulties for these compounds.

A statistical evaluation of the data is given in Table 4. The statistical parameters include the medians of the data from NILU and EMD and the median differences as well as the modified median absolute difference estimator, M.MAD, as described in the EMEP manual (EMEP, 1995) and the coefficient of variation, CoV, defined as CoV=(M.MAD)/(NILU's median). The analyses from the laboratory at CCC/NILU were regarded the reference in these calculations.

M.MAD expresses the spread of the data and approaches the standard deviation if the population has a normal distribution. CoV expresses the relative spread of the data, and, similar to the M.MAD, approaches the relative standard deviation for a normal distributed population. Both parameters are non-parametric statistics which make them particularly useful for trace gas measurements which normally show a non-normal distribution in the data. As recommended (EMEP, 1995) some extremes were removed from the statistical analyses, thus the statistical results for o-xylene and ethylbenzene are not really representative as most of the analyses turned out to be highly different between the two laboratories.

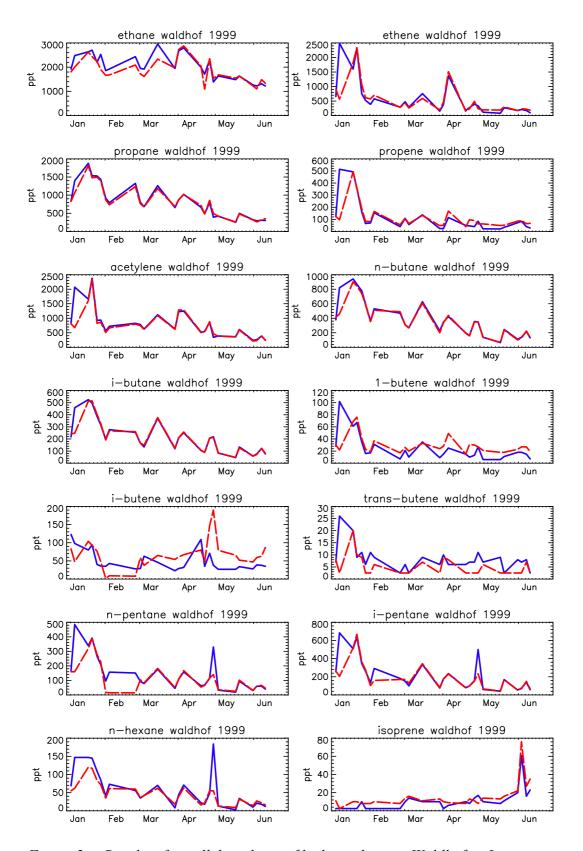


Figure 2: Results of parallel analyses of hydrocarbons at Waldhof in January – June 1999. Red dashed line marks canisters analysed by UBA. Blue full line marks the same canisters subsequently analysed by CCC/NILU. Unit: pptv.

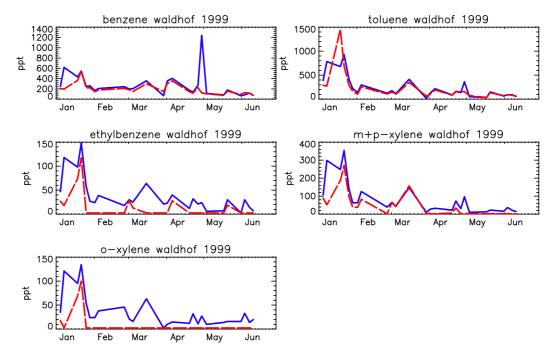


Figure 2, cont.

Table 4: Results from parallel analyses of hydrocarbons at DE002, Waldhof, during Jan-June 1999. The columns give the median of all samples as analysed by NILU and UBA, respectively, as well as the median difference and the modified median absolute difference estimator (M.MAD) and the coefficient of variation, CoV, defined as CoV=(M.MAD)/(NILU's median). A few outliers were removed from this analysis. Unit: pptv.

	median NILU	median UBA	median difference	M.MAD	CoV
ethane	1934.000	1765.000	-26.500	186.805	0.097
ethene	356.000	364.000	45.000	100.074	0.281
propane	793.000	748.000	-8.000	46.701	0.059
propene	68.000	84.000	14.000	18.532	0.273
acetylene	678.500	645.000	-27.000	33.358	0.049
n-butane	347.500	343.000	-6.500	15.567	0.045
i-butane	202.000	199.500	0.500	8.154	0.040
1-butene	16.500	26.500	8.500	5.189	0.314
i-butene	38.000	63.000	20.000	25.204	0.663
trans-butene	7.000	2.500	-2.000	2.965	0.424
n-pentane	94.500	103.000	2.000	6.672	0.071
i-pentane	163.000	162.500	0.500	12.602	0.077
n-hexane	43.000	40.500	-3.500	11.119	0.259
isoprene	10.000	11.000	5.000	3.706	0.371
benzene	205.000	179.000	-27.000	28.169	0.137
toluene	145.000	143.500	-16.000	31.875	0.220
ethylbenzene	27.500	15.500	-11.250	10.749	0.391
mxylene	64.500	40.500	-17.000	18.162	0.282
o-xylene	14.500	2.500	-12.000	4.448	0.307

The statistical calculations show that the CoV is less than 15% for a number of compounds. However, for ethene and propene the CoV is rather high even though the time series indicate a good agreement. This emphasises the importance of accompanying methods (as visual inspection of plots) in addition to tabulating the statistical parameters.

The conclusion from this evaluation is that for many of the components the agreement is satisfactory (and for some, as acetylene, remarkably good). The reason for the analytical problems with some of the aromats and alkenes should be studied further. The reason for the few outliers (i.e. totally different results between the laboratories) should also be cleared out. Based on such an evaluation supplied with similar studies for other laboratories, a list of selected hydrocarbons to report in the future accompanied with estimates of the total precision of each single component should be agreed on.

#### 3.2 Parallel analysis of carbonyls between NILU and EMD at Donon

Parallel sampling and analyses of VOC have been carried out at FR0008, Donon, since the French laboratory at EMD (Ecole Mines des Douai) started VOC sampling in 1997. The parallels of hydrocarbons were ended in 1998, whereas the parallels of carbonyl compounds continued until mid April 1999 when CCC/NILU's sampling was ended. In 1999 the parallel measurements were carried out by using both parallel sampling and parallel analyses. Separate sampling devices and DNPH cartridges were mounted and the samples were taken for the same time periods. The exposed cartridges were then shipped to the responsible laboratories which analysed them independently.

The time series of the carbonyls analysed by NILU and EMD are given in Figure 3 and a statistical evaluation of the parallel data are given i Table 5, similar to the statistical evaluation for hydrocarbons at Waldhof in the previous section. The results from the laboratory at CCC/NILU were regarded the reference in these calculations as well.

One outlier was taken out of the statistical calculations for each of propanal, benzaldehyde and glyoxal although they are shown in Figure 3. The time series clearly shows best agreement between the two laboratories for formaldehyde. For other compounds the differences are larger although a clear correlation is seen for most of the components. For some compounds the comparison is not possible as different detection limits obviously have been applied by the two laboratories. This regards in particular methyl vinyl ketone and methacrolein. All NILU's data for methacrolein were below detection limit and this compound was thus let out of the statistical analyses. However, also for propenal, butanal, benzaldehyde and glyoxal a large number of the samples from the laboratory at CCC/NILU were flagged as below detection limit, whereas data values were provided by the French laboratory for the same days.

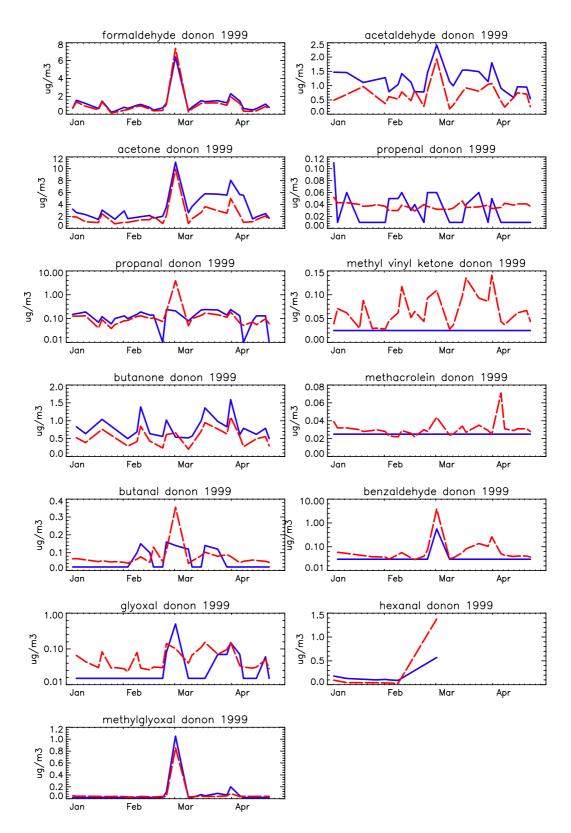


Figure 3: Results of parallel sampling and analyses of carbonyl compounds at Donon by NILU (blue full line) and EMD (red dashed line) in January–April 1999. Note the logarithmic axis for propanal, benzaldehyde and glyoxal (due to outliers for these compounds).

Table 5: Results from parallel sampling and analyses of carbonyl compounds at FR0008, Donon during Jan-April 1999. The columns give the median of all samples as analysed by NILU and EMD, respectively, as well as the median difference and the modified median absolute difference estimator (M.MAD) and the coefficient of variation (CoV). A few outliers were removed from this analysis. Unit: μg/m³.

	median	median	median	M.MAD	CoV
	NILU	EMD	difference		
formaldehyde	0.800	0.690	-0.177	0.104	0.130
acetaldehyde	1.120	0.692	-0.561	0.291	0.259
acetone	2.625	1.529	-1.197	1.027	0.391
propenal	0.010	0.037	0.025	0.010	1.038
propanal	0.130	0.099	-0.033	0.025	0.194
mvk	0.025	0.062	0.037	0.037	1.483
butanone	0.740	0.540	-0.262	0.085	0.115
butanal	0.020	0.057	0.030	0.015	0.741
benzaldehyde	0.030	0.041	0.011	0.014	0.469
glyoxal	0.015	0.034	0.015	0.015	0.988
hexanal	0.110	0.040	-0.066	0.010	0.094
methylglyoxal	0.020	0.039	0.017	0.007	0.371

The results show higher values when measured by CCC/NILU than EMD for formaldehyde, acetaldehyde, acetone and butanone. The CoV are smallest for hexanal, formaldehyde and butanone which all are below 15%. For the other compounds the CoV s in the range 15-100%. The precision of the carbonyl measurements (sampling and analyses) by NILU's laboratory on its own has been previously estimated at better than 15% for most compounds based on parallel sampling and analyses at NO0001 Birkenes. It is of course not surprising that the differences are larger when using different sampling tubes and different laboratories

Based on these results it is recommended to carry out a more detailed laboratory intercomparison for carbonyls taking into account all previous results for parallel sampling as well. As for the hydrocarbons, the aim should be to agree on detection limits and the selection of individual components to report to EMEP in the future.

#### 3.3 Results from the AMOHA project

In the EU FP5 project AMOHA (Accurate Measurements of Hydrocarbons in the Atmosphere) a large number of laboratories in Europe participated in parallel sampling and analyses of hydrocarbons in ambient air. Main results from the three different sampling periods (different times of the day) are shown in Figure 4 – Figure 6. The results show that except for a few laboratories the agreement is within  $\pm 25\%$  of the median for the lighter alkanes. For some aromats and unsaturated hydrocarbons as well as the  $C_6$ - $C_7$  alkanes a large spread in the values are seen, indicating measurement difficulties with these compounds. The spread in the results were, however, much less for laboratories using a NPL standard for

calibration. This is seen in Figure 7 which shows the results from sampling period 2 only for the sites which use the same NPL standard for calibration. Thus, it may be concluded that a large part of the differences seen among the laboratories reflects the use of different calibration gases. When using the same NPL standard the results from this intercomparison are very satisfactory.

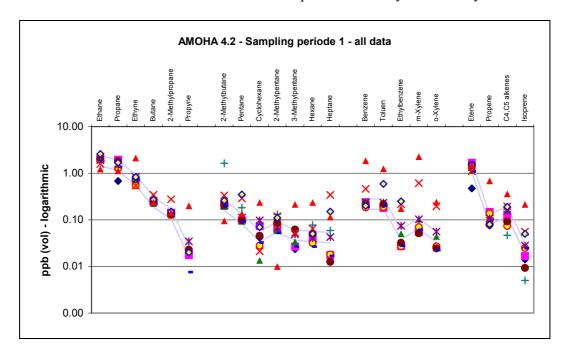


Figure 4: Results of the hydrocarbon measurement intercomparison (sampling period 1) in the EU FP5 project AMOHA. The symbols mark the average results from individual laboratories for a wide range of individual species given on top of the panel. The blue lines mark the median  $\pm 25\%$ .

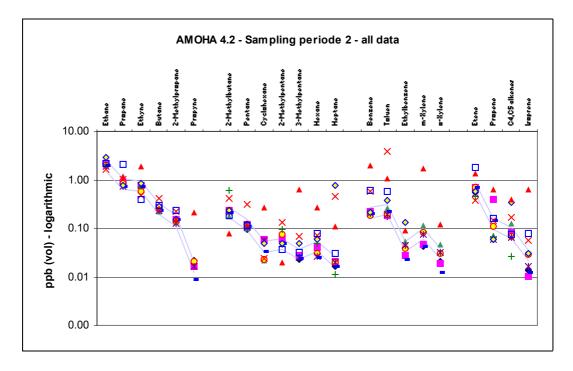


Figure 5: Same as Figure 4 for sampling period 2.

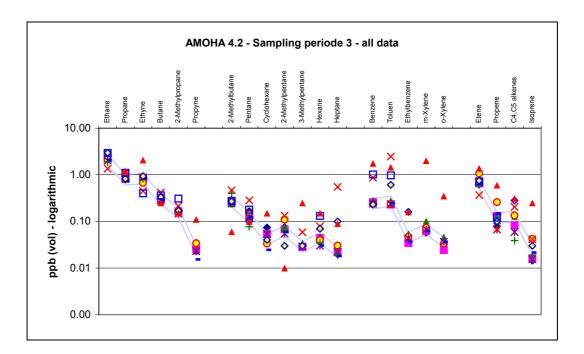


Figure 6: Same as Figure 4 for sampling period 3.

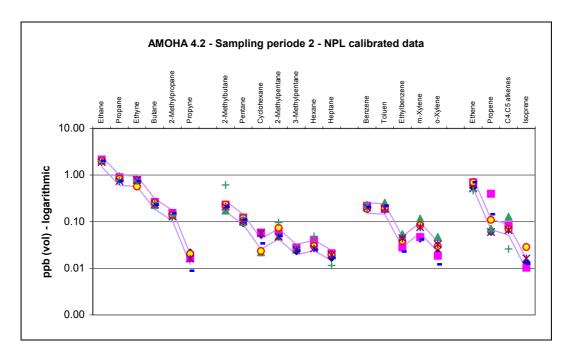


Figure 7: Same results as in Figure 5 but only for the laboratories which use a standard gas from NPL for calibration.

## 4. VOC concentrations in 1999 and long-term trends

#### 4.1 Regional distribution of hydrocarbons

Monthly mean and median concentrations of the individual hydrocarbons and carbonyls for 1999 are tabulated in Appendix A. The monthly statistics were not calculated if the number of samples were below four. Time series of all compounds during 1999 are given in Appendix B.

Figure 8–Figure 17 shows maps with the stations' median concentrations of 10 light hydrocarbons for the winter months January, February, November and December in 1999 taken together. Although the number of sites obviously is too low to give a clear picture of the regional background distribution of hydrocarbons in Europe, some characteristics are indicated by these results. Note, however, that there were no measurements at Zeppelin in November–December, thus the medians shown for Zeppelin will be relatively higher as the general concentration level is higher in January–February. Similarly, there were no measurements at Peyrusse Vieille in January–February, thus giving relatively lower medians than the other sites.

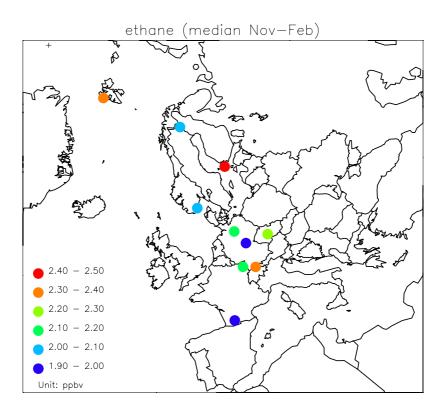


Figure 8: Median concentration of ethane at EMEP sites in the winter months November, December, January and February 1999 taken together.

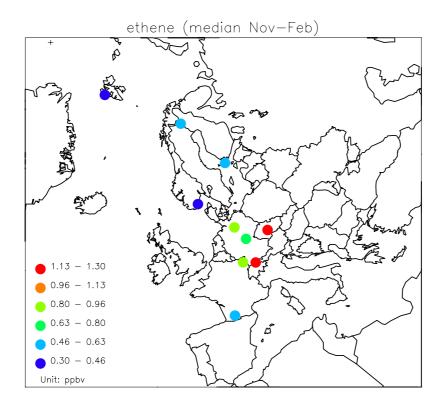


Figure 9: Median concentration of ethene at EMEP sites in the winter months November, December, January and February 1999 taken together.

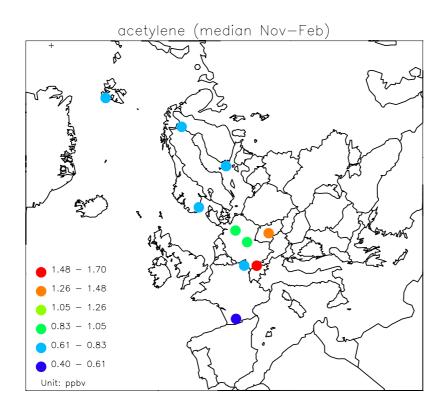


Figure 10: Median concentration of acetylene at EMEP sites in the winter months November, December, January and February 1999 taken together.

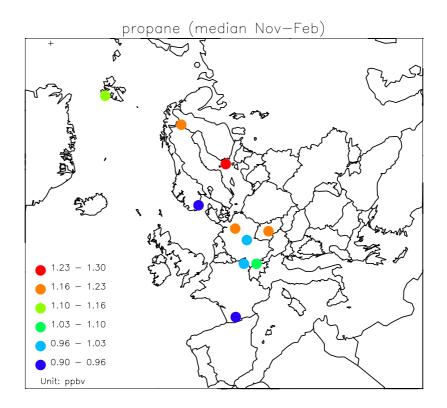


Figure 11: Median concentration of propane at EMEP sites in the winter months November, December, January and February 1999 taken together.

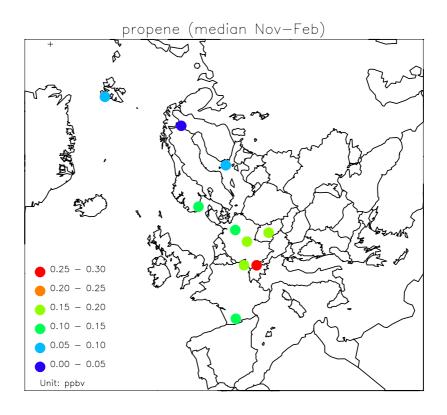


Figure 12: Median concentration of propene at EMEP sites in the winter months November, December, January and February 1999 taken together.

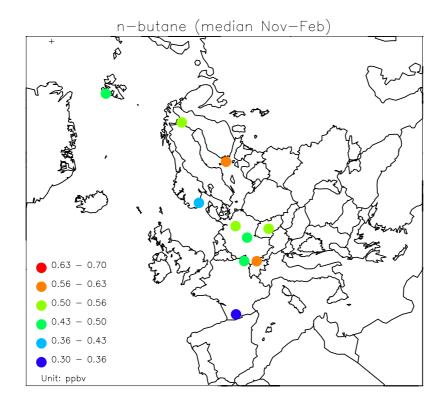


Figure 13: Median concentration of n-butane at EMEP sites in the winter months November, December, January and February 1999 taken together.

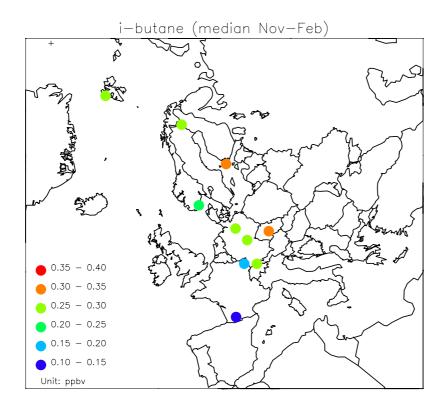


Figure 14: Median concentration of i-butane at EMEP sites in the winter months November, December, January and February 1999 taken together.

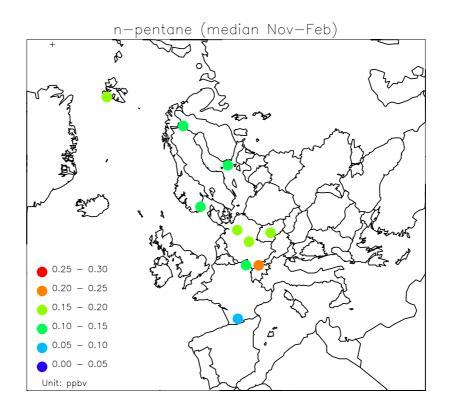


Figure 15: Median concentration of n-pentane at EMEP sites in the winter months November, December, January and February 1999 taken together.

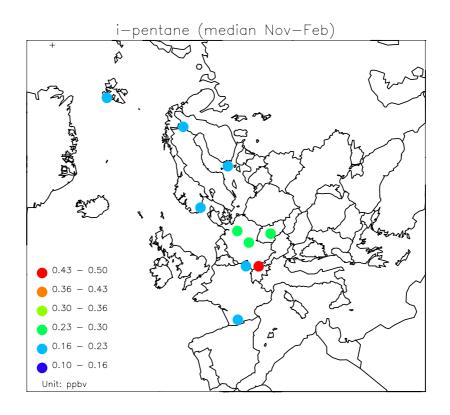


Figure 16: Median concentration of i-pentane at EMEP sites in the winter months November, December, January and February 1999 taken together.

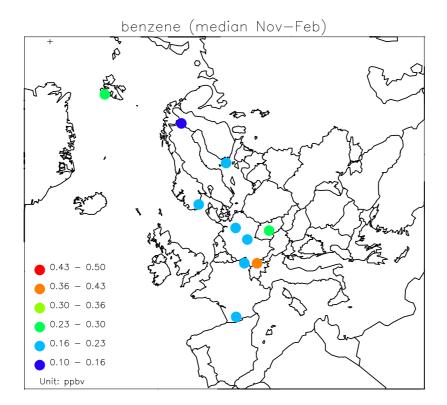


Figure 17: Median concentration of benzene at EMEP sites in the winter months November, December, January and February 1999 taken together.

As noted in previous reports, the measurements indicate that hydrocarbons become fairly well mixed in Europe in winter. Components indicative of natural gas emissions, ethane and propane, were highest at Utö when looking at the median for these months. For other components, e.g. acetylene, benzene and ethene the measurements indicate generally higher concentrations in southeast. In January and February the ethane and propane concentrations were particularly high at the northern sites (Pallas, Utö and Zeppelin Mountain). This may either indicate particularly high emissions of natural gas or more frequent and efficient atmospheric transport from regions with such emissions. To distinguish between these two effects numerical transport models would be needed. As a few examples, Figures 18, 19 and 20 show the air mass back trajectories calculated with the NOAA Hysplit code for three selected days with particularly high concentrations of ethane and propane at Zeppelin, Pallas and Utö, respectively. The trajectories for these three days indicate transport from Russia or Eastern Europe.

As the potential to form ozone varies considerably among the individual hydrocarbons, a detailed study to evaluate the distribution of VOC emission source types in Europe, based on measured VOC concentrations and transport pattern would be interesting but is beyond the scope and time limit of this report.

## NOAA AIR RESOURCES LABORATORY Backward Trajectories Ending- 12 UTC 08 FEB 99

FNL Meteorological Data

Verifical Motion Method - OMEGA

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Figure 18: Air mass back trajectories arriving at Zeppelin Mountain 8 February 1999 as calculated with the NOAA Hysplit code.



# NOAA AIR RESOURCES LABORATORY

Backward Trajectories Ending- 12 UTC 19 FEB 99

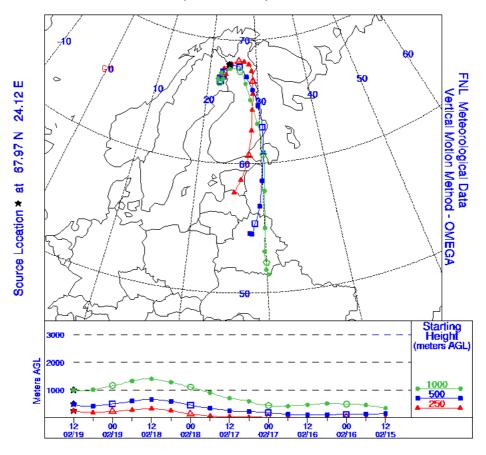


Figure 19: Air mass back trajectories arriving at Pallas 19 February 1999 as calculated with the NOAA Hysplit code.

## NOAA AIR RESOURCES LABORATORY

Backward Trajectories Ending- 12 UTC 29 JAN 99

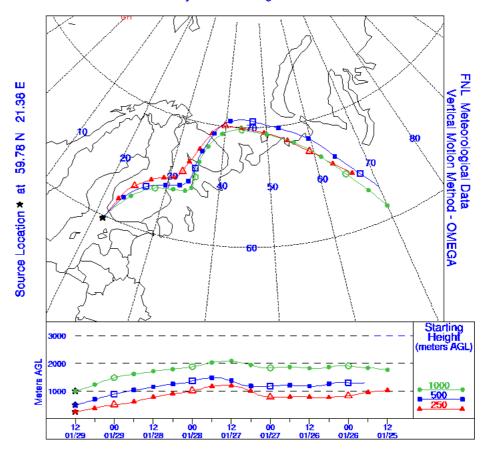


Figure 20: Air mass back trajectories arriving at Utö 29 January 1999 as calculated with the NOAA Hysplit code.

#### 4.2 Trends in hydrocarbons based on EMEP measurements

The compliance monitoring aspect is essentially the question: Are the trend in the measurement values of the trace gases consistent with the emission changes reported by the Parties to the Convention? In practice however, answering this question faces a number of difficulties. Most obvious, the large scale meteorology variations from year to year are decisive for the concentrations measured, thus effectively masking the trend in the underlying emissions. To distinguish between these two processes is practically impossible without detailed numerical dispersion models to validate and evaluate the measurements. On the other hand, with sufficiently long time series of trace gas measurements, indications of long term changes in emissions may be sought.

Figure 21 and Figure 22 show the variations in the median concentrations of selected hydrocarbons for the winter months January-March from 1988 to 1999

for the Nordic sites and for two continental sites, respectively. This time period was selected as it is the time of the year least affected by photochemical degradation and also the time when the overall regional differences in concentrations becomes most smoothed due to winter time mixing processes. These results indicate a general downward trend in several compounds, although the annual variation and spread in data values are large thus making the interpretation difficult and uncertain. The winter medians for the Nordic sites, with Birkenes having the longest time series back to 1988, indicate no trend in propane and acetylene and a slight downward trend in other components. The Birkenes data indicate clearly higher concentrations for most compounds except propane and acetylene in the first three years, 1988-1990, followed by a period of more stable concentrations in the 1990-ies. Unfortunately, the Finnish measurements started in the mid 1990-ies and the data from Zeppelin Mountain is sparse, not really allowing a trend analyses. The data indicate slightly higher values the last 1-2 years, particularly for ethane as noted above. This may, however, well be due to interannual variations in the large scale meteorological pattern.

The reason for the differences in annual variations among the different compounds is unclear. It might indicate less effective emission reductions in natural gas emissions than for other compounds. This is qualitatively consistent with the more effective cleaning of car exhaust due to the use of three-way catalysts on newer vehicles.

For the two continental sites, Waldhof and Košetice, the time series (from 1992) are really too short to make statements about the trends in concentrations. However, except for the peak year 1996, these data indicate a decrease as well. Interestingly, the winter medians for these two sites, which are about 500 km apart in the mid of the European continent, are strikingly similar, indicating their role as rural sites

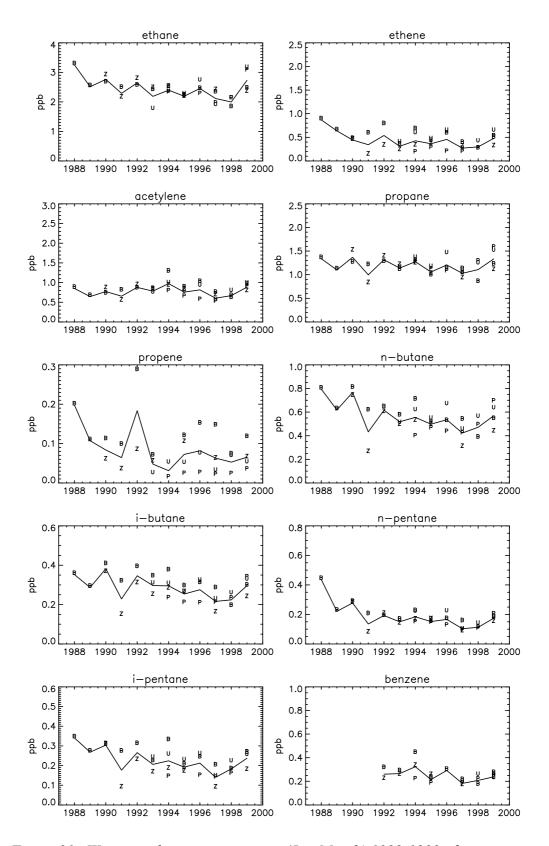


Figure 21: Winter median concentrations (Jan-March) 1988-1999 of hydrocarbons observed at the Nordic sites Zeppelin Mountain (Z), Pallas (P), Utö (U) and Birkenes (B) as well as the average of the stations' medians (line).

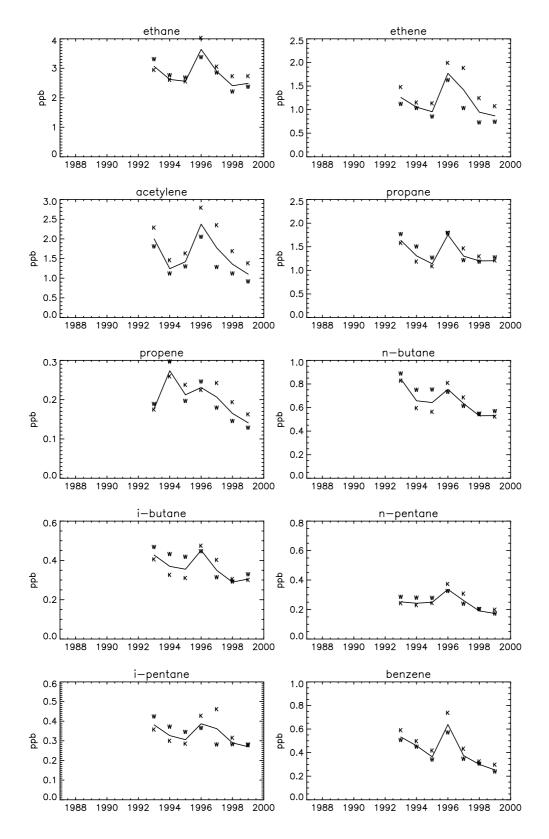


Figure 22: Winter median concentrations (Jan-March) 1988-1999 of hydrocarbons observed at the continental European sites Waldhof (W) and Košetice (K) as well as the average of the stations' medians (line).

## 4.3 Studies of VOC concentration trends within other European projects<sup>1</sup>

This chapter give a brief overview of the results of trend studies based on results from the EUROTRAC TOR-2 project and on other data. The studies are divided to the type of data (rural and urban). A general "complaint" is that there is a lack of observational data. It is certainly true that the size of the VOC databases is considerably smaller than of  $NO_x$  and  $O_3$ , but in the course of time interesting and useful time series of VOC have been constructed. Some of the data has already been used in trend studies, others await further evaluation.

#### 4.3.1 Urban and semi-urban measurements

In the United Kingdom 26 hourly speciated C<sub>2</sub>-C<sub>8</sub> hydrocarbons are routinely measured at 11 urban and 1 rural site since 1994 (Derwent, 1999; Derwent et al, 2000). The majority of the hydrocarbon concentrations show good correlation with benzene indicating that motor vehicle exhaust and evaporative emissions are the major source of hydrocarbon emissions at the UK urban background sites. The concentrations of the majority of the hydrocarbons have shown significant decreases, typically in the order of 20-40% between 1996 and 1998. At sites heavily influenced by industrial emissions, some of the hydrocarbon concentrations have shown reduced trends, no trends or sometimes upward trends.

In Sweden an urban network of benzene and other aromatics was initiated in 1992 (Lindskog, 2001). During the winter half-year weekly averaged samples of 8 speciated VOCs are taken at 10 urban sites. In the course of time the number of urban sites has increased to 39 in 1999/2000. The average of the 10 urban sites show a 60% drop in winter half-year concentrations from the winter 92/93 to the winter 99/00 (Svanberg and Lindskog, 2001). The data of the aromatics indicate a much stronger reduction than the trend in the urban total VOC emissions, which are calculated at -38% over the same period. It is not clear yet whether the discrepancy is due to a reduced share of aromatics in traffic exhaust, or that the urban sites are substantially influenced by other source categories, which have undergone much more change.

In the Netherlands the longest records of continuous VOC measurements are at Moerdijk nearby an industrial plant for which the sites were designed (Thijsse, 1983). Hourly data of C<sub>2</sub>-C<sub>5</sub> are collected for the 1981-91 period, daily data for the 1992-99 period. In the second period ethene, propene and acetylene are measured each day, C<sub>6</sub>-C<sub>12</sub> once every fourth day. A method to filter out the data contaminated by the refinery worked satisfactorily for the longer lived species, but seemed less appropriate for a very reactive component as propene (Roemer, 2001). Typical traffic components like acetylene and ethene showed a downward trend of about 50% over the 1981-99 interval (Figure 23), which is in excellent agreement with the trends according to the traffic emission inventory. The decrease starts in the mid 1980s and accelerates in the 1990s. For benzene and toluene downward trends of 3-4%/yr are found over the 1992-99 interval.

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<sup>&</sup>lt;sup>1</sup> This text is based on the TNO report R2001/244 ("Trends of ozone and precursors in Europe – status report TOR-2 Task Group 1"). The same text will also appear in the annual TOR-2 report for the year 2000.

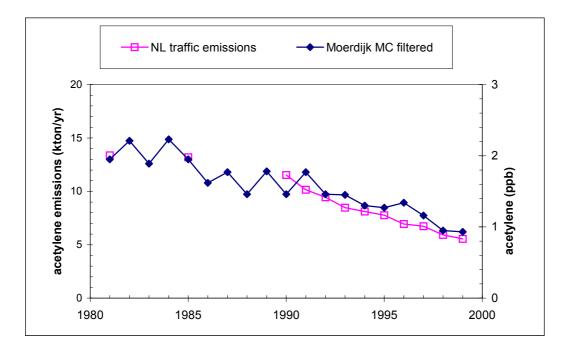


Figure 23: Trend of filtered Moerdijk MC acetylene concentrations (ppb) and trend of Netherlands traffic acetylene emissions (kton/yr) over the 1980-1999 period.

The urban data, and the non-urban data in The Netherlands and Germany have in common that the trends are downward, and very substantial, and that there is a good agreement with the emission inventory data. Urban data sites are predominantly traffic sites, and the agreement shows that the traffic emission information is consistent (in relative terms) with the observations.

In Switzerland long-term continuous measurements of VOC are available for Zurich (urban) and Dübendorf (near a road in an industrial area). At both locations VOC concentrations decreased with about 50% in the period 1986-98 (Kuebler et al., 2001). According to the Swiss emission inventory (SAEFL, 1995) the national Swiss VOC emission has dropped in the 1985-98 period by –42%. For the two main source categories, traffic and industry the reductions are –70% and –29% respectively. It is important to note that in these emission estimates the last few years are based on future projections.

#### 4.3.2 Rural measurements

At the Wank mountain site in southern Germany measurements of VOCs were carried out by grab samples during periods from March '87 to July '90 (part 1), and by means of an automatic device in the period January '96 to June '98 (part 2). In the first part samples were taken in the morning while in the second period the observations were done at night. Due to differences in sampling strategy and due to improvements in analytical techniques it is not possible to derive a trend from these two data sets.

At the site of K-puszta in Hungary VOCs are measured since 1988. Total NMVOC concentrations stayed fairly constant in the 1988-91 period and then

dropped steeply to much lower values in 1992 which is attributed to the economic crises that occurred in Eastern Europe (Haszpra et al., 2001). Comparing the 1988-90 period with 1992-93 the decrease in concentrations is in the order of 50-70% in the NE-SE-SW sectors and much smaller in the NW sector facing western Europe (and a part of Hungary). Since 1993 the concentrations stayed at a fairly low level, but since 1997 an increase seems eminent.

In general, the start of the VOC emission reduction is a few years ahead of NO<sub>x</sub>. Furthermore, the indication is that the traffic emission reduction is stronger than the national emission reduction, which suggest that the main other source (industry) is trailing compared to traffic.

#### 5. Conclusions and recommendations

The results of regular measurements of VOC at 11 rural European sites during 1999 have been presented. Light hydrocarbons have been measured at all sites and carbonyls at 4 of the sites. The data capture is satisfactory for most sites. Compared to the original plans for the EMEP VOC monitoring program the number of measuring sites are far less than the original recommendations. The carbonyl sampling is particularly sparse due to the requirements of manual sampling (attendance twice pr day) and also due to the generally less expertise with the measurement technique. More laboratories should be encouraged to initiate carbonyl sampling aided by the experimental and analytical support by CCC/NILU.

Parallel analysis and/or sampling were continued during parts of 1999 at DE002 Waldhof and FR008 Donon. The results indicate satisfactory results for most hydrocarbons at Waldhof analysed in parallel by UBA and CCC/NILU, with some outliers though. The butenes and some of the aromats were an exception to this with larger discrepancies between the two laboratories. Poorer agreement was found for the parallel sampling and analyses of carbonyls at Donon in general, although the results for formaldehyde were fairly good. For some of the carbonyls application of different detection limits at the two laboratories was the reason for major discrepancies.

It is highly recommended that a more thorough study of the parallel VOC data collected through the years is carried out with the aim to define a revised list of individual species to be reported in the future with a given accuracy. This should also be viewed in light of the original recommendations at the start of the VOC monitoring programme and in light of the specific need of the modellers.

The result of the VOC monitoring data for 1999 indicate high concentrations of species indicative of natural gas, ethane and propane, at the northeastern sites (Finland, Spitsbergen), whereas the highest concentrations of e.g. ethene and acetylene, tracers of traffic exhaust, was highest in southeast. It would be desirable to carry out a more detailed evaluation of the link between concentration level and transport regime as an attempt to identify the differences in VOC source types in Europe.

A simple evaluation of inter-annual trends in VOC was performed, looking at the development of the winter median concentrations at the EMEP sites using data from the start of the monitoring. At most sites the time series are too short or too sparse to draw statements of the long-term trends. At Birkenes where VOC have been measured since 1988 the data indicate slight reductions in most compounds. Due to the large variations from year to year it is difficult to make a quantitative estimate of the trend. The results show, however, considerably higher concentration of several VOC in the late 1980-ies compared to the late 1990-ies, qualitatively consistent with the reported overall drop in VOC emissions in Europe the last ten years. For propane and actylene no trend is apparent from the time series of the winter medians though. Results from trend analyses within other European projects and mostly from urban data indicate substantial reductions in the VOC concentrations, e.g. about 50% in the period 1981-1999 for Dutch and German data.

It is recommended to extend the trend studies mentioned above. In particular, it is essential to extract the interannual meteorological variations as far as possible to be able to evaluate the underlying trend in emissions. This, however, inevitable, requires some sort of long-term numerical modelling.

### 6. Acknowledgement

We would like to thank all people involved in the sampling and shipment of hydrocarbon canisters and DNPH tubes. We are very grateful for the VOC measurement data provided by Hannele Hakola (FMI), Stefan Reimann (EMPA), Patrice Coddeville (EMD), Jiri Honzak (CHMI), Rita Juneck (UBA) and Marta Mitosinkova (SHMI) who are responsible for the chemical analyses at the different EMEP VOC sites and who have reported the data to CCC. The AMOHA project was funded by the EU FP5 programme, project number SMT4-CT96-2075.

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## Appendix A

Monthly mean and median concentrations (first and second line, respectively) of hydrocarbons (pptv) and carbonyls (ng/m³) in 1999

Monthly mean and median concentrations (first and second line, respectively) of hydrocarbons (pptv) in 1999

					ETHAN	E						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Zeppelin	2075	2700	2443	1871	1361	-	-	-	-	-	-	-
	2025	2670	2323	1900	1348	-	-	-	-	-	-	-
Pallas	2822	3479	3194	2528	1601	1129	808	734	942	1200	1510	1749
	2841	3200	3199	2573	1690	1162	802	704	946	1077	1494	1814
Birkenes	2515	2237	2639	2112	1551	1267	1013	869	1424	1497	1573	1762
Directics	2430	2185	2593	2176	1554	1295	888	795	1274	1313	1438	1701
Utö	3256	2959	3019	2237	1711	1240	864	723	1109	1264	1660	2169
	2899	3236	2748	2212	1623	1143	915	735	1030	1161	1751	1721
Waldhof	2291	2256	2264	2141	1469	1147	1180	904	1301	1426	2083	2056
	2336	2201	2335	2347	1548	1112	1215	875	1084	1376	1968	1975
Schmücke	2165	2137	1885	1801	1373	1138	903	822	1083	1553	1888	1955
Ochmacke	2092	2147	1737	1860	1359	1124	890	772	955	1355	1716	1962
Košetice	2974	2679	2730	2431	1651	1181	933	903	1431	1511	2096	2035
	2427	2675	2741	2435	1561	1206	924	831	1105	1032	1594	1915
Tänikon	3134	2738	2718	2290	1701	1390	1142	1029	1339	1752	2489	2172
	2623	2792	2699	2238	1631	1435	1126	1050	1315	1660	2196	1965
Peyrusse Vieille						_	840	800	989	1220	1994	1927
i cylusse vielle	_	_	_	_	_	_	750	825	970	1205	1840	1910
Donon	1951	3159	2390	2210	1499	1256	1071	834	1159	1523	2036	2181
	1965	2810	2380	2230	1425	1250	1050	780	1130	1505	1820	2260
					ETHEN	E						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Zeppelin	289 279	353 330	260 265	101 92	91 100	-	-	-	-	-	-	-
	213	330	200	32	100	_	_	_	_	_	_	_
Pallas	757	525	388	140					44-			
	756				71	87	109	74	117	274	370	400
D: 1	756	537	331	89	71 44	87 80	109 78	74 71	11 <i>7</i> 124	274 132	370 241	313
Birkenes				89	44	80	78	71	124	132	241	313
Birkenes	729 563	537 312 227	331 539 519									
	729 563	312 227	539 519	89 377 353	294 285	213 206	78 324 251	71 303 296	124 426 389	132 477 370	241 520 352	313 473 388
Utö	729 563 1398	312 227 864	539 519 576	89 377 353 247	294 285 189	80 213 206 171	78 324 251 153	71 303 296 165	124 426 389 232	132 477 370 467	241 520 352 628	313 473 388 743
	729 563	312 227	539 519	89 377 353	294 285	213 206	78 324 251	71 303 296	124 426 389	132 477 370	241 520 352	313 473 388
	729 563 1398 563 1353	312 227 864 867 821	539 519 576 569 669	89 377 353 247 187 546	294 285 189 206 259	80 213 206 171 163 290	78 324 251 153 149 270	71 303 296 165 187 192	124 426 389 232 180 500	132 477 370 467 205 692	241 520 352 628 289 1238	313 473 388 743 529 813
Utö	729 563 1398 563	312 227 864 867	539 519 576 569	89 377 353 247 187	294 285 189 206	213 206 171 163	78 324 251 153 149	71 303 296 165 187	124 426 389 232 180	132 477 370 467 205	241 520 352 628 289	313 473 388 743 529
Utö Waldhof	729 563 1398 563 1353 1282	312 227 864 867 821 597	539 519 576 569 669 688	89 377 353 247 187 546 465	294 285 189 206 259 221	213 206 171 163 290 229	78 324 251 153 149 270 225	71 303 296 165 187 192 179	124 426 389 232 180 500 509	132 477 370 467 205 692 657	241 520 352 628 289 1238 1087	313 473 388 743 529 813 707
Utö	729 563 1398 563 1353	312 227 864 867 821	539 519 576 569 669	89 377 353 247 187 546	294 285 189 206 259	80 213 206 171 163 290	78 324 251 153 149 270	71 303 296 165 187 192	124 426 389 232 180 500	132 477 370 467 205 692	241 520 352 628 289 1238	313 473 388 743 529 813
Utö Waldhof Schmücke	729 563 1398 563 1353 1282 1487 1403	312 227 864 867 821 597 761 724	539 519 576 569 669 688 467 399	89 377 353 247 187 546 465 502 273	294 285 189 206 259 221 269 213	80 213 206 171 163 290 229 243 227	78 324 251 153 149 270 225 244 226	71 303 296 165 187 192 179 243 231	124 426 389 232 180 500 509 362 236	132 477 370 467 205 692 657 1008 658	241 520 352 628 289 1238 1087 994 759	313 473 388 743 529 813 707 960 752
Utö Waldhof	729 563 1398 563 1353 1282 1487 1403	312 227 864 867 821 597 761 724	539 519 576 569 669 688 467 399 845	89 377 353 247 187 546 465 502 273 464	294 285 189 206 259 221 269 213 225	80 213 206 171 163 290 229 243 227 242	78 324 251 153 149 270 225 244 226 379	71 303 296 165 187 192 179 243 231	124 426 389 232 180 500 509 362 236 456	132 477 370 467 205 692 657 1008 658 621	241 520 352 628 289 1238 1087 994 759	313 473 388 743 529 813 707 960 752
Utö Waldhof Schmücke	729 563 1398 563 1353 1282 1487 1403	312 227 864 867 821 597 761 724	539 519 576 569 669 688 467 399	89 377 353 247 187 546 465 502 273	294 285 189 206 259 221 269 213	80 213 206 171 163 290 229 243 227	78 324 251 153 149 270 225 244 226	71 303 296 165 187 192 179 243 231	124 426 389 232 180 500 509 362 236	132 477 370 467 205 692 657 1008 658	241 520 352 628 289 1238 1087 994 759	313 473 388 743 529 813 707 960 752
Utö Waldhof Schmücke	729 563 1398 563 1353 1282 1487 1403 1979 1348 2580	312 227 864 867 821 597 761 724 1150 1370	539 519 576 569 669 688 467 399 845 854	89 377 353 247 187 546 465 502 273 464 384 613	294 285 189 206 259 221 269 213 225 202 515	80 213 206 171 163 290 229 243 227 242 223 448	78 324 251 153 149 270 225 244 226 379 165 446	71 303 296 165 187 192 179 243 231 254 201 448	124 426 389 232 180 500 509 362 236 456 454 519	132 477 370 467 205 692 657 1008 658 621 423 1134	241 520 352 628 289 1238 1087 994 759 1575 1148 1858	313 473 388 743 529 813 707 960 752 1247 1097
Utö Waldhof Schmücke Košetice	729 563 1398 563 1353 1282 1487 1403 1979 1348	312 227 864 867 821 597 761 724 1150 1370	539 519 576 569 669 688 467 399 845 854	89 377 353 247 187 546 465 502 273 464 384	294 285 189 206 259 221 269 213 225 202	80 213 206 171 163 290 229 243 227 242 223	78 324 251 153 149 270 225 244 226 379 165	71 303 296 165 187 192 179 243 231 254 201	124 426 389 232 180 500 509 362 236 456 454	132 477 370 467 205 692 657 1008 658 621 423	241 520 352 628 289 1238 1087 994 759 1575 1148	313 473 388 743 529 813 707 960 752 1247 1097
Utö Waldhof Schmücke Košetice	729 563 1398 563 1353 1282 1487 1403 1979 1348 2580	312 227 864 867 821 597 761 724 1150 1370	539 519 576 569 669 688 467 399 845 854	89 377 353 247 187 546 465 502 273 464 384 613	294 285 189 206 259 221 269 213 225 202 515	80 213 206 171 163 290 229 243 227 242 223 448	78 324 251 153 149 270 225 244 226 379 165 446	71 303 296 165 187 192 179 243 231 254 201 448	124 426 389 232 180 500 509 362 236 456 454 519	132 477 370 467 205 692 657 1008 658 621 423 1134	241 520 352 628 289 1238 1087 994 759 1575 1148 1858	313 473 388 743 529 813 707 960 752 1247 1097
Utö Waldhof Schmücke Košetice Tänikon	729 563 1398 563 1353 1282 1487 1403 1979 1348 2580 1251	312 227 864 867 821 597 761 724 1150 1370	539 519 576 569 669 688 467 399 845 854	89 377 353 247 187 546 465 502 273 464 384 613	294 285 189 206 259 221 269 213 225 202 515	80 213 206 171 163 290 229 243 227 242 223 448 407	78 324 251 153 149 270 225 244 226 379 165 446 419	71 303 296 165 187 192 179 243 231 254 201 448 360	124 426 389 232 180 500 509 362 236 456 454 519 420	132 477 370 467 205 692 657 1008 658 621 423 1134 1074	241 520 352 628 289 1238 1087 994 759 1575 1148 1858 1448	313 473 388 743 529 813 707 960 752 1247 1097 1228 921
Utö Waldhof Schmücke Košetice Tänikon Peyrusse Vielle	729 563 1398 563 1353 1282 1487 1403 1979 1348 2580 1251	312 227 864 867 821 597 761 724 1150 1370 1375 1188	539 519 576 569 688 467 399 845 854 1221 1065	89 377 353 247 187 546 465 502 273 464 384 613 585	294 285 189 206 259 221 269 213 225 202 515 465	80 213 206 171 163 290 229 243 227 242 223 448 407	78 324 251 153 149 270 225 244 226 379 165 446 419 314 300	71 303 296 165 187 192 179 243 231 254 201 448 360 350	124 426 389 232 180 500 509 362 236 456 454 519 420 332 325	132 477 370 467 205 692 657 1008 658 621 423 1134 1074	241 520 352 628 289 1238 1087 994 759 1575 1148 1858 1448 868 755	313 473 388 743 529 813 707 960 752 1247 1097 1228 921 737 570
Utö Waldhof Schmücke Košetice Tänikon	729 563 1398 563 1353 1282 1487 1403 1979 1348 2580 1251	312 227 864 867 821 597 761 724 1150 1370	539 519 576 569 669 688 467 399 845 854	89 377 353 247 187 546 465 502 273 464 384 613	294 285 189 206 259 221 269 213 225 202 515	80 213 206 171 163 290 229 243 227 242 223 448 407	78 324 251 153 149 270 225 244 226 379 165 446 419 314	71 303 296 165 187 192 179 243 231 254 201 448 360 360	124 426 389 232 180 500 509 362 236 456 454 519 420 332	132 477 370 467 205 692 657 1008 658 621 423 1134 1074	241 520 352 628 289 1238 1087 994 759 1575 1148 1858 1448 868	313 473 388 743 529 813 707 960 752 1247 1097 1228 921 737

					PROPA	NE						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Zeppelin	984 917	1489 1429	1096 995	579 557	227 201	-	-	-	-	-	-	-
	0	0	000									
Pallas	1665	1713	1467	896	305	157	121	104	277	535	908	1154
	1558	1505	1575	935	326	137	91	99	280	436	914	1232
Birkenes	1269	1020	1261	757	432	296	347	287	557	619	666	841
	1201	956	1275	805	436	270	255	253	489	496	646	813
Utö	1657	1521	1400	737	363	188	149	165	406	572	911	1282
	1438	1594	1104	717	364	186	134	110	346	461	941	1095
Waldhof	1406	1354	1074	750	417	270	330	353	547	724	1074	1036
	1439	1232	1157	801	417	273	305	329	469	638	999	1134
Cohmiiako	1107	1106	071	704	257	272	207	244	402	625	075	060
Schmücke	1197 1149	1196 1137	871 805	704 692	357 325	272 255	287 273	244 230	403 323	635 546	875 796	969 943
Košetice	1419 1122	1298 1324	1128 1061	816 764	504 384	331 358	324 268	300 299	559 496	708 431	1023 883	1084
	1122	1324	1001	704	304	336	200	299	490	431	003	1188
Tänikon	1484	1286	1035	699	494	448	406	377	537	752	1226	955
	1203	1365	985	669	453	398	389	394	496	700	1084	851
Peyrusse Vielle	_	_	_	_	_	_	222	440	323	455	948	922
,	-	-	-	-	-	-	170	360	290	465	1010	920
Donon	801	1396	996	724	394	348	243	264	286	713	981	982
Donon	790	1335	910	780	305	280	255	200	350	650	820	1030
	JAN	FEB	MAR	APR	P <b>ROPE</b> I MAY	NE JUN	JUL	AUG	SEP	ОСТ	NOV	DEC
Zeppelin	89	65	103	53	50	-	-	-	-	-	-	-
• •	104	45	73	45	52	-	-	-	-	-	-	-
Pallas	47	32	29	20	22	22	29	13	25	31	44	37
i diid3	44	36	32	18	19	18	24	15	29	19	40	25
Birkenes	153 120	92 88	122 129	133 122	101 98	64 41	134 91	111 111	122 129	111 116	111 112	136 120
	120	00	120	122	30	71	31		123	110	112	120
Utö	139	101	49	37	38	45	41	35	39	60	89	87
	48	108	52	29	43	44	39	39	29	40	65	78
Waldhof	233	145	115	102	73	77	72	45	115	126	218	149
	220	96	111	90	62	66	67	45	74	135	167	139
Schmücke	243	120	73	95	61	63	54	54	106	172	207	198
Communic	223	104	67	54	66	65	48	59	50	158	143	176
12 × 11	000	474	400	0.4	-4		00		00	00	0.47	040
Košetice	236 199	174 166	100 88	64 58	51 41	52 46	36 30	41 32	62 49	92 72	247 149	219 166
	100	100	00	00	• • •	.0	00	02	10		1.10	100
Tänikon	608	291	227	119	104	115	90	91	97	248	409	264
	295	216	177	109	97	73	79	63	81	236	300	213
Peyrusse Vielle	-	-	-	-	-	-	134	122	90	103	140	124
	-	-	-	-	-	-	130	120	90	105	120	110
Donon	140	258	157	97	103	84	128	132	84	175	194	216
200.1	130	230	140	90	85	80	120	100	80	145	120	190

				Α	CETYLE	ENE						
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	677	937	759	453	266	-	-	-	-	-	-	-
	645	941	766	495	251	-	-	-	-	-	-	-
Pallas	895	969	906	524	216	121	74	63	142	290	496	571
	863	935	857	488	210	104	53	68	116	207	375	544
Birkenes	1109	775	1038	732	392	273	277	277	531	593	629	725
	948	754	1169	787	369	254	210	270	526	455	514	667
Utö	1264	1053	951	549	300	220	103	141	246	392	630	803
	925	1027	905	521	284	205	101	93	228	230	377	632
Waldhof	1213	1094	1033	827	440	236	275	262	477	728	1119	908
	982	835	1092	811	483	245	280	278	478	702	995	865
Schmücke	1451	1086	947	854	431	271	287	266	454	836	976	952
	1189	910	852	748	398	280	286	216	383	624	742	855
Košetice	1837	1430	1344	966	564	437	296	352	616	752	1541	1236
	1413	1323	1333	912	563	460	278	328	553	476	1439	1188
Tänikon	3156	2034	1322	867	714	856	508	492	711	1292	2270	1555
	1670	1706	1254	809	661	545	495	498	715	1112	1881	1233
Peyrusse Vielle	-	-	-	-	-	-	140 150	212 210	170 150	-	584 420	568 450
Donon	566	966	828	574	390	248	182	252	251	571	709	660
	605	1005	840	510	285	210	150	210	260	440	650	580
					N-BUTA							
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	403	631	402	154	41	-	-	-	-	-	-	-
	381	607	355	145	37	-	-	-	-	-	-	-
Pallas	915	749	581	282	85	61	41	33	98	198	417	516
	817	708	648	289	85	47	33	32	105	153	383	542
Birkenes	580	424	544	299	188	127	168	143	296	302	288	416
	566	388	557	305	183	126	142	123	257	230	272	430
Utö	761	698	549	250	101	73	71	103	170	221	414	577
	613	727	416	277	94	73	60	102	156	158	412	527
Waldhof	689	626	476	314	171	141	279	209	335	392	599	508
	665	509	517	349	137	128	228	197	309	295	457	481
Schmücke	621	560	395	259	167	153	191	146	240	315	416	487
	590	559	390	255	149	147	157	142	197	255	402	483
Košetice	670	499	444	285	188	155	212	150	277	309	504	490
	483	546	459	263	143	156	123	142	263	185	430	537
Tänikon	1221	741	614	405	400	439	363	361	492	548	810	550
	752	666	527	370	396	323	357	356	462	492	698	471
Peyrusse Vielle	-	-	-	-	-	-	80 70	134 150	87 100	165 165	354 360	322 320
Donon	320	623	719	300	221	226	328	162	180	428	450	462

					I-BUTA	NE						
Zoppolin	JAN 215	FEB 342	MAR 215	APR 90	MAY 27	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Zeppelin	198	330	189	88	26	-	-	-	-	-	-	-
Pallas	426	384	292	146	37	29	18	63	47	99	206	269
	441	349	322	144	33	20	15	19	48	70	200	279
Birkenes	313	223	337	161	92	65	86	72	162	160	161	209
	293	191	332	166	88	69	65	61	145	117	145	215
Utö	414	355	292	129	50	39	38	87	94	127	228	309
	309	387	222	135	46	35	33	35	87	88	215	274
Waldhof	386	347	274	179	102	80	161	119	177	216	315	271
	366	277	293	207	83	76	137	125	194	163	257	241
Schmücke	355	314	225	143	87	79	97	75	138	166	217	262
	330	325	234	148	78	80	84	67	102	137	206	247
Košetice	382	311	259	188	135	87	143	86	168	174	274	280
	267	329	264	161	85	91	77	89	169	98	241	288
Tänikon	545	342	289	176	169	168	145	132	189	239	369	246
	362	326	252	162	162	126	150	138	179	217	316	218
Peyrusse Vielle	-	-	-	-	-	-	36	66	39	-	156	130
	-	-	-	-	-	-	30	70	40	-	170	130
Donon	173	339	277	166	106	114	73	89	80	291	209	196
	165	320	290	160	65	80	70	60	90	225	170	200
					1-BUTE							
Zannalin	JAN 25	FEB 21	MAR 35	APR 16	MAY 16	JUN -	JUL	AUG	SEP	OCT	NOV	DEC
Zeppelin	22	18	30	15	14	-	-	-	-	-	-	-
Pallas	11	4	4	3	10	25	5	3	3	4	3	2
Fallas	10	3	3	3	3	26 26	5 6	3	3	3	3	3 3
Dirkonoo	20	22	20	22	22	10	22	27	22	20	26	25
Birkenes	32 28	23 22	30 32	32 32	23 23	19 22	33 23	27 29	33 35	30 30	26 27	35 30
142												
Utö	21 11	17 18	9 9	5 3	16 11	15 14	8 8	7 3	6 3	7 3	10 3	7 3
	• • • • • • • • • • • • • • • • • • • •	10	J	J		17	J	J	J	Ü	J	Ü
Waldhof	52	79	32	31	26	25	28	21	34	36	57	48
	55	41	28	30	21	27	27	22	27	38	43	46
Schmücke	51	27	25	26	22	23	23	19	26	37	37	48
	51	21	27	22	20	21	22	19	20	33	28	36
Košetice	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Tänikon	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Peyrusse Vielle	_	_	_	_	-	-	34	26	20	18	21	20
-	-	-	-	-	-	-	30	30	20	15	20	20
Donon	16	28	20	12	14	10	26	21	17	23	24	27
				-								
	15	25	20	10	10	10	30	20	20	20	20	30

					I-BUTEI	NE						
Zeppelin	JAN 38 39	FEB 31 30	MAR 43 36	APR 31 30	MAY 32 30	JUN - -	JUL - -	AUG - -	SEP - -	OCT - -	NOV - -	DEC - -
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
Birkenes	47 47	39 39	69 66	64 62	44 44	34 30	62 59	57 55	74 53	50 41	44 43	59 56
Utö	-	-	-	-	-	-	-	-	-	-	-	-
Waldhof	80 87	9 9	77 65	95 74	70 66	63 63	80 67	61 58	81 64	72 77	104 93	158 143
Schmücke	82 84	52 41	51 48	53 52	55 50	63 60	54 52	52 47	54 47	59 54	59 48	73 68
Košetice	-	-	-	-	-	-	-	-	-	-	-	-
Tänikon	-	-	-	-	-	-	-	-	-	-	-	-
Peyrusse Vielle	-	-	-	-	-	-	90 80	64 70	59 60	65 65	40 40	46 40
Donon	19 20	30 30	23 20	16 10	23 20	17 20	34 40	31 20	28 30	36 35	29 30	30 30
					ANS-BU							
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	ОСТ	NOV	DEC
Zeppelin	JAN 8 9	FEB 5 3	MAR 7 6				JUL - -	AUG - -	SEP - -	OCT - -	NOV - -	DEC - -
Zeppelin Pallas	8	5	7	APR 5	MAY 5	JUN -	-	-	-		-	-
	8 9 6	5 3	7 6 9	APR 5 5	MAY 5 6	JUN - - 6	3	- - 3	- - 3	3	3	- - 3
Pallas	8 9 6 3	5 3 3 3	7 6 9 3	APR 5 5 3 3 3	MAY 5 6 4 3	JUN 6 7 10	3 3 17	3 3 12	3 3 14	- - 3 3	- - 3 3	- 3 3
Pallas Birkenes	8 9 6 3 14 13	5 3 3 3 13 11	7 6 9 3 13 14	APR 5 5 3 3 3 15 16 3	MAY 5 6 4 3 11 12 4	JUN 6 7 10 8 8	3 3 17 10	3 3 12 11	3 3 14 12	3 3 11 12	3 3 7 6	3 3 3 12 14
Pallas Birkenes Utö	8 9 6 3 14 13 3 3	5 3 3 3 11 11 3 3	7 6 9 3 13 14 3 3	APR 5 5 5 3 3 3 15 16 3 3 6	MAY 5 6 4 3 11 12 4 3 4 4	JUN 6 7 10 8 8 3 5	3 3 3 17 10 3 3	3 3 3 12 11 3 3	3 3 3 14 12 3 3	3 3 3 11 12 3 3	- - 3 3 7 6 3 3	3 3 3 12 14 3 3
Pallas Birkenes Utö Waldhof	8 9 6 3 14 13 3 3	5 3 3 3 11 11 3 3	7 6 9 3 13 14 3 3	APR 5 5 5 3 3 3 15 16 3 3 6	MAY 5 6 4 3 11 12 4 3 4 4	JUN 6 7 10 8 8 3 5	3 3 3 17 10 3 3	3 3 3 12 11 3 3	3 3 3 14 12 3 3	3 3 3 11 12 3 3	- - 3 3 7 6 3 3	3 3 3 12 14 3 3 5 5
Pallas Birkenes Utö Waldhof Schmücke	8 9 6 3 14 13 3 3	5 3 3 3 11 11 3 3	7 6 9 3 13 14 3 3	APR 5 5 5 3 3 3 15 16 3 3 6	MAY 5 6 4 3 11 12 4 3 4 4	JUN 6 7 10 8 8 3 5	3 3 3 17 10 3 3	3 3 3 12 11 3 3	3 3 3 14 12 3 3	3 3 3 11 12 3 3	- - 3 3 7 6 3 3	3 3 3 12 14 3 3 5 5
Pallas  Birkenes  Utö  Waldhof  Schmücke  Košetice	8 9 6 3 14 13 3 3	5 3 3 3 11 11 3 3	7 6 9 3 13 14 3 3	APR 5 5 5 3 3 3 15 16 3 3 6	MAY 5 6 4 3 11 12 4 3 4 4	JUN 6 7 10 8 8 3 5	3 3 3 17 10 3 3	3 3 3 12 11 3 3	3 3 3 14 12 3 3	3 3 3 11 12 3 3	- - 3 3 7 6 3 3	3 3 3 12 14 3 3 5 5

				N	-PENTA	NE						
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	141	225	123	35	11	-	-	-	-	-	-	-
	138	215	106	37	8	-	-	-	-	-	-	-
Pallas	259	212	152	54	18	15	9	5	17	44	82	93
	264	207	166	54	14	11	6	5	16	30	67	93
Birkenes	198	129	183	89	58	41	70	58	112	110	104	132
	176	111	183	85	56	38	60	57	100	90	78	127
Utö	219	177	156	53	23	20	15	20	36	51	85	99
	199	188	121	45	21	19	15	13	34	36	48	91
Waldhof	285	22	187	106	66	68	128	87	137	176	248	177
	253	17	183	107	63	48	100	85	128	180	177	162
Schmücke	223	187	116	86	75	65	79	63	111	126	150	166
	200	191	114	83	70	56	80	52	85	104	122	162
Košetice	256	174	167	120	111	73	117	79	140	143	191	196
	176	198	174	100	60	71	74	90	144	83	177	196
Tänikon	438	263	244	153	167	179	167	148	213	263	329	194
	261	238	221	139	141	131	151	141	203	233	270	175
Peyrusse Vielle	-	-	-	-	-	-	24 20	36 40	26 30	50 45	114 100	87 90
Donon	88	174	144	108	88	83	76	60	73	168	189	137
	75	160	140	100	55	50	70	30	70	150	150	150
					-PENTA				0=0			5-0
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	162	256	139	40	11	-	-	-	-	-	-	-
	156	249	130	42	7	-	-	-	-	-	-	-
Pallas	426	287	212	82	32	25	14	19	26	60	132	149
	353	280	244	76	19	12	8	20	24	43	115	165
Birkenes	295	186	270	168	113	79	128	105	211	169	158	205
	283	201	269	142	119	78	115	103	187	138	116	185
Utö	329	273	214	84	37	28	36	41	59	92	153	191
	271	309	172	85	25	26	31	39	64	54	159	184
Waldhof	385	211	290	167	111	102	247	162	256	242	418	255
	320	176	293	174	66	68	204	158	195	219	258	234
Schmücke	379	286	207	134	122	120	151	130	218	210	248	268
	348	267	221	135	130	109	124	115	161	187	197	262
Košetice	392	265	244	175	176	131	234	138	232	195	325	307
	280	266	268	165	104	127	108	165	248	125	307	282
Tänikon	1054	551	491	314	380	458	371	375	504	519	687	409
	531	430	411	275	377	291	350	355	449	437	571	351
Peyrusse Vielle	-	-	-	-	-	-	52 50	90 110	67 70	123 105	218 220	171 150
Donon	159	294	267	173	160	164	130	134	143	314	324	254
	150	305	240	140	85	90	110	80	140	265	270	270

				N	N-HEXA	NE						
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	59	96	46	13	6	-	-	-	-	-	-	-
	56	89	40	15	3	-	-	-	-	-	-	-
Pallas	50	42	33	10	7	4	4	3	3	14	26	35
	55	40	37	9	3	3	3	3	3	11	26	36
Birkenes	88	55	79	31	21	14	22	34	39	44	48	88
	80	49	75	27	22	16	19	18	30	33	38	76
Utö	45	40	31	10	6	5	10	3	9	20	26	43
	47	43	23	7	3	4	8	3	4	10	17	39
Waldhof	98	73	58	43	24	23	61	30	47	54	93	64
	82	63	62	48	19	19	34	28	47	50	67	62
Schmücke	78	68	40	27	24	21	27	21	40	48	57	59
	73	76	38	25	19	19	23	16	24	37	44	56
Košetice	132	73	53	60	54	35	42	32	54	51	73	62
	76	75	51	63	27	35	29	31	59	28	67	52
Tänikon	134	81	70	39	43	45	41	39	50	66	84	51
	80	70	62	37	39	31	41	34	46	59	68	43
Peyrusse Vielle	-	-	-	-	-	-	24 20	52 40	14 10	13 10	34 30	29 30
Donon	36	75	57	39	25	30	13	24	21	65	60	56
	30	65	50	40	10	10	10	10	20	30	50	40
					SOPRE							
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	3	3	3	3	3	-	-	-	-	-	-	-
	3	3	3	3	3	-	-	-	-	-	-	-
Pallas	3 3	3 3	3 3	3 3	3 3	19 13	50 23	13 16	8	3 3	3 3	3
Birkenes	10	8	10	70	81	14	73	181	213	124	84	11
	10	8	3	12	17	8	31	116	140	122	9	9
Utö	3 3	3 3	3 3	3 3	3 3	8	4 3	3 3	4 3	4 3	3 3	3 3
Waldhof	8	6	12	17	15	44	110	81	58	29	18	12
	9	8	13	10	14	34	96	81	56	29	16	8
Schmücke	11	4	7	10	28	32	59	27	35	20	9	12
	12	3	6	9	20	29	51	25	34	19	7	11
Košetice	16	9	6	10	67	35	102	62	56	11	19	10
	9	7	3	7	30	34	68	46	57	10	13	3
Tänikon	27	12	12	11	33	46	80	65	61	26	19	15
	16	10	11	10	32	38	69	63	57	24	18	13
Peyrusse Vielle	-	-	-	-	-	-	990 990	1050 1150	770 780	115 120	15 10	10 10
Donon	40	23	38	113	344	556	983	954	769	209	83	17
	35	15	30	100	200	400	1090	680	620	175	40	10

					BENZE	NE						
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	205	270	218	167	85	-	-	-	-	-	-	-
	194	271	214	161	72	-	-	-	-	-	-	-
Pallas	264	240	233	129	55	40	24	20	36	67	126	150
	254	264	234	122	53	32	12	22	36	49	97	158
Birkenes	312	203	307	213	130	95	87	91	185	194	182	217
	266	194	333	223	128	81	64	95	171	143	141	196
Utö	326	231	223	131	66	41	46	31	91	107	170	206
	257	198	232	123	71	40	45	30	65	63	112	169
Waldhof	304	282	258	213	124	85	146	110	166	202	305	216
	253	212	253	221	129	77	136	108	169	192	271	207
Schmücke	381	281	233	217	120	98	107	93	149	249	264	236
	297	229	224	188	110	100	97	76	116	175	196	217
Košetice	452	314	271	232	149	101	107	122	174	188	326	249
	332	299	267	195	138	99	97	110	149	123	335	248
Tänikon	885	538	371	230	209	200	170	176	249	354	593	381
	462	445	323	217	196	163	166	159	257	320	462	314
Peyrusse Vielle	-	-	-	-	-	-	108 110	96 90	79 70	128 125	214 190	186 180
Donon	181	305	242	180	116	113	99	120	102	211	257	217
	185	305	240	170	95	80	90	100	120	170	250	220
					TOLUE	NE						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Zeppelin	233 255	188 139	69 63	91 39	97 11	-	-	-	-	-	-	-
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
Birkenes	341	196	245	146	111	93	116	140	236	165	168	264
	334	193	252	156	106	90	79	131	199	163	151	216
Utö	-	-	-	-	-	-	-	-	-	-	-	-
Waldhof	472	268	232	154	104	80	298	155	217	240	377	216
	288	191	213	158	75	58	150	148	151	212	275	198
Schmücke	368	270	169	144	128	147	147	116	239	234	274	276
	345	234	165	138	99	145	162	102	182	218	241	271
Košetice	356	244	179	263	119	110	107	116	131	121	224	190
	326	217	178	177	90	96	84	107	130	87	206	184
Tänikon	1469	719	588	291	412	373	353	381	503	656	1040	617
	592	527	462	280	305	269	321	261	384	565	719	474
Peyrusse Vielle	-	-	-	-	-	-	70 60	118 120	61 70	108 110	183 180	151 120
Donon	143	288	239	143	125	124	102	149	116	293	296	236
	140	280	180	110	70	60	100	90	100	210	220	210

					IYLBEN			4110	055	0.07	NOV	550
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	13	17	10	4	6	-	-	-	-	-	-	-
	13	18	8	3	6	-	-	-	-	-	-	-
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
Birkenes	40	22	41	22	17	16	11	10	13	22	25	51
	37	22	41	24	16	16	3	7	3	20	20	35
Utö	-	-	-	-	-	-	-	-	-	-	-	-
Waldhof	41	15	20	13	13	7	27	26	27	40	62	30
	25	3	13	3	3	3	16	26	24	36	44	25
Schmücke	58	31	19	14	15	15	18	15	30	33	42	43
	62	20	19	14	16	10	14	11	17	31	35	46
Košetice	54	28	27	25	15	15	14	17	21	15	34	26
	51	29	27	23	12	16	10	14	20	14	34	25
Tänikon	234	117	110	58	62	59	58	60	78	120	172	102
	107	89	96	57	53	47	58	48	63	110	127	80
Peyrusse Vielle	-	-	-	-	-	-	10 10	14 10	10 10	23 20	33 25	27 20
Donon	25	53	47	26	26	22	23	24	28	54	49	40
	25	50	30	30	20	10	20	10	20	40	40	40
				M-	+P-XYL	ENE						
Zeppelin	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
	22	26	13	6	8	-	-	-	-	-	-	-
	21	26	12	6	7	-	-	-	-	-	-	-
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
Birkenes	93	42	82	37	33	40	16	24	27	42	51	154
	91	29	87	39	26	46	3	14	3	47	44	84
Utö	-	-	-	-	-	-	-	-	-	-	-	-
Waldhof	120 93	41 3	48 42	14 3	9	10 3	53 33	45 42	36 17	62 57	99 55	54 44
Schmücke	146	76	33	18	17	20	37	26	47	51	64	74
	171	55	25	16	14	14	38	16	27	49	45	66
Košetice	103	61	47	40	27	30	28	30	37	38	68	54
	92	70	41	43	22	27	19	18	35	33	52	51
Tänikon	-	-	-	-	-	-	-	-	-	-	-	-
Peyrusse Vielle	-	-	-	-	-	-	25 25	32 30	20 20	45 25	64 45	57 40

O-XYLENE													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
Zeppelin	10	13	11	4	5	-	-	-	-	-	-	-	
	9	13	11	3	6	-	-	-	-	-	-	-	
Pallas	-	-	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
Birkenes	36	19	33	28	14	17	7	13	11	13	18	60	
	32	12	37	25	13	14	3	4	3	11	14	38	
Utö	-	-	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
Waldhof	31	14	3	3	3	3	18	3	11	30	35	16	
	10	3	3	3	3	3	3	3	3	27	3	3	
Schmücke	46	16	3	3	3	13	9	3	18	16	16	22	
	52	3	3	3	3	3	3	3	3	16	3	3	
Košetice	44	21	23	18	14	13	13	13	16	17	26	24	
	33	19	21	17	9	13	10	9	16	15	21	22	
Tänikon	224	102	83	40	51	50	46	48	49	100	161	99	
	96	73	65	40	43	33	43	31	41	86	117	74	
Peyrusse Vielle	-	-	-	-	-	-	12	16	14	23	34	29	
	-	-	-	-	-	-	10	20	10	20	25	20	
Donon	19	45	37	21	25	19	20	26	26	53	49	42	
	20	40	30	20	20	10	20	10	20	35	40	40	

Monthly mean and median concentrations (first and second line, respectively) of carbonyls (ng/m³) in 1999

				FOF	RMALDE	HYDE						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	284	275	320	394	508	644	1134	973	830	409	407	270
	270	265	340	400	540	550	860	835	805	360	430	240
Waldhof	_	580	625	699	834	816	2158	2051	1906	815	776	538
	-	390	475	650	875	805	1980	1860	1740	685	710	460
Košetice	868	885	999	880	979	1056	1661	1751	1801	1333	1050	1086
	660	750	960	720	960	1035	1660	1630	1760	1270	1040	720
Donon	738	658	1541	1040	1891	1807	1839	1273	1338	1001	729	698
	690	551	1006	866	1440	2040	2189	1093	958	1019	574	746
				ACE	ETALDE	HYDE						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	-	448	548	509	670	673	1659	1146	868	563	494	500
	-	395	600	500	620	670	1170	890	815	580	440	510
Waldhof	-	755	828	863	841	759	1000	1057	1054	691	814	652
	-	530	680	690	850	770	1020	900	1280	500	860	600
Košetice	1084	957	1116	1127	1538	1331	1358	1492	1509	1019	993	1019
	760	810	1110	1030	1410	1380	1270	1550	1460	1030	1100	750
Donon	535	589	734	661	783	831	717	520	555	660	509	436
	491	571	783	706	905	668	682	455	614	769	466	465
					ACETO	NE						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	764	-	1594	2639	4214	3269	5256	5896	4297	7550	5662	2130
	770	-	1730	2800	3700	2850	4190	5770	3685	8490	3720	1540
Waldhof	-	-	2893	3480	6014	3377	3691	3301	3341	2305	2364	2484
	-	-	2775	3465	4830	3115	3380	3180	2970	2225	1780	2610
Košetice	1764	1510	2512	2527	3189	2914	2904	3208	4433	2446	3076	3818
	1300	1570	2510	2200	2980	2700	3120	2550	4890	2190	2730	2610
Donon	1302	1396	2888	2706	4005	3867	3414	3295	3647	2269	1782	1157
	1084	1356	2536	2072	3534	3830	3158	2584	3298	2468	1568	1116
				F	PROPA	NAL						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	54	73	76	74	118	99	208	88	90	74	34	43
	50	75	70	70	120	100	130	70	65	70	10	40
Waldhof	-	128	127	129	114	98	148	213	184	138	128	82
	-	90	110	130	120	100	150	180	220	110	130	80
					050	000	047	070	007	040	404	147
Košetice	122	143	199	208	350	283	317	372	367	246	161	171
Košetice	122 120	143 125	199 190	208 180	330	283	280	390	36 <i>7</i> 440	195	161	100
Košetice Donon												

				Е	BUTANC	NE						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	-	232	295	269	268	338	409	369	320	136	276	212
	-	230	245	260	290	345	310	290	295	180	230	240
Waldhof	_	390	543	576	590	511	304	596	725	471	491	498
	-	260	425	490	590	545	340	610	780	430	465	440
Košetice	410	505	831	876	848	706	650	847	979	604	557	514
	330	485	850	720	750	665	620	850	850	620	520	430
Donon	343	438	538	601	915	1105	1120	811	959	570	376	323
	271	424	632	555	819	951	1121	633	712	507	367	325
					BUTAN	AL						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	47	77	80	66	100	74	126	61	47	36	20	39
	50	75	80	60	90	70	90	55	45	20	20	40
Waldhof	_	85	85	79	64	60	68	97	78	76	79	52
	-	70	80	85	65	60	60	80	100	70	80	50
Košetice	96	115	128	126	198	164	158	183	206	109	73	68
	90	115	120	110	170	160	150	130	240	100	70	60
Donon	57	71	98	63	94	78	77	64	61	64	56	48
	55	57	80	57	90	63	84	58	47	42	53	44
					PENTAN	ΙΔΙ						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	57	58	116	97	182	233	198	90	98	69	30	29
	60	50	100	100	180	260	240	70	85	40	20	20
Waldhof	_	113	233	304	314	346	47	31	22	22	20	19
	-	130	240	300	295	340	30	20	20	20	20	20
Košetice	78	99	156	144	141	129	77	109	116	84	22	24
	90	95	150	130	120	110	50	60	100	50	20	20
Donon	_	_	_	_	_	_	_	_	_	_	_	_
	-	-	-	-	-	-	-	-	-	-	-	-
					GLYOX	ΔΙ						
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Birkenes	15	15	50	31	76	44	20	42	15	31	18	15
	15	15	50	30	70	30	15	15	15	30	15	15
Waldhof	_	15	38	80	46	54	236	191	134	31	24	15
	-	15	35	30	40	45	160	15	140	25	15	15
Košetice	35	67	63	62	44	40	66	48	56	49	66	71
	15	55	30	50	30	35	60	40	50	40	40	15
Donon	40	49	81	80	82	83	77	31	36	23	35	24
	30	30	71	34	57	67	74	23	24	23	24	26

## Appendix B

# Time series of VOCs measured in 1999 and reported to EMEP-CCC

