EMEP/CCC-Report 8/2002

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

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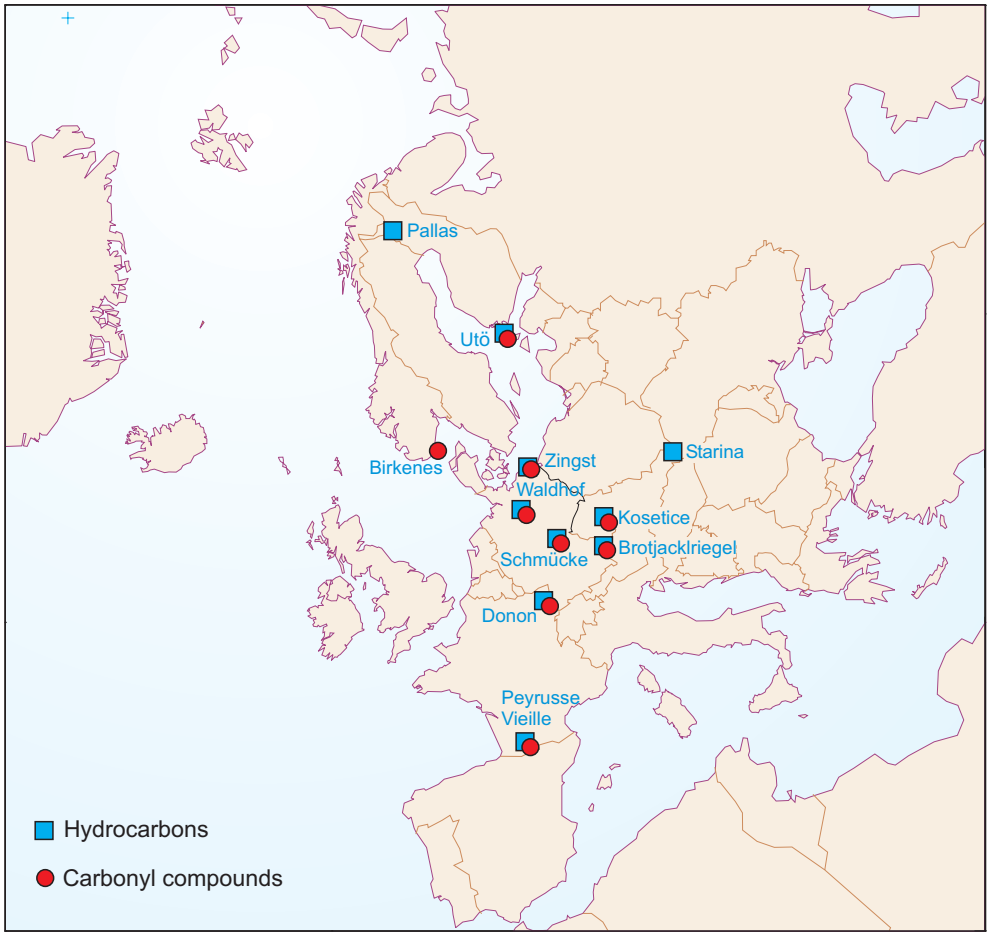

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VOC measurements 2000

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Summary

This report presents measurements of VOC carried out during 2000 at EMEP monitoring sites. The first carbonyl analyses made by UBA at four German sites are included in the report. Furthermore, carbonyl sampling was also initiated at Utö in Finland in 2000, and the first measurement data are presented in this report.

In 2001 CCC visited UBA's laboratories in Germany and the results and conclusions of the discussions and comparisons is summarized in this report. The visit was a very fruitful and efficient way of identifying and solving technical problems related to the sampling and chemical analyses and will be continued with visits to other laboratories participating in the EMEP VOC monitoring activity.

The parallel analyses of hydrocarbons by UBA and NILU at Waldhof indicate that both institutes are in general within the data quality objectives (DQO) limits for most of the data although some outliers are seen. For the carbonyls UBA's data should be regarded as preliminary and only formaldehyde, acetaldehyde and acetone are reported. The comparison with NILU's parallel data for these compounds indicate an overall good agreement between the two laboratories' data although UBA's values were systematically higher for formaldehyde and acetaldehyde.

For the small VOC network as a whole, monthly statistics show that in general the concentration level and seasonal variation in 2000 was of the same order as in the year before. For several of the components, the mean concentrations in the first months of the year were lower than in 1999, indicating a winter with more efficient mixing. The December concentrations were, however, fairly high at many of the continental sites, indicating a month with more stagnant weather pattern. The concentration level of several compounds was particularly high at Starina during several months, and the concentrations of acetone at Birkenes in summer were also substantially higher than measured previously.

In general the measurements indicate that hydrocarbons become fairly well mixed in Europe in winter. Components indicative of natural gas emissions, ethane and propane, were higher in north and east, whereas e.g. ethene, propene and acetylene were higher in central and eastern parts of the continent. N- and i-butane that stem from a number of different emission sources also showed high concentrations to the north.

A simple trend evaluation of the hydrocarbons was carried out. Whereas the Finnish data do not indicate any clear concentration trends during 1993-2000, the data from Germany and the Czech Republic clearly suggest a decline in the hydrocarbons. The drop in the median concentrations during these 7 years is of the order of 20-50% with the largest downward tendency for benzene and the C_4 and C_5 compounds and the smallest change for propane. These changes correspond well with the changes in daily measured NO_2 concentrations during winter at the same sites, indicating that the emission reductions in the

hydrocarbons during 1993-2000 have been of the same order as the emission reductions for NO_x.

The source regions for the peak episodes of individual hydrocarbons at the different sites were mapped by use of 3-dimensional back trajectories. This indicated that each station had it's own characteristic source regions, differing considerably from one station to another and stressing the importance of a station network of a minimum number of sites. This method didn't segregate very well between the source regions for the individual compounds although some differences were seen for the two compounds propane and acetylene.

VOC measurements 2000

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.

In 1999 the so-called Gothenburg protocol, the Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, was adopted by the Executive Body of UN-ECE. The Protocol sets emission ceilings for 2010 for four pollutants: sulphur, NO_x, VOCs and ammonia. These ceilings were negotiated on the basis of scientific assessments of pollution effects and abatement options. Parties whose emissions have a more severe environmental or health impact and whose emissions are relatively cheap to reduce will have to make the biggest cuts. Once the Protocol is fully implemented, Europe's sulphur emissions should be cut by at least 63%, its NO_x emissions by 41%, its VOC emissions by 40% and its ammonia emissions by 17% compared to 1990. The Protocol also sets tight limit values for specific emission sources (e.g. combustion plant, electricity production, dry cleaning, cars and lorries) and requires best available techniques to be used to keep emissions down. VOC emissions from such products as paints or aerosols will also have to be cut.

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 (LV10), Košetice (CS03), Waldhof (Langenbrügge) (DE02), Tänikon (CH32) and Donon (FR08). 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 ±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 ±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. Previous results from the EMEP VOC programme have been presented in annual reports (e.g. Solberg et al., 2001). An EMEP expert meeting on VOC measurements was organised in Berlin, 1994 (EMEP/CCC, 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 (Lindskog et al., 1995; Solberg et al., 1996; Hov et al., 1997; Solberg et al., 2001).

2. Status of the measurement programme in 2000

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 are given in Table 2.

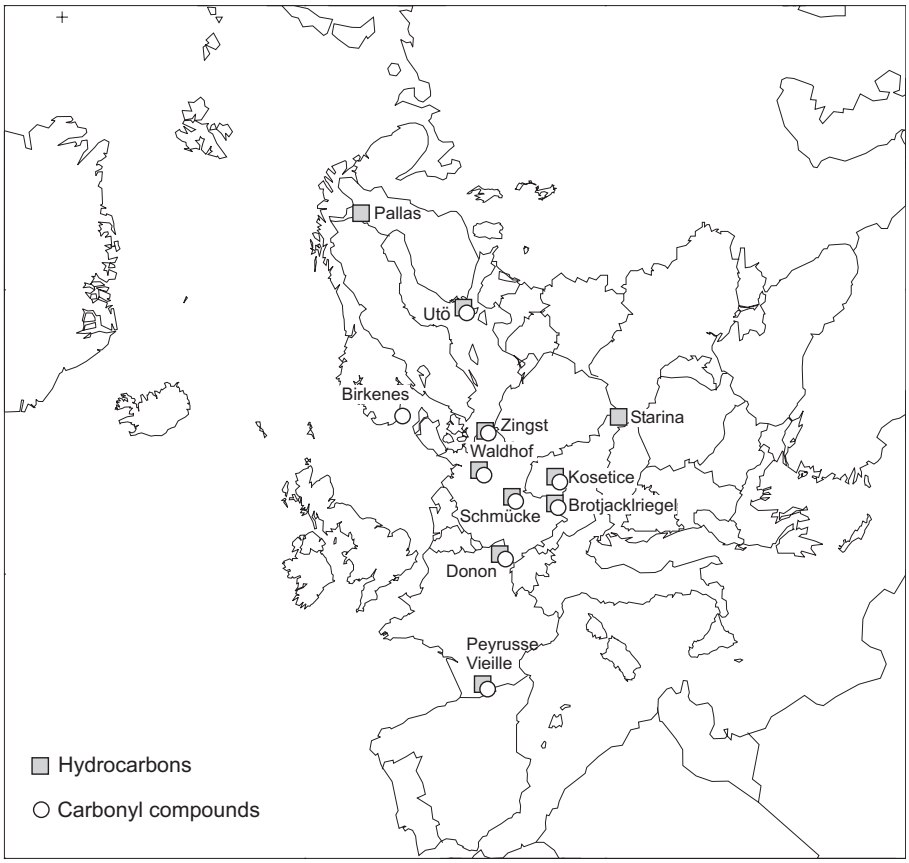


Figure 1: Monitoring sites for VOC in 2000.

As indicated by Table 2, data for 11 measurement sites for VOC have been reported to CCC and 9 of these with carbonyls. FMI started carbonyl sampling at Utö in 2000 and the samples are analysed by NILU. UBA started VOC measurements including both hydrocarbons and carbonyls at DE09 Zingst and DE05 Brotjacklriegel in January 2000. The carbonyl data analysed and reported by UBA should be regarded as preliminary, and only formaldehyde, acetaldehyde and acetone are reported.

Table 2: Status of the VOC monitoring programme in 2000. 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[1)]	Lab.[2)]	Carb[1)]	Lab.[2)]	Comments
Pallas	FI96	Reg.	FMI	n.m.	-	
Utö	FI09	Reg.	FMI	Reg.	NILU	Carbonyl sampling started in May 2000
Birkenes	NO01	n.r.	NILU	Reg.	NILU	Technical lab. problems with analyses during spring
Waldhof	DE02	Reg.	UBA	Reg.	NILU (UBA)	Problems with old ozone scrubber at last part of year (NILU's data)
Schmücke	DE08	Reg.	UBA	Reg.	UBA	Carbonyl monitoring started in 2000
Zingst	DE09	Reg.	UBA	Reg.	UBA	New VOC site
Brotjacklriegel	DE05	Reg.	UBA	Reg.	UBA	New VOC site
Košetice	CS03	Reg.	CHMI	Reg.	NILU	
Starina	SK06	Reg.	SHMI	n.m.	-	Problems with GC operation at the beginning of the 3rd quarter of the year. Some canisters analysed by CHMI.
Donon	FR08	Reg.	EMD	Reg.	EMD	
Peyrusse Vieille	FR13	Reg.	EMD	(Scat.)	EMD	Carbonyl monitoring started in March but no carbonyl data used due to station renovation

1) Reg. = regularly, Scat. = scattered, n.m. = not measured., n.r. = not reported
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)

Table 3 gives the number of valid samples of hydrocarbons and carbonyls (after inspection and removal of outliers). According to EMEP's recommendations, the samples should be taken twice a week, implying that 104 samples per year correspond to 100% data cover.

Table 3: The number of samples of hydrocarbons (HC) and carbonyls (Carb) in 2000.

Station	Number of samples	
	HC	Carb
Pallas	98	-
Utö	98	52[a)]
Birkenes	-	77
Zingst	87	104
Waldhof (NILU)	-	95
Waldhof (UBA)	87	104
Schmücke	87	91
Brotjacklriegel	84	104
Košetice	102	75
Starina	63	-
Peyrusse Vieille	95	-
Donon	98	98

a) Carbonyl measurements started in May at Utö

A 90% data completeness, i.e. 94 samples pr year, of daily values is given as data quality objective according to the EMEP manual (EMEP/CCC, 1996) and that is fulfilled at about half of the VOC sites. The number of carbonyl samples analysed by NILU was low due to technical and logistical problems affecting samples during March-June (varying between the stations). Furthermore, there were problems with the GC operation at the beginning of the third quarter of 2000 at Starina. In order not to loose all results from this period, the laboratory at CHMI analysed a few exposed canisters.

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). However, the number of sites with carbonyl measurements was higher in 2000 than previously due to the monitoring initiated at Utö and at Zingst and Brotjacklriegel.

2.2 Analytical procedures and quality control

The procedures for sampling and chemical analyses were similar in 2000 as in 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, 1996). 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). A detailed presentation of the sampling and analyses performed by the laboratories at EMD (France), CHMI (Czech Republic) and SHMI (Slovakia) has been given in previous annual reports and is not repeated here.

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, 1996) 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 concentrations of 3-buten-2-one, 2-methylpropenal, 2-butanone and butanal have for many years been difficult to interpret. No systematic and explainable pattern has been found and inter-laboratory comparisons between EMD, UBA and NILU have indicated analytical problems. Laboratory studies at CCC indicate that unsaturated carbonyl compounds are not chemical stable in the prepared sample solution. Ongoing studies will provide a validation of the EMEP-method performance against unsaturated carbonyl compounds. Furthermore, LC/MS studies indicate possibilities of chromatographic interference in the C_4 carbonyl compound range. Ongoing studies will provide a validation of the chromatographic performance of the EMEP-method.

2.3 Visit at Waldhof – Carbonyl measurements and QA-initiative

For several years two carbonyl samplers have been running in parallel at Waldhof in Germany. Both samplers have been used to expose 2,4-DNPH coated silica cartridges according to the EMEP-manual. One cartridge has been sent to CCC for chemical analysis and the other one has been analysed by UBA in Germany (Berlin).

In beginning of December 2001 CCC visited the Waldhof site and the UBA laboratory in Berlin. The intention by the visit was to discuss routines and protocols on the sampling site and the laboratory. As a part of the visit a selection of the measured carbonyl data from 2000-2001 was compared and evaluated. A comparison of the year 2000 measurements of formaldehyde, acetaldehyde and acetone can be in found elsewhere in this report.

The main conclusion is that the Waldhof site and the UBA laboratory perform carbonyl measurements at the same level as CCC. The agreement between the results from UBA's and CCC's laboratories is very satisfactory.

A number of points regarding the sampling and analyses were raised, though. Firstly, a problem with the ozone scrubber was discovered for the first and the last part of the 2000 samples. The problem causes a negative artefact and is most pronounced for formaldehyde and acetaldehyde. This can be seen when the ozone scrubbers are changed in the beginning of April and in the beginning of September. In the period between April and September the agreement between the two data sets is good. Before April and after September the negative artefact is seen for the CCC scrubbers. As a routine, at Waldhof the scrubbers are tested against ozone scrubbing effect before discarding, and the CCC scrubbers did still perform satisfactory in ozone scrubbing when the scrubbers were discarded. However, there are indications of altered performance as the scrubber tended to be active on carbonyl scrubbing due to systematic low carbonyl concentrations in the actual time. In the chromatograms of some samples a relatively large unknown peak appears which can be related to the performance of the ozone scrubber. These samples will be further explored with LC/MS in addition to a more extensive scrubber validation experiment.

Then, secondly, it was realized and agreed that the use of a so-called *ternary gradient* (in the chromatogram analyses) at UBA will make UBA's chromatograms and NILU's chromatograms more comparable. Furthermore, this will also lead to a better chromatographic resolution for C_4 carbonyl compounds. In

addition, better resolution between the reagent peak and the formaldehyde peak will be achieved.

Thirdly, it was agreed that parallel sampling should be performed as a part of the QA work during 2–4 campaigns (6–8 samples) each year to compare UBA's measurements with NILU's measurements.

2.4 C_2-C_9 hydrocarbons

In December 2001 CCC visited UBA's site Schmücke where UBA is performing measurements of C_2-C_9 hydrocarbons in canisters from five different sites (Schmücke, Zugspitze, Waldhof, Brotjacklriegel and Zingst). Every Thursday the instrument is used to do online measurements at Schmücke for a period of 24 hours.

During the audit the instrumentation for both canister sampling and canister cleaning was discussed. For the cleaning procedure a slight change for further cleaning of the purge gas was suggested and agreed on.

The routines for maintenance work like changing of drying tubes and CO_2 scrubber where discussed as well as calibration routines.

The synthetic standard gas is from NPL (UK) – concentrations of the individual hydrocarbons are between 1 and 8 ppb. 50 ml of the standard mixture is used for daily calibration – once a month a 500 ml aliquot is analysed. For on-line analysis or canister sampling 500 ml of air is analysed. Because of the fact that different volumes of standard and sample are analysed the precision and reproducibility of the volume measurements done by mass flow controllers are crucial.

The chromatograms of the instrument indicated one minor problem. Too much methane from the sample was pre-concentrated resulting in ethane being "rider peak" in the end of a big methane peak. This problem could be solved just by adjusting the temperatures for the pre-concentration step.

The results of parallel measurements between UBA and NILU were discussed. The results did agree very well.

The overall impression of the staff, the site, the instrumentation and the routines as well as the achieved results were very good.

3. Results from parallel analysis

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

Parallel sampling and analysis of hydrocarbons started in 1997 and was continued until June 1999. The 1998 data has been presented previously (Solberg, 1999). The 1999 data are discussed here. A total of 27 data points have been obtained for the period January to June 1999.

The evaluation of the comparison between the two institutes should be based on the data quality objective (DQO) as well as on the principal limitations of the sampling procedure and the analysis method in connection with the physio-chemical properties of the individual substance under consideration:

As the DQO for the hydrocarbon analysis has not been finally decided within EMEP, the DQO assumed here is ±25% for concentrations above 100 ppt and ±25 ppt for concentrations below 100 ppt. This choice takes into account that an overall fractional error does not make sense near the detection limit and is in agreement with expert recommendations.

Regarding analysis, the GC-method is specified for C_2 to C_5 hydrocarbons and is useful up to C_7, but fails for C_8 and higher. Therefore larger differences are expected for hydrocarbons with more than 7 C-atoms.

Furthermore, the individual hydrocarbons differ considerably in atmospheric concentration: Alkenes in general have much lower concentrations than alkanes or aromatics. Additionally, at least the higher alkenes are more reactive than alkanes and therefore are more susceptible to concentration changes in the sampling canister. Therefore, larger differences are expected for alkenes than for alkanes.

Regarding sampling procedure, in contrast to 1998, where the same sample was analysed by the two institutes, in 1999 two samples where taken, one of which was analysed at UBA, the other one at NILU. The samples where taken “almost” parallel, which means, that the sampling was started within about 5 minutes. If concentrations have a large variability with time, differences between the two samples are expected.

As can be seen from the scatter-plots (Figure 2) both institutes are in general in agreement within the DQO-limits for most of the data. Discrepancies occur where they are expected from the discussion above. Biases and deviations from a slope of 1, which has been observed in 1998 for some hydrocarbons, are not present in the 1999 data. The agreement found for butanes and pentanes can be attributed as nearly perfect. Although within the DQO-limits, larger scatter is observed for ethane, ethene, and propene. Some deviations are visible for the butenes, which may be attributed to the low concentrations, analytical reasons or to stability problems in the sampling canisters. Large discrepancies are expected for hydro-carbons with more than 7 C-atoms, as the method is not adapted here. This is actually the case as can be seen from the scatter plots for ethylbenzene and the xylenes. There are three obvious outliers in the data (7 January, 8 March, and 29 April). The reason for these are not clear although they could be explained by the peculiarities of the sampling procedure: High concentrations and composition of these samples could indicate an impact from “nearby “ sources (motor cars), which can not be avoided completely, even at so called “rural sites”. Under these circumstances, samples that are taken with a few minutes offset in time are expected to show considerable different concentrations. However, other explanations could not be ruled out.

In conclusion, the agreement between the hydrocarbon measurements of NILU and UBA is within the range that can reasonably be expected.

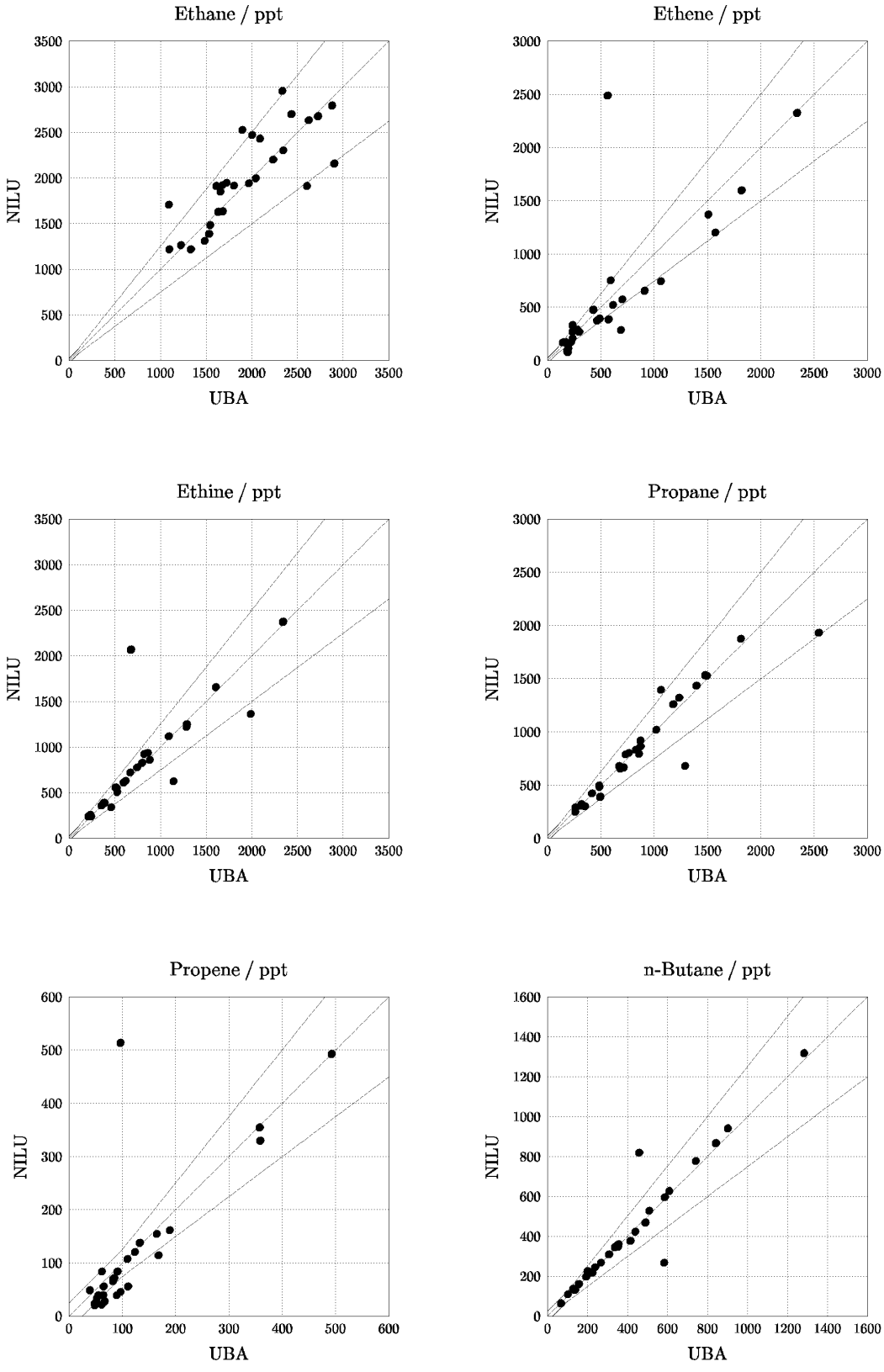
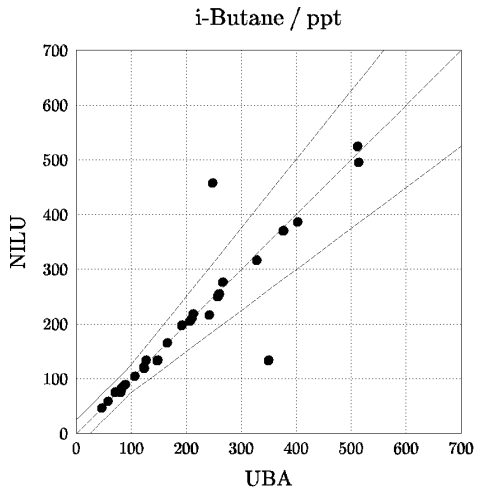
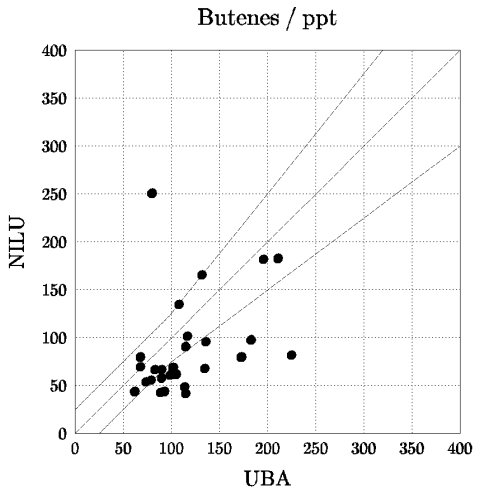
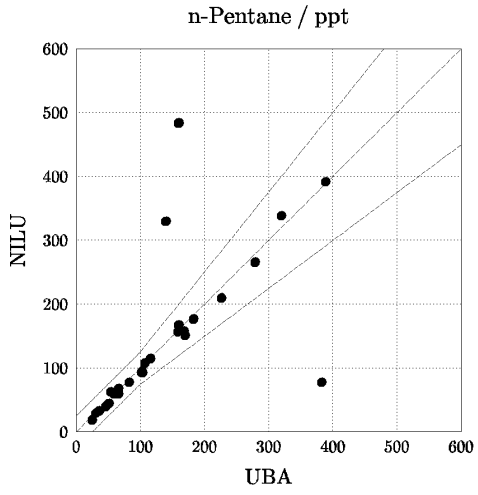
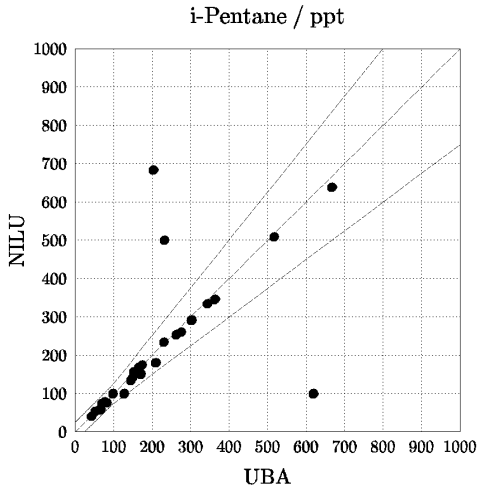


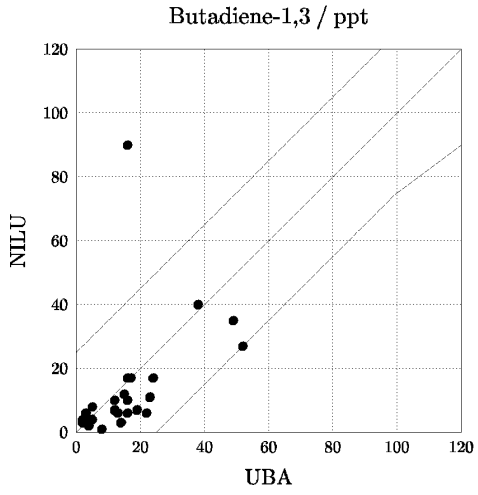
Figure 2: Scatter-plots of hydrocarbon parallel samples at Waldhof analysed by UBA and NILU. The slope-1-line and the DQO-limits (max (±25%, ±25 ppt)) are indicated by dashed lines.











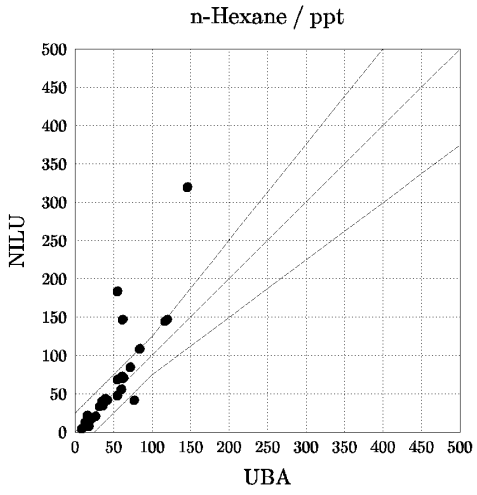


Figure 2 (contd.)

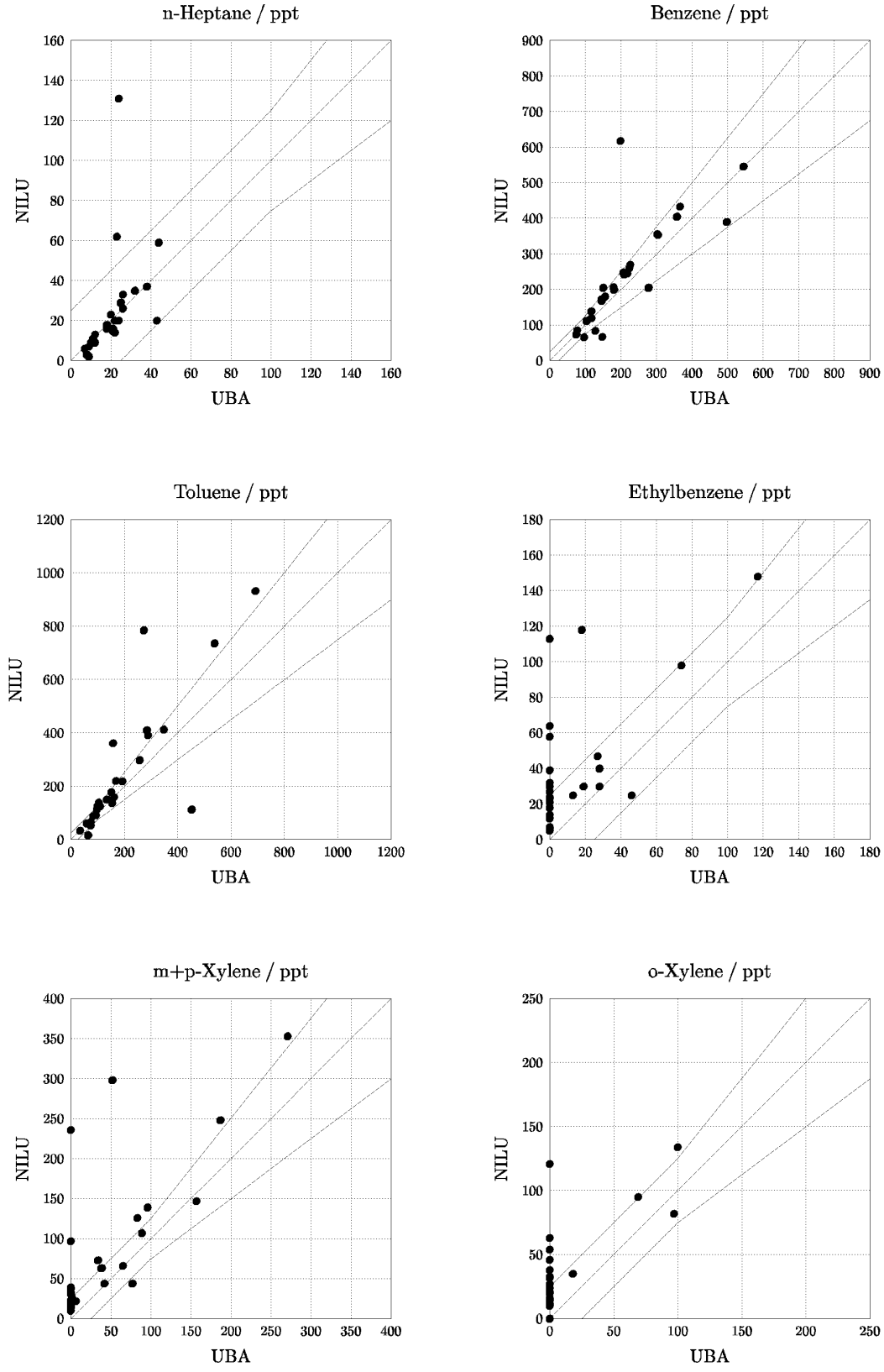


Figure 2 (contd.)

3.2 Parallel analysis of carbonyl compounds at Waldhof by NILU and UBA

Figure 3 shows the results of the parallel analysis of formaldehyde, acetaldehyde and acetone at Waldhof by NILU's and UBA's laboratories. A few results with extreme values of acetone from UBA's analyses during the first three months of 2000 were removed from the dataset. Also for Zingst and Schmücke a small number of acetone outlier values for the same time period were taken out of the data. A statistical summary of the parallel analysis is given in Table 4. The statistical parameters include the medians of the data from NILU and UBA and the median differences as well as the modified median absolute difference estimator, M.MAD, as described in the EMEP manual (EMEP/CCC, 1996) and the coefficient of variation, CoV, defined as CoV=(M.MAD)/(NILU's median). The analyses from the laboratory at NILU were regarded the reference in these calculations.

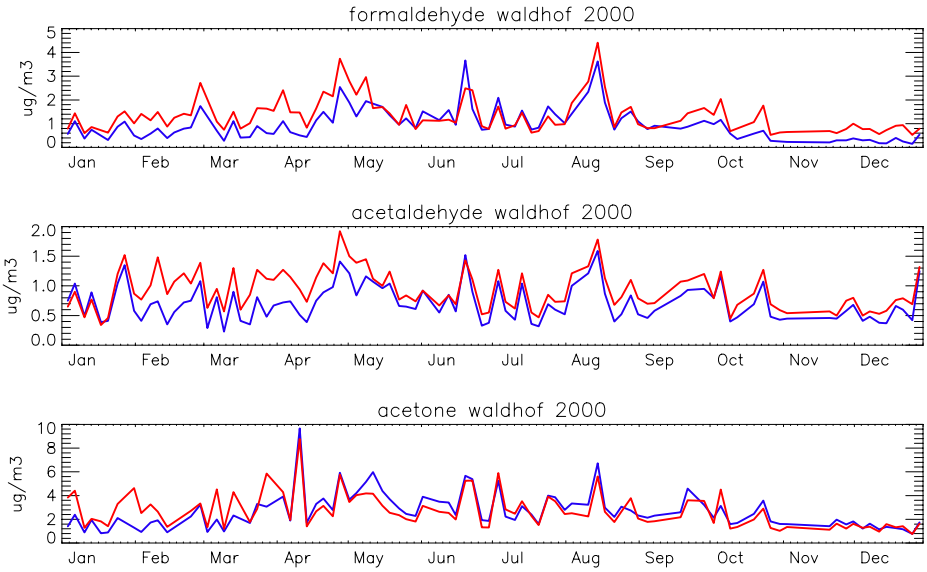


Figure 3: Results of parallel sampling and analyses of carbonyl compounds at Waldhof by NILU (blue line) and UBA (red line) in 2000.

M.MAD expresses the spread of the data and equals 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 that make them particularly useful for trace gas measurements that normally show a non-normal distribution in the data.

As indicated by Figure 3, there is an overall good agreement between the two laboratories' data although UBA's values are systematically higher for formaldehyde and acetaldehyde (Table 4). Furthermore, the results indicate that the agreement between the time series improved during the year. Whereas a marked bias is evident during February-May, the bias is clearly reduced after May. UBA started their carbonyl measurements in the last months of 1999 and was still in a learning and development phase by the beginning of 2000, and the 2000-data should thus be regarded as preliminary.

Table 4: Results from parallel sampling and analyses of carbonyl compounds at DE02, Waldhof during 2000. 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). A few outliers were removed from this analysis. Unit: μg/m³.

	median NILU	median UBA	median difference	M.MAD	CoV
formaldehyde	0.820	1.13	0.425	0.393	0.479
acetaldehyde	0.665	0.850	0.190	0.133	0.201
acetone	2.310	2.450	-0.240	0.504	0.218

4. VOC concentrations in 2000 and long-term trends

4.1 Regional distribution of hydrocarbons

Monthly mean and median concentrations of the individual hydrocarbons and carbonyls for 2000 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 2000 are given in Appendix B.

The tables with monthly data show that in general the concentration level and seasonal variation in 2000 was of the same order as in the previous year. For several of the components, the mean concentrations in the first months of the year were somewhat lower than in 1999, indicating a winter with more efficient mixing. The December concentrations were, however, fairly high at many of the continental sites, indicating a month with more stagnant weather pattern. The concentrations at Starina were particularly high during February-May for most compounds, and for the rest of the year for certain compounds as benzene, butanes, pentanes. Starina, at the eastern part of the continent, is located in a region frequently receiving air masses with anthropogenic pollution from regions to the west. The number of samples from Starina was, however, low due to the GC problems mentioned above, and it is a question whether this could have affected also the concentration level in the first part of the year.

The general concentration level of the carbonyl compounds was also of the same order as the year before. One exception, though, is acetone at Birkenes that showed much higher concentrations in summer 2000 than previously. The reason for this difference is not clear. The mean (and median) concentrations for June and July of more than 8 μg/m^3 acetone at Birkenes seems very high compared with the other stations and compared with previous years of measurements. One possible explanation is unknown local sources and efforts should be taken to investigate the reason for these extreme values.

Figure 4–Figure 13 shows maps with the stations' median concentrations of 10 light hydrocarbons for the winter months January, February, November and December in 2000 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.

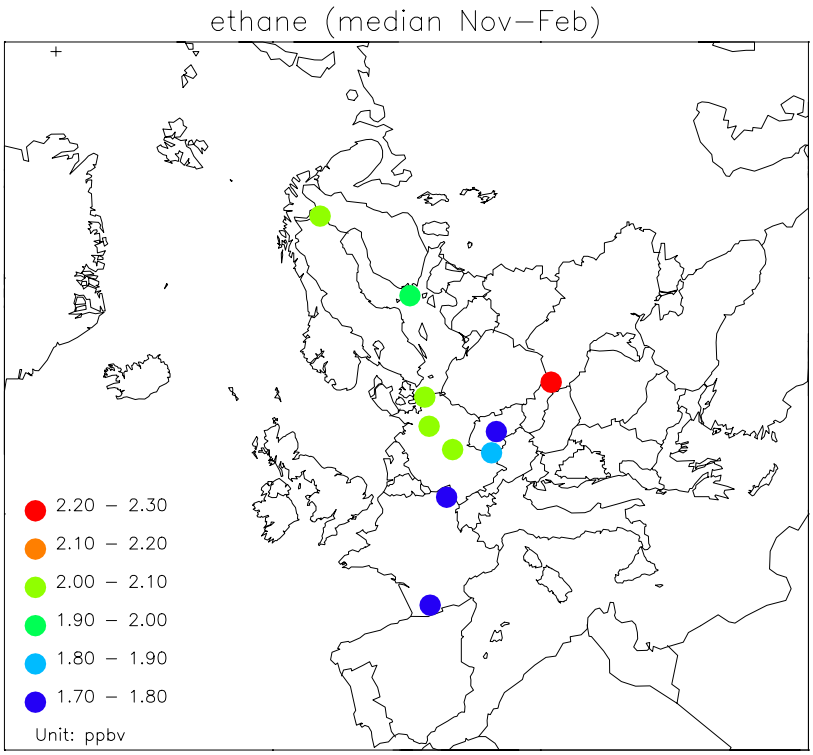


Figure 4: Median concentration of ethane at EMEP sites in the winter months November, December, January and February 2000 taken together.

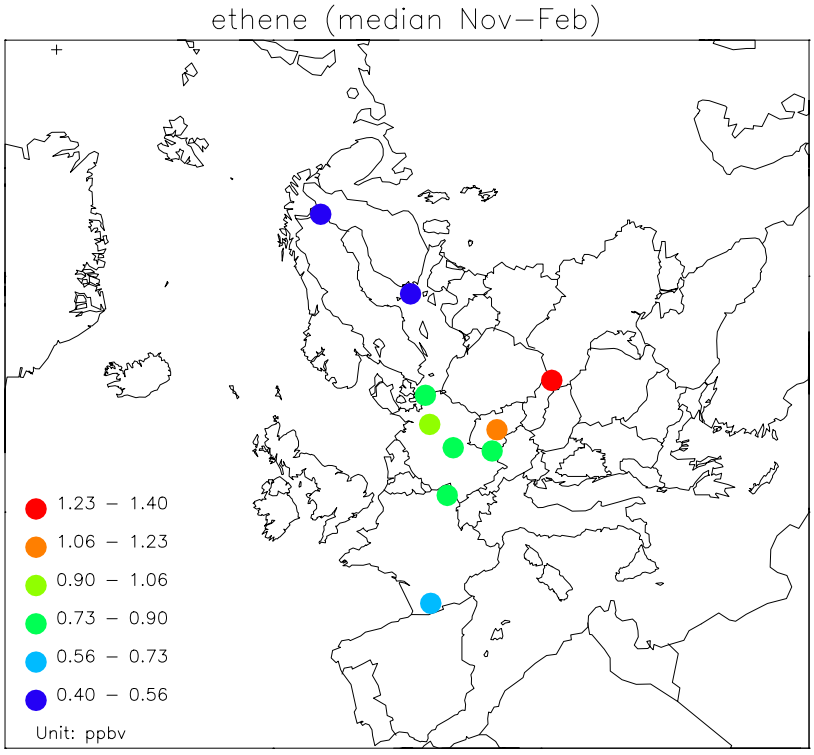


Figure 5: Median concentration of ethene at EMEP sites in the winter months November, December, January and February 2000 taken together.

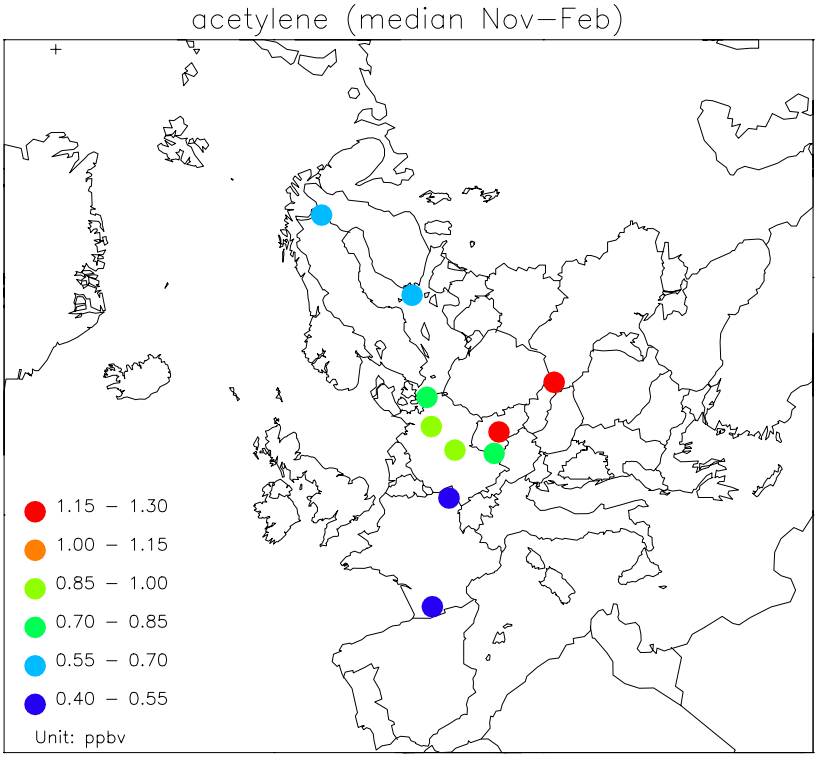


Figure 6: Median concentration of acetylene at EMEP sites in the winter months November, December, January and February 2000 taken together.

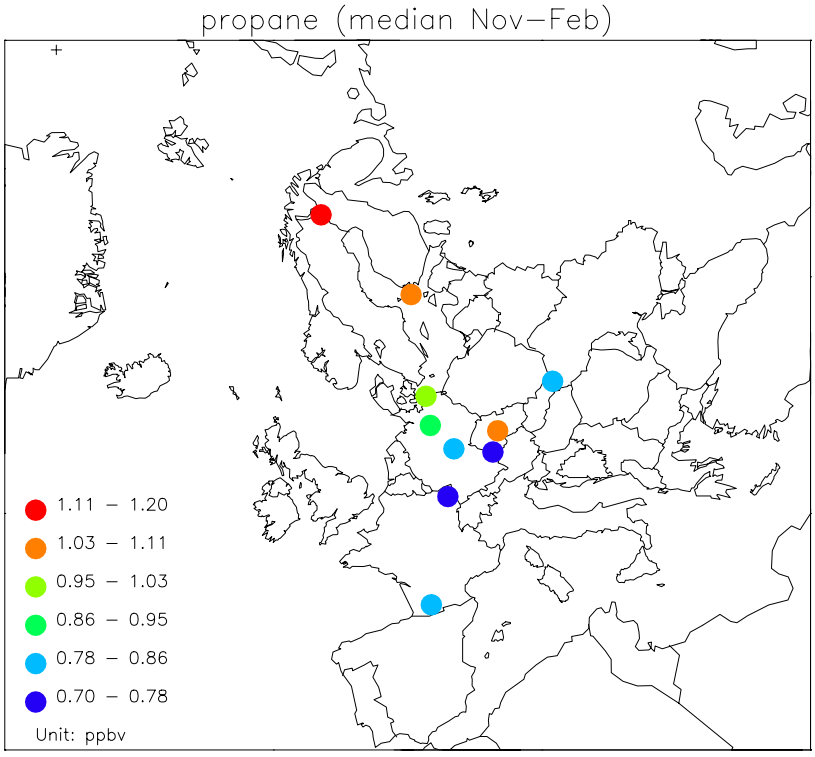


Figure 7: Median concentration of propane at EMEP sites in the winter months November, December, January and February 2000 taken together.

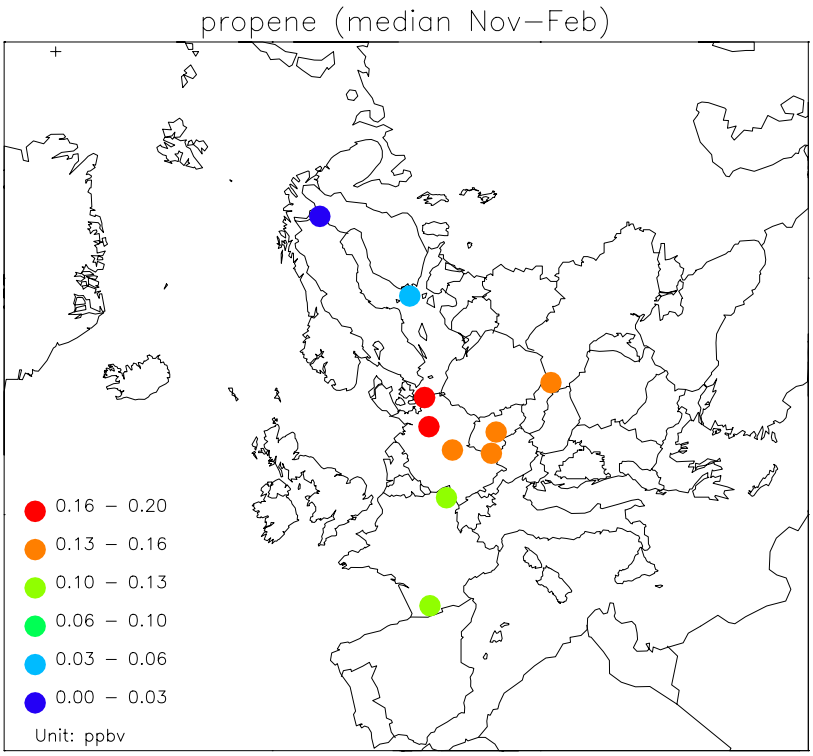


Figure 8: Median concentration of propene at EMEP sites in the winter months November, December, January and February 2000 taken together.

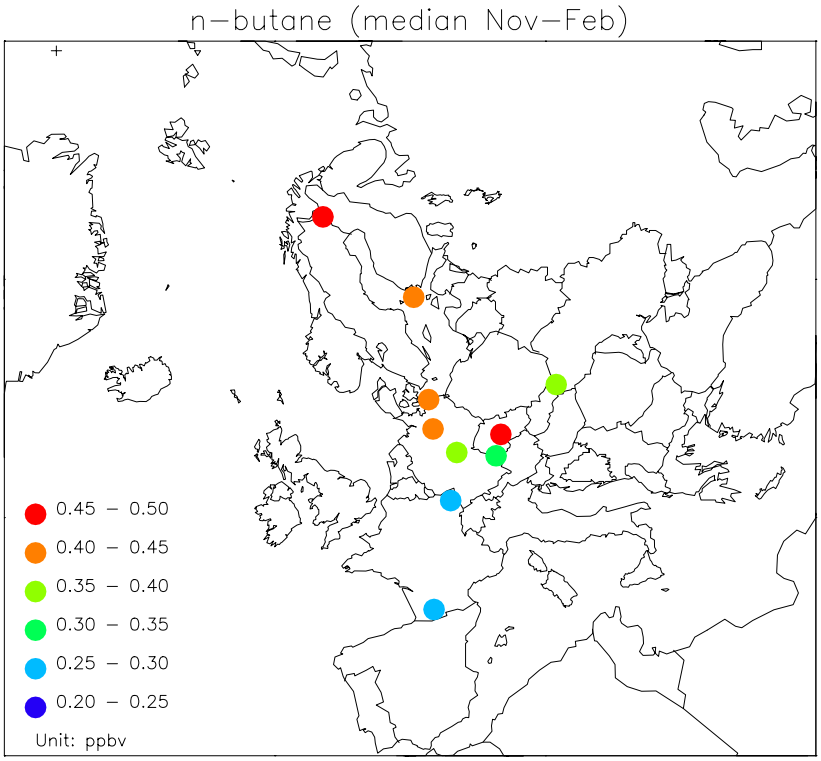


Figure 9: Median concentration of n-butane at EMEP sites in the winter months November, December, January and February 2000 taken together.

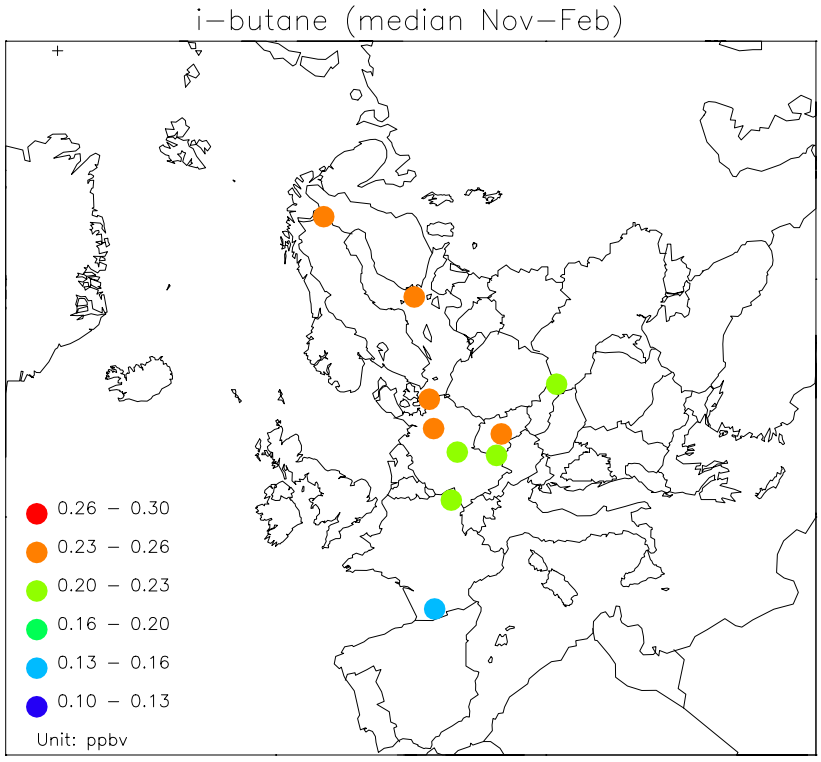


Figure 10: Median concentration of i-butane at EMEP sites in the winter months November, December, January and February 2000 taken together.

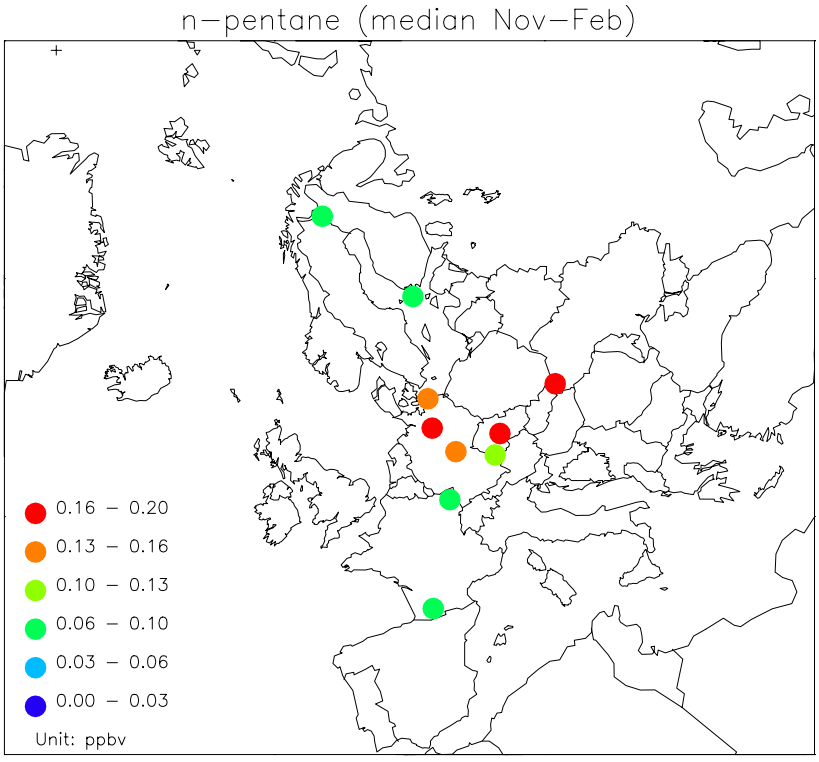


Figure 11: Median concentration of n-pentane at EMEP sites in the winter months November, December, January and February 2000 taken together.

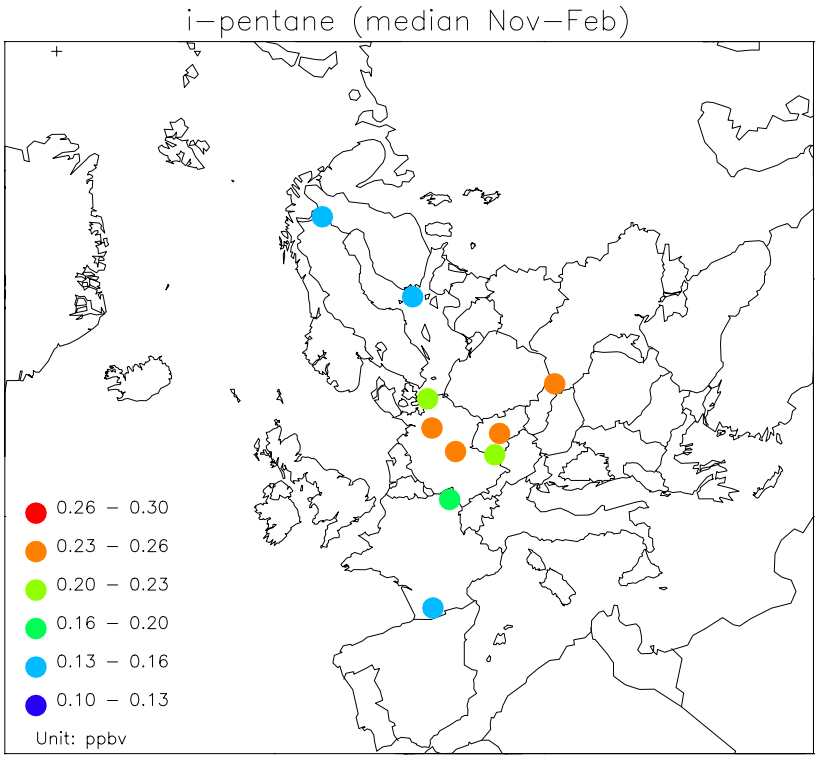


Figure 12: Median concentration of i-pentane at EMEP sites in the winter months November, December, January and February 2000 taken together.

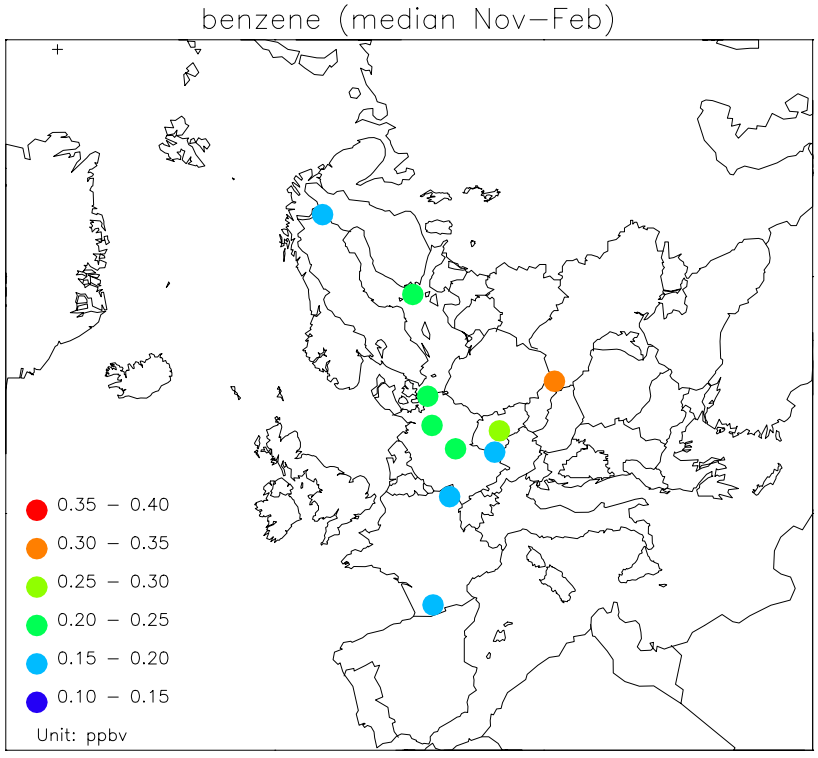


Figure 13: Median concentration of benzene at EMEP sites in the winter months November, December, January and February 2000 taken together.

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 higher in north and east, whereas e.g. ethene, propene and acetylene were higher in central and eastern parts of the continent. n- and i-butane that stems from a number of different emissions sources also show high concentrations to the north (Pallas).

4.2 Trends in VOC

Compared to other components measured in EMEP, long-term trends in VOC are particularly difficult to evaluate. This is due to the less frequent sampling – sampling twice a week – combined with a large natural spread in the measurement values, particularly for the hydrocarbons with a sampling time of 20 min, as well as the fact that VOC monitoring has a shorter history. The hydrocarbon monitoring in EMEP started in 1992 and for carbonyls in 1993. Thus, apparent trends in measured VOC will hardly be significant in a strictly statistical or meteorological perspective at this stage. Still, it is valuable to study how VOC concentrations change with time and how they compare with other components measured in EMEP.

Figure 14 and Figure 15 show the variations in the median concentrations of selected hydrocarbons for the winter months January-March from 1993 to 2000 for the Finnish sites (Pallas and Utö) and for Waldhof and Košetice, respectively. Figure 16 shows the similar results for the two continental sites for the summer months May-August. A least-squares fit to the average of the stations winter medians is included in the figures.

Firstly, these results show a clear difference between the Finnish sites on one hand and the two continental sites on the other. The Finnish data does not indicate any clear trends during these years. The same was true for the summer months (Finnish data not plotted). The data from the two continental sites clearly suggest a decline of several of the selected compounds in both seasons. The drop in the median concentrations during these 7 years is of the order of 20-50% with the largest downward tendency for benzene and the C_4 and C_5 compounds and the smallest change for propane. These values are of the same range or lower (i.e. smaller changes) than found for a number of urban networks (Roemer et al., 2001) and compare fairly well with the reported national emission reductions of VOC.

That the results differ from the northeast corner of Europe to the central part of the continent reflect the proximity to the main emission areas. The two sites in Finland will mostly be influenced by cleaner background air masses and only occasionally exposed to pollution episodes with transport from the continent. Waldhof and Košetice on the other hand, are both located in receptor regions from major emission regions. Filtering the measurement data by e.g. trajectories could in principle be used to segregate between the different air masses, however, with the sampling frequency of two samples pr week, the number of data is too small to carry out such an analysis for the trend evaluation.

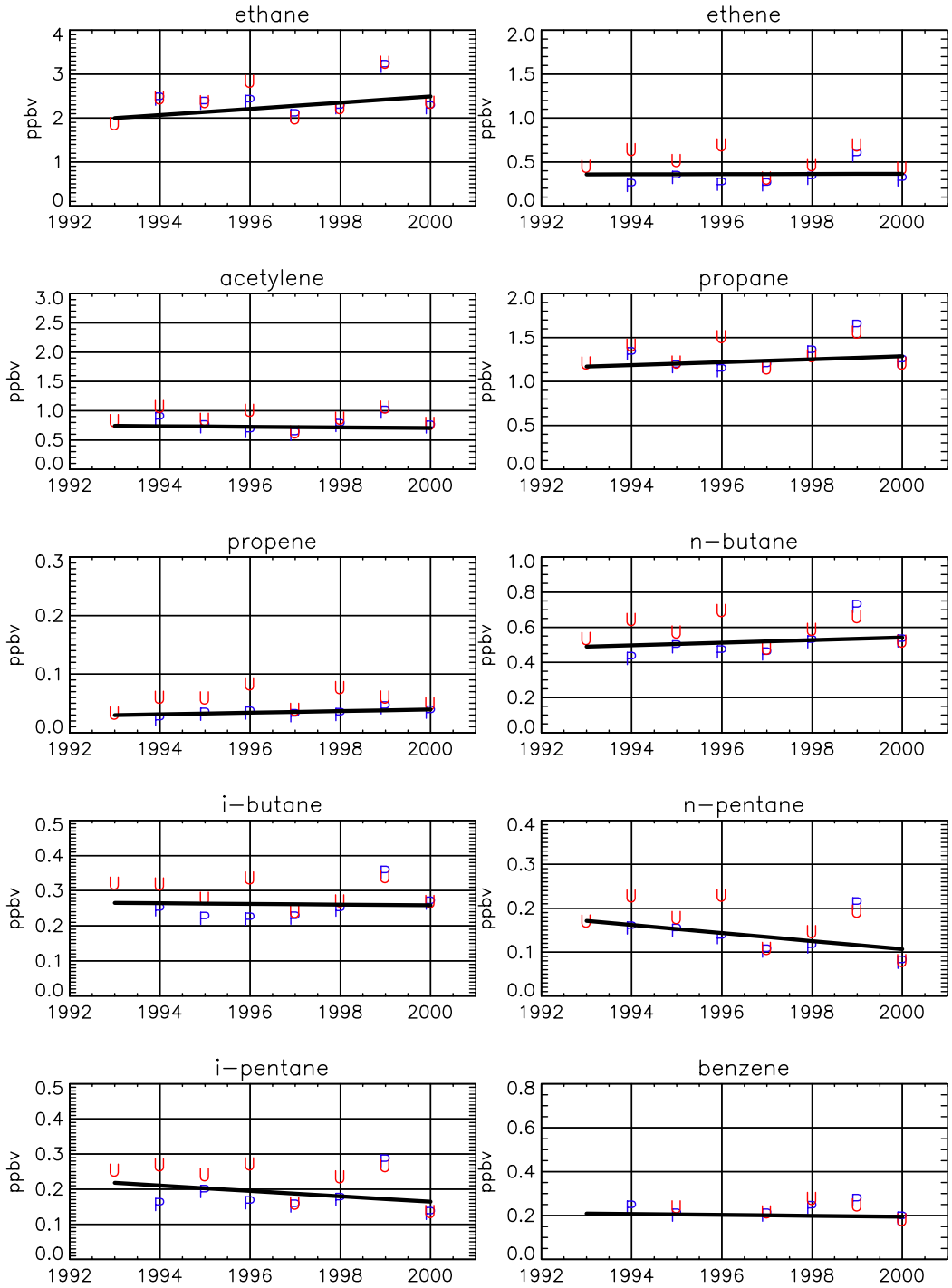


Figure 14: Winter medians (Jan-March) of selected hydrocarbons at Utö and Pallas in Finland from 1993-2000. A linear least-squares fit to the average of the winter medians is given by the solid line. Unit: ppbv.

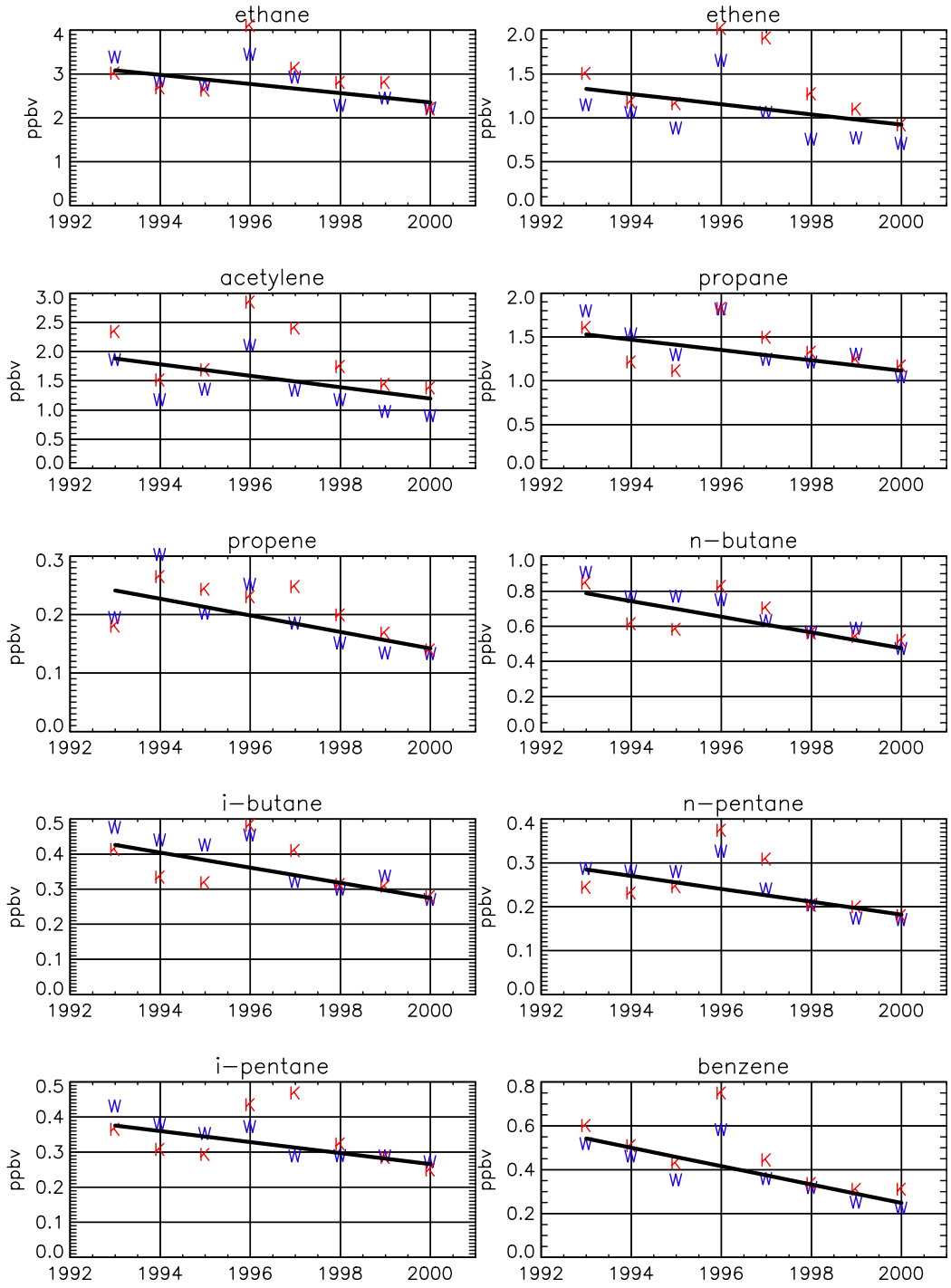


Figure 15: Winter medians (Jan-March) of selected hydrocarbons at Košetice and Waldhof from 1993-2000. A linear least-squares fit to the average of the winter medians is given by the solid line. Unit: ppbv.

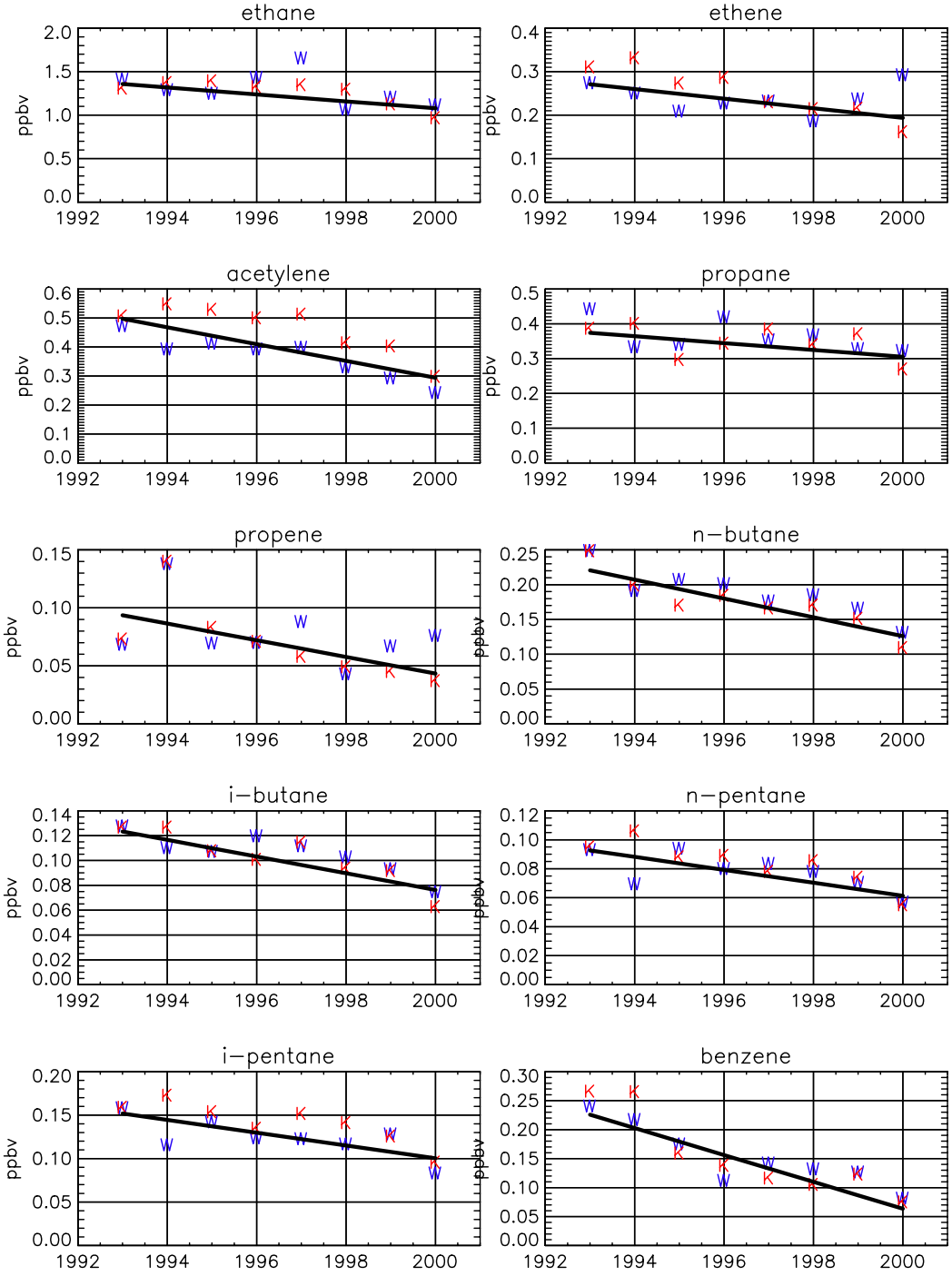


Figure 16: Summer medians (May-August) of selected hydrocarbons at Košetice and Waldhof from 1993-2000. A linear least-squares fit to the average of the summer medians is given by the solid line. Unit: ppbv.

For carbonyls, the number of monitoring sites is even less than for the hydrocarbons, and the time period with monitoring is also shorter. Additionally, analyses from spring and partly the summer in 2000 were unfortunately lost for several of the sites as mentioned above. Figure 17 shows the summer (June-August) medians of formaldehyde, acetaldehyde and acetone at Birkenes and Waldhof (NILU's analyses) during the period 1993-2000. Only the last part of

June is included in the Birkenes data for 2000. The data indicate a large variation from year to year and no clear trend for formaldehyde and acetone, although the acetone values at Birkenes for 2000 were much higher than the previous years. For acetaldehyde, however, the summer medians indicate a general increase, particularly at Birkenes. There is no obvious reason to expect an increase in acetaldehyde. The indications of a general decrease in hydrocarbon concentrations and indications of reduced peak ozone concentrations reported elsewhere should rather suggest a drop in acetaldehyde as well. Whether this trend is simply a result of infrequent sampling, or reflecting meteorological variations from one year to another or caused by other effects are at the moment an open question.

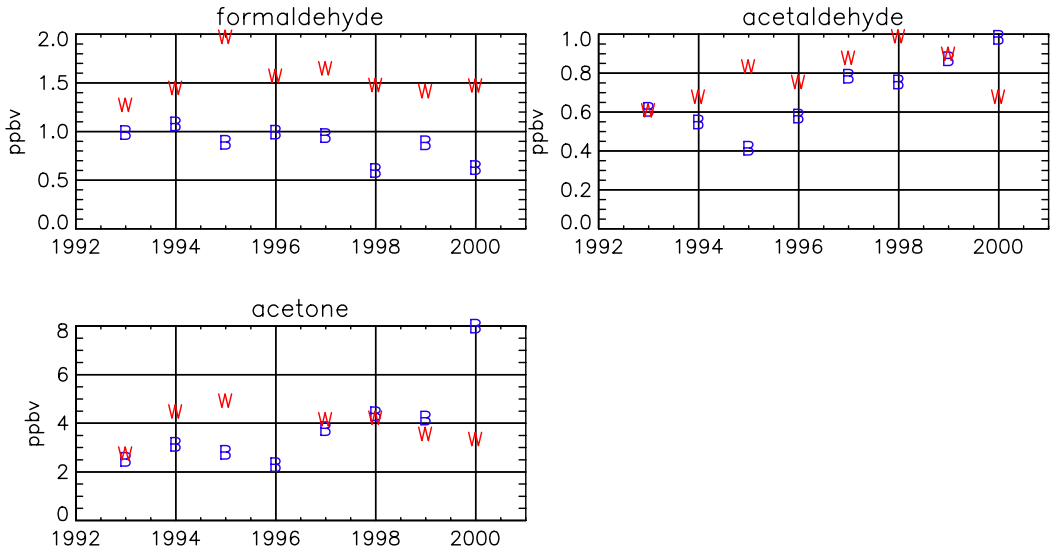


Figure 17: Summer medians (June-August) of selected carbonyl compounds at Birkenes and Waldhof from 1993-2000. Unit: μg/m^3.

There is, as noted, a large uncertainty inherent in long-term concentration trends estimated in the simple analyses above. Furthermore, the meteorological variations from year to year implies that these estimates shouldn't be regarded representative for the long-term trends in emissions, although at urban sites the atmospheric concentration data may correlate well with the emission data (Roemer et al., 2001). To partly circumvent this, the trend in measured NO_2 concentrations calculated in the same way and using only the days with hydrocarbon sampling was used as a reference.

Figure 18 shows the results when the individual hydrocarbons are plotted together with NO_2 and when all medians are scaled relative to the 1993 medians. These results suggest a fairly good agreement between NO_2 and several of the hydrocarbons, particularly for n-butane and i-pentane. For the hydrocarbons the winter of 1996 was obviously a peak year that is not reflected in the NO_2 median. Apart from this year, the agreement in the medians between NO_2 and the hydrocarbons is very good also for ethene and ethane. For toluene the results indicate a close agreement with NO_2, and a stronger decline in toluene than in NO_2.

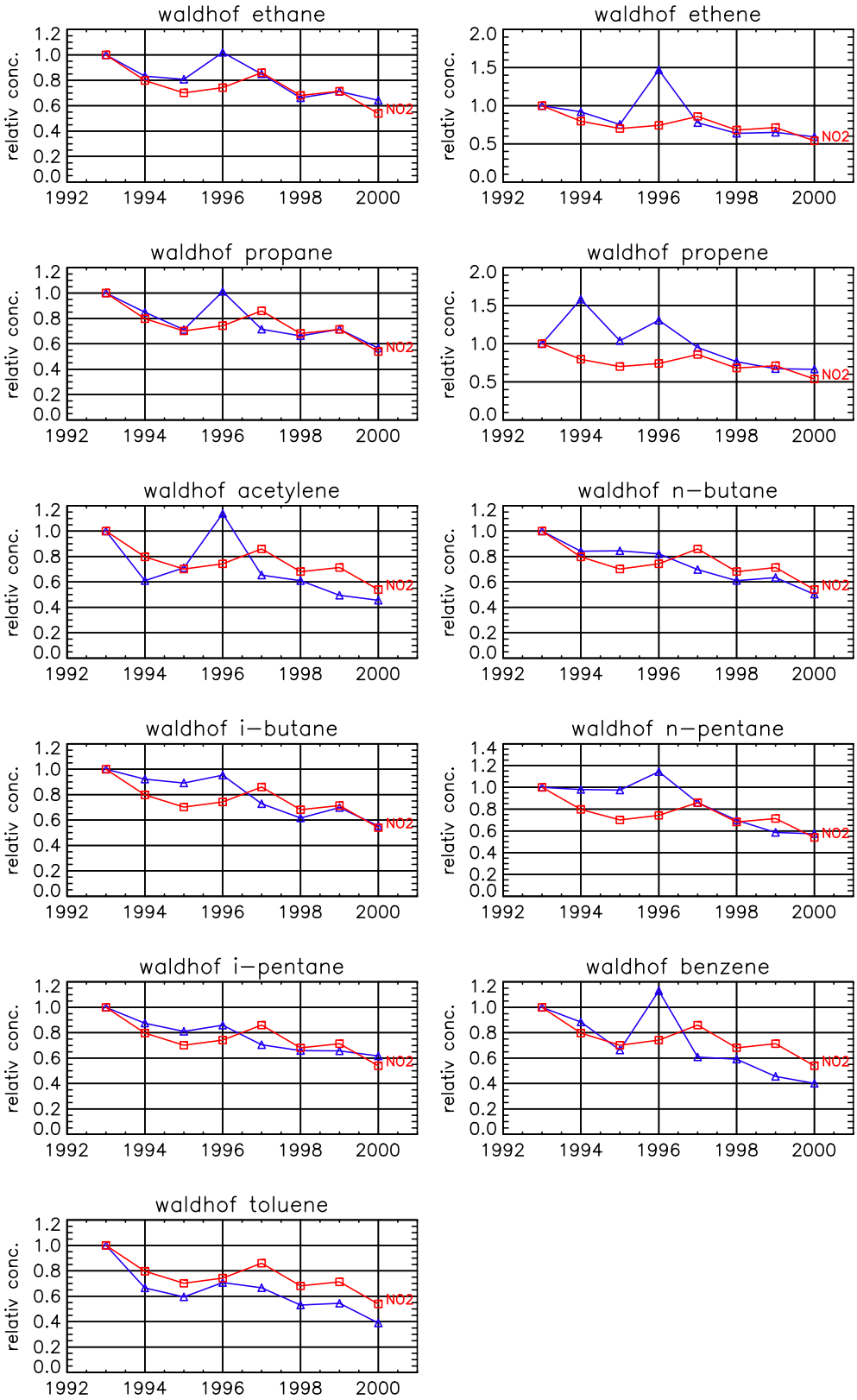


Figure 18: Winter medians (Jan-March) of selected hydrocarbons (blue) at Waldhof from 1993-2000 together with the accompanying winter medians of NO_2 (red) at the same site based on 24h samples from the days with hydrocarbon sampling only. All data are given relative to the medians in 1993.

In our view, these results indicate that the emission reductions in the given hydrocarbons during the 7-year's period 1993-2000 have been of the same order as the emission reductions for NO_x. For benzene and toluene the results suggest an even higher emission reduction than for NO_x. These are potentially important findings, as the VOC emission data are generally believed to be more uncertain than the data on NO_x emissions. Furthermore, there have been few attempts to verify the reported trends in VOC emissions based on rural VOC monitoring data.

These results and statements should be considered with substantial care for several reasons. Firstly, the findings for Waldhof are strictly speaking only representative of the main source areas for this site located in Northern Germany. The correlation between NO_2 and hydrocarbons at Košetice was much less evident than for Waldhof. Secondly, the agreement with seasonal medians of NO_2 and hydrocarbons was much less evident for other periods of the year than these three winter months. Partly, though, this is as expected, as atmospheric oxidation processes may mask these results when using periods outside winter.

In the discussion above the change in responsible laboratories has not been mentioned. Whereas the Finnish data have all been analysed by the national laboratory at FMI, the data from 1997 and onwards at Košetice was analysed by CHMI in the Czech Republic and from 1999 and onwards at Waldhof from UBA's laboratory in Germany. In the first years the samples were analysed by CCC's laboratory at NILU at both these sites. Biases in the laboratories' analyses may then affect the trends shown. No corrections were made to take this into account, as we think this is not straightforward to correct for. For the main components included in this trend analysis the results of extended parallel sampling, as well as the results from the EU project AMOHA have shown a good agreement between the laboratories. Table 5 (adopted from last year's EMEP VOC report) show the results for the parallel analyses between UBA and NILU for the compounds included in the trend study. Except for propene and benzene, all UBA's median values are within 10% from NILU's medians. The coefficient of variation (CoV) indicates however larger spread in data values for ethene, propene, benzene and toluene. For the butanes and pentanes that are among the components with the clearest downward trends, the agreement between the two laboratories is almost perfect.

Thus, it is not likely that the downward trends from 1993-2000 indicated by these results are caused by changes in responsible laboratories (or in other analytical changes). For benzene, the strong decline apparent during this period, may to a limited extent be exaggerated by a bias between UBA's and NILUS laboratories, though.

Table 5: 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
n-pentane	94.500	103.000	2.000	6.672	0.071
i-pentane	163.000	162.500	0.500	12.602	0.077
benzene	205.000	179.000	-27.000	28.169	0.137
toluene	145.000	143.500	-16.000	31.875	0.220

4.3 Source regions and trajectory calculations

To investigate the region of origin for the hydrocarbons, air mass back trajectories were calculated to the VOC stations for all of 2000 by use of the FLEXTRA trajectory model (Stohl and Koffi, 1998). These trajectories are 3-dimensional and were calculated for 4 days' length backwards from the receptor sites based on meteorological data given every 6 h.

The 20 most pronounced peak episodes of each of the hydrocarbons were selected based on the difference between the actually observed value and an average seasonal function (based on a sine fit) through the year, but neglecting the months May-August as the photochemistry may mask the episodes in summer. The hourly positions of the trajectories arriving at noon on these 20 days were then mapped into the EMEP grid cells (150 km based) and used to construct maps showing the total residence time inside each grid cell. Only trajectory positions below 2000 m height were used in this mapping. The results for the fairly long-lived compounds propane and acetylene for 5 sites are shown in Figure 19.

The resulting maps indicate that the source regions of the peak episodes vary considerably between the stations. This emphasizes the need for a minimum number of VOC monitoring sites. Whereas the Utö episodes were more influenced by transport from SE and E, Waldhof received episodes from W and SW (UK and France), Košetice from SE and the region all around, except Germany, Donon from Germany and France, and Peyrusse Vieille from the SW Mediterranean plus UK. Thus, each station apparently shows its own characteristic in terms of source regions for the selected episodes. Furthermore, less differences are seen when comparing the mapped residence times for propane vs. acetylene, which is to be expected, as many of the episodes will be the same

for these two compounds. Some details are evident, however. For Utö, more of the propane episodes were linked to transport from E, SE, and N whereas acetylene episodes were associated with transport more from W and from the nearby regions. For Waldhof the propane episodes' source regions were more widespread, whereas for acetylene the source regions were more confined to emission regions in Germany and UK.

This analysis refers to the episodes seen in 2000 and the number of data and trajectories are obviously too few to make conclusions bout the climatological average conditions, which would require several years of trajectory information.

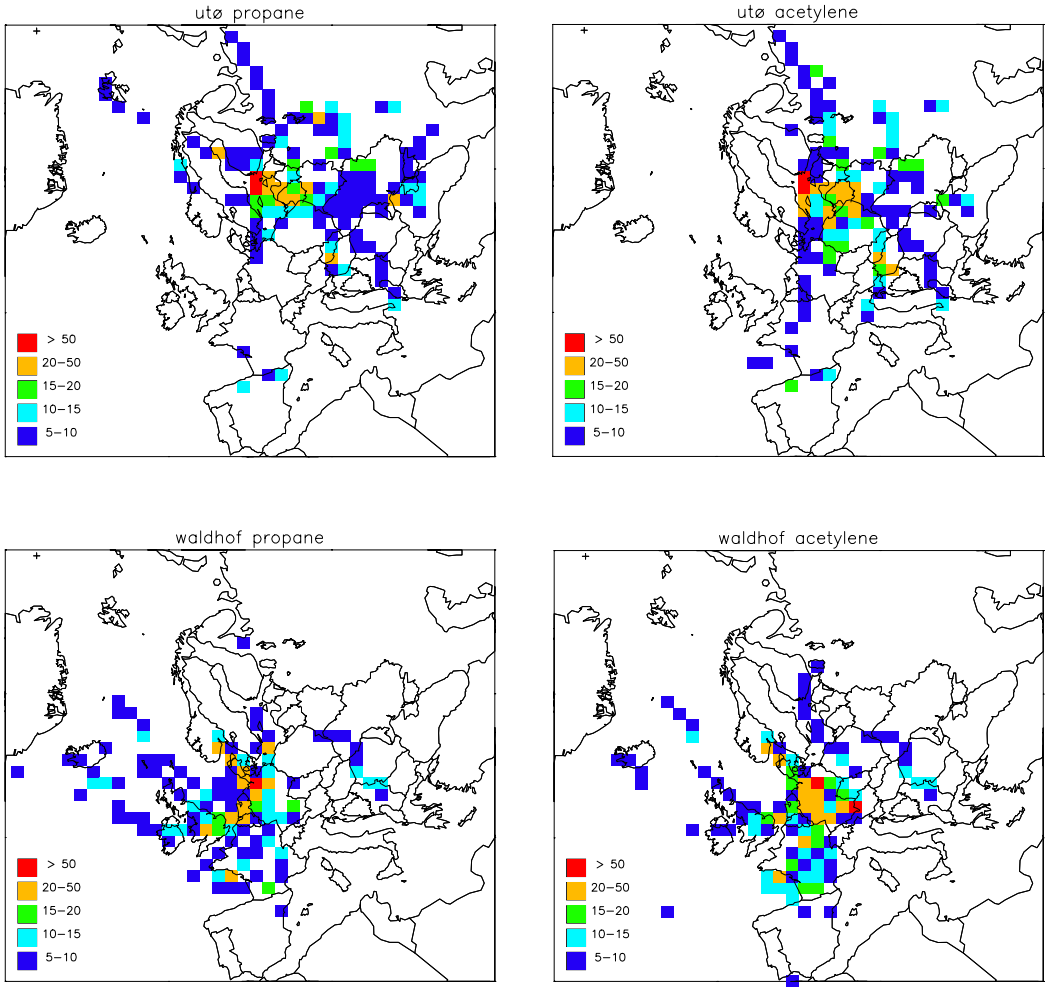


Figure 19: Hourly residence time, i.e. hourly trajectory crossings, for the 20 peak episodes based on 4 days back trajectories.

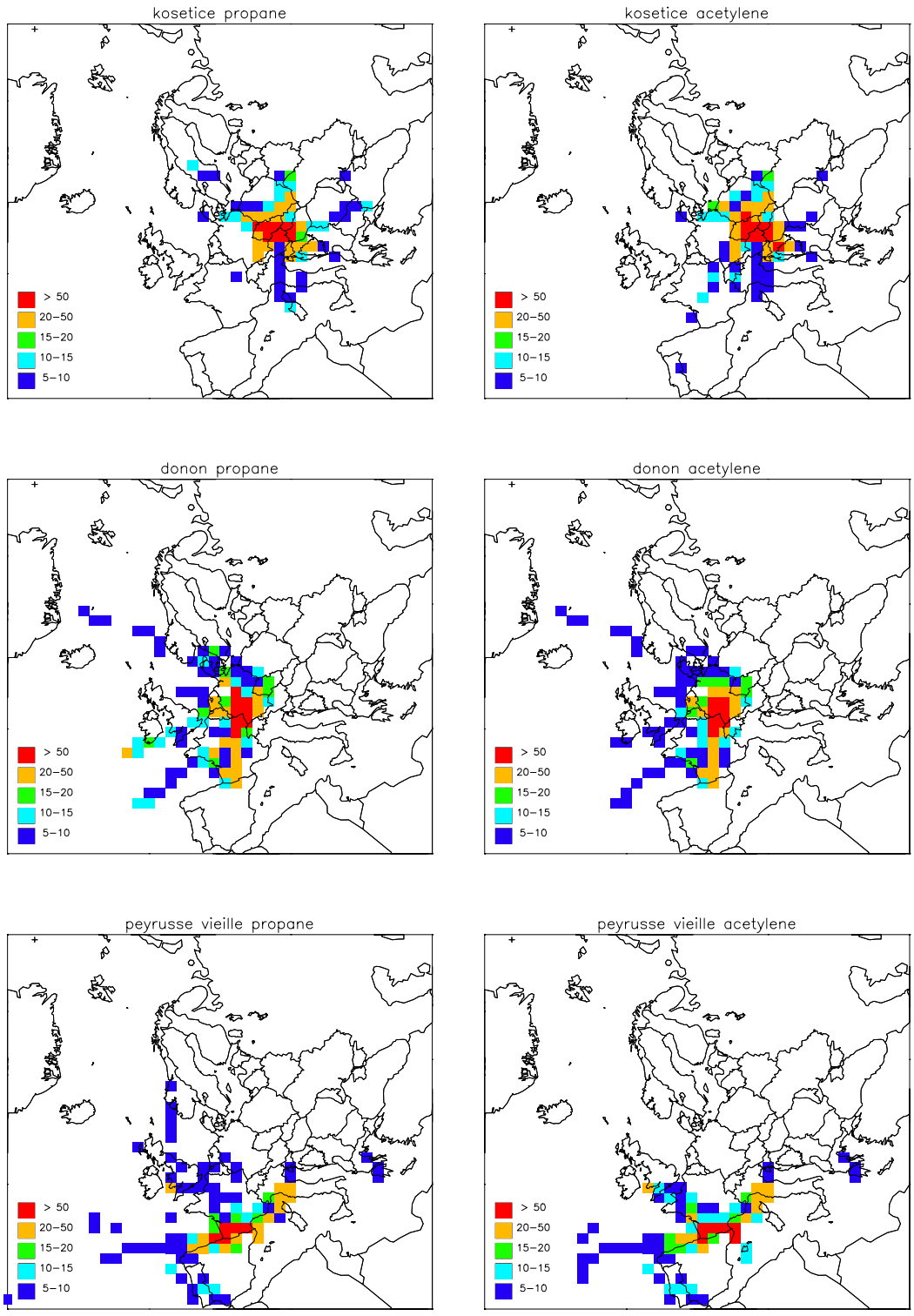


Figure 19, cont.

5. Conclusions

Measurements of VOC at 11 EMEP sites in 2000 have been presented including new monitoring activity in Germany and Finland. Based on a visit in 2001 by CCC to UBA a number of problems and improvements to the technical procedures were sorted out. The parallel analyses of hydrocarbons by UBA and NILU at Waldhof indicate that both institutes are in general within the DQO-limits (data quality objectives) for most of the data although some outliers are seen. For the carbonyls UBA's data should be regarded as preliminary and only formaldehyde, acetaldehyde and acetone are reported.

Monthly statistics have shown overall similar concentration levels as in the year before though with low mean concentrations in the first months of the year and fairly high concentrations in December. The concentration level of several compounds was particularly high at Starina during several months, and the concentrations of acetone at Birkenes in summer were also substantially higher than measured previously.

A simple trend evaluation indicated a marked decline in hydrocarbon concentrations from 1993 to 2000 in Germany and the Czech Republic as opposed to no evident trend in Finland. The estimated drop in the median concentrations during these 7 years was of the order of 20-50% with the largest downward tendency for benzene and the C_4 and C_5 compounds and the smallest change for propane. These changes corresponded well with the change in daily NO_2 concentrations during winter, indicating that the emission reductions in the hydrocarbons during1993-2000 have been of the same order as the emission reductions for NO_x.

The source regions for the peak episodes of individual hydrocarbons at the different sites were mapped by use of 3-dimensional back trajectories indicating that each station had it's own characteristic source regions. Some differences between the source regions for propane and acetylene were also indicated by these calculations.

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), Patrice Coddeville (EMD), Jiri Honzak (CHMI) 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 work was partly funded by the EU FP5 project TROTREP, project number EVK2-CT-1999-00043.

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Appendix A

Monthly mean and median concentrations of hydrocarbons and carbonyls in 2000

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

ETHANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	2119	1993	1991	1311	1076	769	642	559	673	1273	1666	–
	2157	2007	2060	1309	1049	732	701	548	645	1123	1555	–
Utö	2030	2181	2287	1825	1137	846	709	582	791	1450	1679	1896
	2063	2301	2363	1921	1202	810	751	641	705	1473	1372	1622
Zingst	2035	2153	2285	2067	1423	1068	878	853	–	–	2023	2567
	2045	2005	2188	2007	1477	1069	834	823	–	–	2024	2232
Waldhof-UBA	2148	2132	2102	2055	1435	1104	985	845	–	–	2039	2668
	1962	2114	2088	1968	1393	1105	1031	853	–	–	2004	2165
Schmücke	2173	2093	2058	1998	1448	1074	959	837	–	–	1702	2555
	2077	2096	2081	1838	1476	1093	997	823	–	–	1736	2303
Brotjacklriegel	1891	1919	2007	1799	1327	1023	771	853	–	–	1628	2175
	2030	1977	2049	1759	1303	959	743	823	–	–	1713	2002
Starina	1831	2957	3862	3963	2069	1137	1014	–	–	–	–	3626
	1770	3035	3705	3535	1745	1160	1210	–	–	–	–	3510
Košetice	2334	1970	2225	1766	1316	1095	694	752	1119	1322	1459	2123
	2167	1946	2233	1743	1306	1058	772	643	1027	1242	1392	1439
Donon	2235	1903	2147	1791	1374	997	806	799	1116	1179	1304	1805
	2220	1850	2060	1730	1350	1070	770	765	1090	1160	1370	1625
Peyrusse Vieille	2517	1796	1936	1663	1194	818	748	847	1236	1332	1390	1485
	2490	1760	1970	1580	1240	930	740	810	1240	1295	1370	1520

ETHENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	389	325	141	80	70	60	78	66	126	269	727	–
	305	259	162	51	75	56	80	64	104	235	654	–
Utö	353	477	332	302	160	113	161	141	293	605	770	1045
	326	415	326	296	157	88	93	154	221	593	715	756
Zingst	1036	766	725	516	385	308	335	334	–	–	1158	1749
	817	673	645	489	344	270	347	369	–	–	1115	1245
Waldhof-UBA	1184	835	586	508	374	301	331	351	–	–	1089	1934
	901	618	473	433	308	278	275	336	–	–	941	1245
Schmücke	1238	739	455	572	317	195	238	221	–	–	813	1642
	1176	680	417	256	288	174	233	166	–	–	687	1062
Brotjacklriegel	1472	757	620	453	307	313	291	334	–	–	700	1039
	1687	696	596	392	317	304	267	369	–	–	676	838
Starina	1152	950	1248	742	629	521	364	–	–	–	–	3486
	1360	970	1325	720	705	470	320	–	–	–	–	3300
Košetice	1503	1103	778	339	202	140	175	207	457	744	1152	1912
	1441	779	707	297	204	148	150	138	392	685	876	1448
Donon	1340	700	868	393	232	214	308	252	363	401	494	1015
	1470	670	760	300	220	210	240	225	250	270	425	855
Peyrusse Vieille	1384	506	459	304	316	316	334	387	423	537	516	659
	1300	500	390	250	320	320	320	380	320	485	410	595

PROPANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	1105	1189	1156	586	249	125	114	116	182	546	1209	–
	1132	1244	1172	533	249	125	74	95	165	463	913	–
Utö	1111	1220	1192	702	284	155	192	152	306	770	997	1170
	1114	1186	1105	723	250	155	154	142	237	697	722	929
Zingst	1044	1022	1000	706	383	265	252	343	–	–	879	1155
	1090	971	1057	695	403	196	240	271	–	–	917	1107
Waldhof-UBA	1154	1026	887	751	389	325	385	335	–	–	877	1334
	993	964	963	660	307	270	390	300	–	–	808	920
Schmücke	1096	975	828	662	380	236	326	281	–	–	666	1027
	1064	987	841	618	406	224	324	290	–	–	701	856
Brotjacklriegel	954	893	843	561	295	264	235	343	–	–	548	768
	945	924	867	534	308	244	211	271	–	–	546	725
Starina	794	880	988	1115	666	427	100	–	–	–	–	1848
	680	800	885	1070	460	420	110	–	–	–	–	1810
Košetice	1211	1055	913	665	356	289	215	285	537	844	753	1506
	1197	1070	959	650	299	262	184	245	451	953	674	1136
Donon	1029	780	890	537	320	258	210	194	263	406	503	878
	1020	800	870	460	250	300	260	160	260	340	530	680
Peyrusse Vieille	1284	744	719	489	287	151	154	224	399	490	577	695
	1220	760	720	445	290	130	150	230	340	505	570	715

PROPENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	48	29	20	24	20	25	25	21	22	28	68	–
	32	29	23	23	20	21	24	20	20	30	49	–
Utö	42	48	38	54	38	31	39	40	49	86	93	137
	34	43	46	54	37	28	32	39	35	83	88	126
Zingst	220	135	128	101	88	88	82	72	–	–	197	263
	190	106	112	93	78	69	82	75	–	–	170	195
Waldhof-UBA	263	148	125	78	77	70	89	70	–	–	222	294
	200	116	122	66	63	63	79	54	–	–	167	209
Schmücke	319	123	76	95	55	53	47	43	–	–	144	239
	167	125	69	56	49	42	46	40	–	–	115	151
Brotjacklriegel	264	139	101	67	66	83	76	72	–	–	111	129
	269	122	105	57	64	85	62	75	–	–	115	105
Starina	142	85	93	110	151	171	213	–	–	–	–	562
	120	65	60	70	140	130	200	–	–	–	–	540
Košetice	202	151	138	46	44	29	34	35	66	137	199	370
	192	114	110	43	32	32	30	32	48	110	155	338
Donon	225	117	144	86	76	62	78	68	90	107	114	191
	235	110	140	70	70	60	70	60	80	80	100	155
Peyrusse Vieille	214	103	91	88	86	93	96	113	141	132	134	145
	190	100	80	80	80	80	90	110	95	120	120	130

ACETYLENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	612	707	600	439	217	87	77	57	154	355	678	–
	600	673	630	388	212	78	58	49	115	249	587	–
Utö	590	763	729	635	280	153	162	137	324	671	715	948
	593	717	702	675	300	145	145	118	265	634	671	864
Zingst	945	961	958	848	408	237	186	272	–	–	936	1544
	808	797	930	753	371	162	165	182	–	–	878	1085
Waldhof-UBA	1079	938	772	785	381	256	209	252	–	–	830	1597
	942	843	706	780	382	223	204	219	–	–	770	1100
Schmücke	1269	1035	796	752	482	230	276	320	–	–	778	1490
	1155	954	732	635	403	212	313	317	–	–	729	1031
Brotjacklriegel	1367	941	907	824	407	303	223	272	–	–	627	920
	1520	910	898	845	434	295	221	182	–	–	634	783
Starina	1054	880	967	563	526	329	343	–	–	–	–	3660
	1150	705	890	300	525	270	290	–	–	–	–	3900
Košetice	1499	1371	1207	829	392	285	220	334	678	1157	1155	2693
	1427	1169	1199	776	364	281	215	285	518	1121	839	2013
Donon	873	582	661	424	256	147	100	154	190	430	287	596
	820	630	610	370	220	140	100	120	230	340	255	440
Peyrusse Vieille	849	484	460	308	188	103	106	153	200	393	331	360
	840	510	460	270	200	100	110	170	190	370	250	375

N-BUTANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	483	473	479	227	63	32	31	27	63	230	519	–
	481	505	445	146	67	33	15	21	51	186	379	–
Utö	466	522	438	338	107	54	104	58	132	310	423	528
	438	523	384	318	91	39	70	47	124	291	319	422
Zingst	478	446	401	261	140	124	108	150	–	–	428	534
	475	389	366	244	135	71	105	127	–	–	409	495
Waldhof-UBA	622	434	421	301	149	149	147	154	–	–	436	632
	580	383	399	248	116	137	126	139	–	–	363	453
Schmücke	544	458	315	336	205	115	178	172	–	–	358	488
	457	452	301	278	184	102	197	185	–	–	359	378
Brotjacklriegel	438	395	346	182	122	117	150	150	–	–	297	320
	417	402	347	176	106	103	141	127	–	–	229	296
Starina	399	397	430	808	413	374	271	–	–	–	–	918
	310	335	395	915	380	410	240	–	–	–	–	1050
Košetice	550	441	425	244	138	152	92	128	266	404	349	699
	525	448	410	226	108	108	81	101	213	410	263	530
Donon	459	297	394	261	174	116	106	116	134	179	379	500
	480	290	390	280	180	130	70	85	100	120	200	275
Peyrusse Vieille	468	261	228	134	88	48	54	83	121	138	208	255
	480	270	240	110	110	50	50	80	105	155	180	260

I-BUTANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	256	254	225	141	35	24	22	21	38	123	280	–
	253	264	219	97	38	20	19	17	39	110	200	–
Utö	254	276	234	152	64	32	54	32	74	165	233	303
	232	274	212	157	45	26	51	25	66	154	179	249
Zingst	254	242	245	158	84	72	53	78	–	–	234	295
	266	218	215	146	81	47	46	64	–	–	217	274
Waldhof-UBA	336	241	281	167	80	82	83	81	–	–	235	357
	327	218	217	143	62	73	72	64	–	–	193	246
Schmücke	293	250	162	157	99	57	87	81	–	–	183	271
	278	248	156	134	82	57	87	79	–	–	182	205
Brotjacklriegel	251	232	207	118	68	68	70	78	–	–	135	185
	244	233	203	116	63	57	71	64	–	–	123	169
Starina	231	212	345	623	384	483	201	–	–	–	–	530
	160	185	225	540	220	250	110	–	–	–	–	470
Košetice	310	241	240	142	83	82	47	74	148	245	197	421
	296	244	212	131	78	59	39	62	123	250	149	308
Donon	313	203	267	149	99	78	48	66	100	129	132	376
	310	210	260	150	80	90	50	50	100	70	120	195
Peyrusse Vieille	263	146	153	91	49	27	23	23	44	80	106	166
	260	150	140	75	60	30	20	25	40	75	90	175

1-BUTENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	3	3	4	6	3	3	3	3	3	3	3	–
	3	3	3	3	3	3	3	3	3	3	3	–
Utö	3	3	3	3	3	3	3	3	3	3	3	3
	3	3	3	3	3	3	3	3	3	3	3	3
Zingst	53	38	32	30	25	23	21	21	–	–	49	51
	57	36	32	33	25	24	20	20	–	–	48	40
Waldhof-UBA	71	43	53	30	25	20	22	22	–	–	53	61
	70	38	46	30	24	19	21	20	–	–	41	46
Schmücke	52	34	26	30	19	17	14	18	–	–	38	54
	48	31	24	24	16	13	14	15	–	–	33	38
Brotjacklriegel	77	50	39	27	25	27	25	21	–	–	37	34
	68	47	36	26	23	27	19	20	–	–	36	33
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Donon	30	12	15	7	7	6	5	7	8	11	11	25
	35	10	10	5	5	5	5	5	5	5	5	20
Peyrusse Vieille	38	16	13	15	11	18	12	21	29	30	23	29
	40	20	10	10	10	20	10	20	20	25	20	20

TRANS-2-BUTENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	4	3	3	8	3	3	3	3	3	3	3	–
	3	3	3	3	3	3	3	3	3	3	3	–
Utö	3	3	3	3	3	3	3	3	3	3	3	3
	3	3	3	3	3	3	3	3	3	3	3	3
Zingst	6	3	5	4	5	4	5	4	–	–	6	7
	6	3	3	3	3	3	3	3	–	–	6	7
Waldhof-UBA	12	5	12	6	5	3	6	4	–	–	8	10
	8	5	6	5	5	4	3	3	–	–	6	8
Schmücke	9	6	5	11	4	3	5	5	–	–	8	13
	8	3	6	9	3	3	3	3	–	–	3	9
Brotjacklriegel	11	9	8	3	3	4	4	4	–	–	11	6
	11	8	7	3	3	3	3	3	–	–	7	7
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Donon	6	5	6	5	8	5	5	7	6	6	5	8
	5	5	5	5	5	5	5	5	5	5	5	5
Peyrusse Vieille	6	5	6	5	5	5	5	6	6	5	5	5
	5	5	5	5	5	5	5	5	5	5	5	5

CIS-2-BUTENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	3	3	3	5	3	3	3	3	3	3	3	–
	3	3	3	3	3	3	3	3	3	3	3	–
Utö	3	3	3	3	4	3	3	3	3	3	3	3
	3	3	3	3	3	3	3	3	3	3	3	3
Zingst	7	5	3	4	5	4	4	4	–	–	7	11
	7	4	3	3	3	3	3	3	–	–	8	12
Waldhof-UBA	11	6	11	5	4	4	5	4	–	–	10	14
	9	5	7	5	3	3	4	3	–	–	11	12
Schmücke	8	5	4	9	4	4	4	4	–	–	8	15
	7	3	3	8	3	3	3	3	–	–	6	11
Brotjacklriegel	11	8	7	4	3	3	3	4	–	–	13	9
	9	8	6	3	3	3	3	3	–	–	10	9
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Donon	5	5	5	5	7	5	5	7	6	5	5	7
	5	5	5	5	5	5	5	5	5	5	5	5
Peyrusse Vieille	7	5	5	5	5	5	5	5	7	5	5	5
	5	5	5	5	5	5	5	5	5	5	5	5

N-PENTANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	55	68	78	27	7	5	6	10	21	86	213	–
	53	68	67	15	7	3	3	9	14	56	135	–
Utö	58	67	65	52	26	8	14	18	82	150	171	227
	60	70	59	47	16	6	10	12	73	153	129	188
Zingst	149	139	124	83	64	57	38	64	–	–	165	198
	152	117	142	82	59	28	39	51	–	–	171	175
Waldhof-UBA	190	147	144	134	59	72	66	76	–	–	195	251
	186	133	141	129	42	73	55	50	–	–	136	199
Schmücke	177	154	95	98	68	51	79	77	–	–	132	170
	136	143	88	87	72	41	70	68	–	–	121	141
Brotjacklriegel	168	137	116	158	62	74	73	64	–	–	150	128
	155	130	110	82	54	72	60	51	–	–	84	121
Starina	174	187	312	1038	494	330	261	–	–	–	–	426
	120	190	215	205	255	190	220	–	–	–	–	390
Košetice	193	146	154	98	61	56	48	69	148	192	155	289
	185	152	142	90	55	51	45	46	105	181	111	217
Donon	160	97	112	74	53	49	28	59	70	69	63	129
	150	90	110	50	60	60	20	30	80	40	55	85
Peyrusse Vieille	122	63	48	28	26	18	57	54	48	48	62	74
	110	60	50	20	30	20	20	50	40	55	60	75

I-PENTANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	130	115	115	57	12	5	8	10	22	111	264	–
	114	108	112	24	13	3	3	3	20	79	204	–
Utö	112	137	106	153	41	19	44	37	97	167	302	329
	114	147	91	100	28	12	17	17	75	182	315	342
Zingst	223	219	192	148	138	143	95	125	–	–	268	319
	218	213	179	157	152	88	88	113	–	–	284	266
Waldhof-UBA	356	218	243	206	98	109	99	111	–	–	276	363
	312	198	243	204	65	115	74	94	–	–	245	261
Schmücke	293	253	147	183	127	88	142	151	–	–	225	312
	282	219	133	157	134	83	134	134	–	–	196	245
Brotjacklriegel	272	235	199	138	108	121	108	125	–	–	216	198
	277	205	189	161	91	87	102	113	–	–	163	202
Starina	278	182	233	1035	493	338	349	–	–	–	–	660
	170	150	175	195	265	220	150	–	–	–	–	580
Košetice	285	220	230	169	107	102	86	140	235	342	266	465
	281	203	175	175	87	98	83	91	206	325	199	370
Donon	280	148	241	144	133	102	66	136	167	130	136	251
	275	160	220	130	150	90	50	90	110	70	115	180
Peyrusse Vieille	274	123	112	71	67	53	56	80	274	142	113	138
	280	125	100	55	80	50	50	80	190	135	90	140

N-HEXANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	31	30	26	13	4	3	3	3	3	25	61	–
	31	32	26	3	3	3	3	3	3	18	38	–
Utö	30	31	25	20	11	3	5	5	17	40	51	80
	30	31	24	23	3	3	3	3	6	40	38	53
Zingst	55	48	46	28	31	33	23	34	–	–	61	76
	59	40	51	30	30	16	21	31	–	–	60	75
Waldhof-UBA	74	51	47	53	23	21	24	24	–	–	61	94
	71	45	52	34	15	17	19	13	–	–	51	68
Schmücke	59	53	33	32	23	17	29	24	–	–	44	67
	53	50	30	30	21	14	27	21	–	–	44	56
Brotjacklriegel	65	47	43	21	27	29	26	34	–	–	38	44
	60	45	38	20	22	21	26	31	–	–	31	39
Starina	124	72	70	124	104	106	170	–	–	–	–	162
	100	70	65	120	64	70	80	–	–	–	–	130
Košetice	60	49	55	30	24	18	16	28	59	72	58	115
	58	46	37	22	15	17	18	18	62	64	38	91
Donon	43	18	36	19	16	12	6	11	23	14	16	33
	40	20	30	20	10	5	5	5	5	5	15	20
Peyrusse Vieille	34	19	12	11	6	11	10	14	9	14	15	21
	30	20	10	8	5	10	10	10	5	10	15	20

ISOPRENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	4	4	4	4	4	9	30	13	4	4	6	–
	4	4	4	4	4	4	15	10	4	4	4	–
Utö	4	4	4	5	4	15	21	26	5	–	–	–
	4	4	4	4	4	4	10	29	4	–	–	–
Zingst	11	8	11	45	166	313	311	290	–	–	23	20
	8	7	10	21	154	223	294	281	–	–	20	17
Waldhof-UBA	15	12	13	14	30	43	84	58	–	–	34	25
	14	13	16	14	31	35	46	57	–	–	26	25
Schmücke	14	11	7	29	42	35	27	41	–	–	23	21
	15	13	6	27	40	29	25	36	–	–	23	14
Brotjacklriegel	21	19	26	32	249	339	165	290	–	–	47	23
	17	18	16	23	175	174	121	281	–	–	39	24
Starina	289	–	292	106	150	577	663	–	–	–	–	42
	150	–	130	80	90	320	350	–	–	–	–	30
Košetice	5	6	10	14	57	92	84	119	27	26	13	20
	3	3	10	11	39	74	83	48	20	18	7	11
Donon	24	27	40	111	523	984	592	1206	644	126	49	45
	25	30	30	70	240	590	620	1225	440	130	35	35
Peyrusse Vieille	6	6	13	45	321	1061	1400	1080	734	130	11	6
	5	5	5	20	280	850	1130	670	635	135	10	5

BENZENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	145	176	169	116	54	21	22	47	87	211	390	–
	155	163	167	103	48	22	18	57	85	157	342	–
Utö	134	164	183	165	76	36	46	57	142	242	364	423
	135	154	167	154	68	30	36	47	124	252	312	343
Zingst	216	216	213	196	99	68	52	71	–	–	244	385
	175	192	211	167	96	45	52	53	–	–	217	268
Waldhof-UBA	250	226	183	181	105	71	79	71	–	–	211	414
	237	218	165	170	86	58	62	67	–	–	204	248
Schmücke	301	236	187	187	120	64	84	88	–	–	198	353
	283	222	180	145	107	64	90	90	–	–	189	238
Brotjacklriegel	318	218	202	168	95	74	61	71	–	–	167	223
	348	219	201	165	95	69	55	53	–	–	160	178
Starina	374	302	282	225	209	126	156	–	–	–	–	682
	380	240	245	225	180	120	130	–	–	–	–	620
Košetice	325	318	287	180	96	66	45	68	110	225	154	335
	289	278	247	165	100	60	45	66	99	208	127	292
Donon	268	177	231	133	82	58	54	54	84	93	110	199
	265	170	220	110	80	60	50	40	70	70	110	155
Peyrusse Vieille	263	161	161	104	73	47	34	27	–	–	–	–
	260	155	160	95	80	50	40	20	–	–	–	–

TOLUENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Utö	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Zingst	204	186	144	115	118	145	60	98	–	–	308	316
	158	152	129	97	111	71	63	82	–	–	288	317
Waldhof-UBA	291	189	169	163	90	87	93	123	–	–	295	415
	208	171	141	168	58	74	76	88	–	–	273	339
Schmücke	375	263	128	176	122	83	170	139	–	–	263	324
	279	210	118	148	110	77	145	117	–	–	246	244
Brotjacklriegel	262	207	169	110	91	88	187	98	–	–	239	191
	254	194	157	93	72	77	115	82	–	–	157	184
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	216	237	240	121	93	70	57	72	134	199	203	284
	210	202	209	124	82	65	54	83	127	213	127	286
Donon	246	129	227	121	101	74	54	92	136	128	121	210
	255	140	210	90	120	70	50	60	100	70	105	150
Peyrusse Vieille	259	103	86	66	60	44	58	69	114	128	123	151
	240	105	60	50	60	50	50	50	80	145	90	155

ETHYLBENZENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Utö	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Zingst	22	22	24	17	18	14	8	9	–	–	40	49
	11	19	27	16	18	11	6	3	–	–	43	37
Waldhof-UBA	51	31	–	29	22	11	11	22	–	–	47	60
	39	28	–	25	16	11	9	19	–	–	42	51
Schmücke	55	31	13	26	17	9	27	18	–	–	43	52
	59	30	3	24	17	9	20	3	–	–	32	39
Brotjacklriegel	35	28	21	14	11	8	8	9	–	–	31	25
	41	24	22	11	12	8	6	3	–	–	15	26
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	33	37	37	24	17	11	10	12	26	42	37	57
	33	38	35	24	14	11	9	9	29	40	27	57
Donon	45	22	49	28	24	14	13	25	29	26	25	43
	50	20	50	30	20	10	10	10	20	10	20	30
Peyrusse Vieille	43	17	11	11	12	8	6	38	25	27	27	25
	40	20	10	5	10	5	5	25	20	30	20	25

M+P-XYLENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Utö	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Zingst	40	27	45	21	28	25	18	11	–	–	95	114
	35	19	39	19	18	20	12	11	–	–	103	89
Waldhof-UBA	76	45	52	31	24	11	14	25	–	–	113	141
	50	39	51	23	16	4	11	14	–	–	99	122
Schmücke	91	55	21	36	22	16	35	20	–	–	112	118
	86	41	17	33	19	16	24	7	–	–	87	89
Brotjacklriegel	55	50	29	20	20	4	15	11	–	–	89	56
	52	35	29	12	13	3	18	11	–	–	45	56
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	68	69	67	45	44	24	25	27	46	88	86	124
	59	59	60	37	28	18	25	24	43	76	69	126
Donon	95	46	94	44	41	19	22	49	56	58	63	101
	100	30	100	30	30	20	20	20	30	40	40	65
Peyrusse Vieille	87	30	22	21	26	22	18	62	59	50	53	54
	70	35	20	10	20	20	20	50	50	45	40	50

O-XYLENE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Utö	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Zingst	13	10	17	10	14	10	4	4	–	–	30	36
	3	3	3	12	14	9	3	3	–	–	27	26
Waldhof-UBA	–	–	–	20	15	8	–	14	–	–	37	53
	–	–	–	17	12	9	–	6	–	–	32	48
Schmücke	27	22	5	19	13	7	17	8	–	–	34	38
	3	17	3	20	3	3	3	3	–	–	22	31
Brotjacklriegel	9	17	3	11	5	4	3	4	–	–	30	10
	3	3	3	12	3	3	3	3	–	–	21	8
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	23	31	29	14	10	9	8	8	21	32	30	49
	20	23	25	15	10	7	7	7	18	29	22	46
Donon	46	21	41	30	22	16	13	24	28	26	25	43
	50	20	50	30	20	10	10	10	20	20	20	30
Peyrusse Vieille	49	21	14	18	16	17	11	40	34	32	27	28
	40	20	20	15	20	20	10	40	35	30	20	30

N-HEPTANE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Utö	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Zingst	28	25	20	18	16	13	12	14	–	–	28	32
	23	23	21	17	16	8	9	14	–	–	26	29
Waldhof-UBA	36	26	25	26	14	12	19	15	–	–	29	41
	31	24	25	27	11	13	12	10	–	–	24	33
Schmücke	42	30	19	20	16	12	17	21	–	–	31	33
	37	25	18	20	16	10	16	16	–	–	26	29
Brotjacklriegel	41	28	24	17	14	18	22	14	–	–	23	20
	39	24	23	18	14	16	19	14	–	–	22	20
Starina	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	18	15	19	10	6	5	3	6	8	16	11	20
	17	14	13	8	6	6	3	3	8	14	10	17
Donon	20	9	14	11	8	5	5	8	9	10	8	14
	20	5	10	10	10	5	5	5	5	5	5	10
Peyrusse Vieille	16	6	5	6	5	5	8	24	9	12	8	9
	10	5	5	5	5	5	5	20	10	10	5	10

Monthly mean and median concentrations (first and second line, respectively) of carbonyls (ng/m^3)

FORMALDEHYDE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	1085	513	565	–	486	534	578	274
	–	–	–	–	440	480	530	–	440	545	400	270
Birkenes	238	295	326	–	–	686	661	447	374	249	251	115
	250	300	320	–	–	750	545	440	370	240	220	115
Zingst	499	443	568	851	1051	1345	1414	1746	1408	1302	920	731
	360	380	440	670	1060	1200	1220	1700	1405	1100	840	645
Waldhof-UBA	972	1458	1208	2009	1807	1434	1050	1949	1111	1154	783	751
	860	1380	1090	1895	1700	1140	950	1710	1005	1070	780	775
Waldhof-NILU	695	765	625	1115	1449	1489	1246	1733	892	610	284	289
	655	710	585	1075	1360	1410	1020	1465	870	580	300	275
Schmücke	–	1355	614	1884	2232	2254	1443	2367	1360	1001	870	925
	–	1335	560	1795	2600	1880	1410	2260	1305	1090	830	950
Brotjacklriegel	257	325	317	418	620	970	528	1049	495	531	491	436
	200	260	280	385	550	820	520	850	445	460	480	445
Košetice	1119	986	800	1219	–	–	–	1867	1420	1351	612	544
	950	785	620	1115	–	–	–	1800	1530	1320	630	450
Donon	921	885	1340	1554	1988	2452	1423	2490	1779	769	549	648
	951	735	1507	1382	1726	2365	1532	2442	1664	608	501	648

ACETALDEHYDE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	857	390	715	–	657	776	857	378
	–	–	–	–	400	340	420	–	650	820	485	310
Birkenes	328	330	362	–	–	1116	1439	629	603	343	388	258
	350	335	380	–	–	1090	1325	620	660	340	330	250
Zingst	704	790	770	1115	1100	845	841	992	969	860	690	601
	730	785	790	975	1110	690	745	960	1000	810	710	550
Waldhof-UBA	798	1104	932	1218	1117	861	797	1037	941	857	667	718
	770	1055	950	1180	1120	840	730	1100	925	790	700	635
Waldhof-NILU	771	663	550	799	911	772	624	869	750	689	524	571
	750	705	480	745	960	850	580	855	830	585	460	450
Schmücke	–	1025	687	1243	1051	896	777	1020	970	636	539	584
	–	1025	710	1170	980	870	820	1040	985	640	570	545
Brotjacklriegel	372	383	413	645	656	629	506	616	546	439	281	286
	350	355	380	650	600	620	490	540	510	410	270	275
Košetice	1131	868	756	1088	–	–	–	2063	1308	1873	696	745
	1260	830	640	1050	–	–	–	2090	1090	1670	710	655
Donon	509	459	1005	1069	1000	954	569	748	786	399	319	459
	518	390	982	855	1170	980	546	735	707	329	295	484

ACETONE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	3337	1592	1946	–	1913	1814	1637	809
	–	–	–	–	1410	1590	1310	–	1870	1770	1250	810
Birkenes	3739	4003	1738	–	–	8620	8756	5786	5386	3794	3704	950
	1110	3755	1820	–	–	8970	8940	5580	5740	3240	2200	895
Zingst	1359	1310	1815	1909	1590	1914	2019	1887	2153	1864	1282	943
	1120	1260	1950	1665	1370	1600	2085	1740	2150	1640	1190	875
Waldhof-UBA	2846	2640	3124	3780	3082	2934	3159	2731	2559	2019	1387	1298
	2670	2685	2880	2905	3150	2630	2840	2510	2070	1690	1350	1355
Waldhof-NILU	1617	1972	2262	4116	3863	3547	3019	3325	2970	2255	1676	1289
	1410	1920	2330	3525	3700	3480	2790	3020	2590	1985	1590	1240
Schmücke	–	1530	1528	2900	2969	2600	2524	2942	2294	1750	1064	1108
	–	1505	1210	2690	3110	2560	2530	2490	2275	1710	1040	1115
Brotjacklriegel	3993	3059	3594	4239	3407	2962	3198	2648	3578	2953	2780	2518
	3950	3420	3650	4395	3700	2990	2900	2640	3310	2780	2430	2255
Košetice	2322	2425	2729	4077	–	–	–	5241	4368	3821	1726	1604
	2140	2470	2100	3755	–	–	–	5570	4330	3560	1920	1395
Donon	1269	1093	2189	2776	4400	4021	2836	3725	3403	1506	990	1184
	1292	945	2054	2484	5327	3940	2936	3538	3308	1394	1002	1119

PROPANAL

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	173	59	140	–	126	95	167	97
	–	–	–	–	80	50	70	–	130	100	95	100
Birkenes	47	67	56	–	–	108	111	66	76	42	92	54
	50	70	60	–	–	130	95	50	70	50	60	60
Zingst	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-UBA	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-NILU	102	90	66	90	111	108	110	134	108	86	82	96
	90	95	50	85	100	80	100	120	120	95	80	75
Schmücke	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Brotjacklriegel	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	153	128	88	158	–	–	–	271	130	333	84	119
	190	100	70	145	–	–	–	240	90	190	90	105
Donon	101	86	150	156	333	213	87	106	108	97	48	74
	95	68	143	133	250	207	76	103	108	56	49	78

BUTANONE

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	380	210	194	–	139	203	203	110
	–	–	–	–	210	180	195	–	150	200	145	110
Birkenes	233	268	310	–	–	170	174	127	131	98	87	74
	240	275	290	–	–	150	170	120	130	100	90	70
Zingst	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-UBA	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-NILU	566	590	541	688	571	549	501	479	260	191	166	178
	560	530	520	680	520	640	520	385	250	185	150	180
Schmücke	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Brotjacklriegel	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	613	786	644	822	–	–	–	427	196	452	171	255
	610	695	485	950	–	–	–	480	200	440	150	220
Donon	410	352	559	606	967	1405	802	1448	1123	260	183	304
	406	401	543	515	842	1236	822	1354	709	216	179	285

BUTANAL

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	143	57	104	–	117	165	172	126
	–	–	–	–	100	60	60	–	110	130	95	120
Birkenes	36	47	46	–	–	72	60	47	46	20	49	40
	40	50	40	–	–	70	55	50	50	20	30	35
Zingst	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-UBA	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-NILU	76	55	32	48	54	54	50	107	68	53	44	60
	70	55	30	45	50	40	50	80	70	45	40	60
Schmücke	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Brotjacklriegel	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	74	66	53	89	–	–	–	206	80	213	50	60
	90	55	40	75	–	–	–	170	20	110	50	55
Donon	56	53	70	74	72	144	41	54	65	24	19	28
	51	55	68	72	77	140	46	50	64	19	20	20

PENTANAL

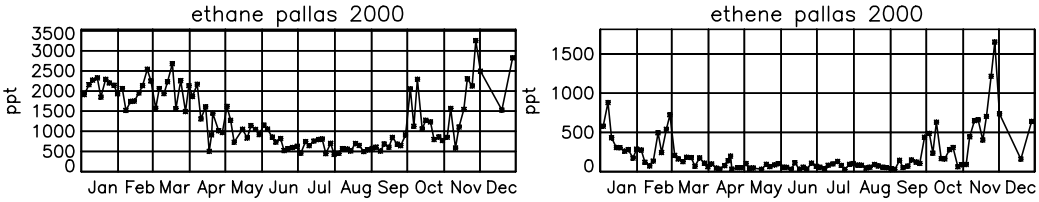
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	253	130	199	–	189	151	110	100
	–	–	–	–	260	100	185	–	190	175	120	100
Birkenes	39	67	62	–	–	164	113	159	130	173	210	80
	30	70	30	–	–	160	95	150	120	160	200	60
Zingst	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-UBA	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-NILU	20	20	18	24	24	23	20	31	48	102	164	148
	20	20	20	20	20	20	20	25	60	80	170	150
Schmücke	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Brotjacklriegel	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	28	76	53	54	–	–	–	89	28	131	76	113
	30	80	50	50	–	–	–	80	20	90	60	105
Donon	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–

GLYOXAL

	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Utö	–	–	–	–	125	53	89	–	81	84	98	111
	–	–	–	–	60	50	70	–	80	90	110	100
Birkenes	15	16	16	–	–	80	118	51	64	25	44	37
	15	15	15	–	–	80	130	50	60	15	50	35
Zingst	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-UBA	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Waldhof-NILU	18	15	46	54	99	112	74	104	74	65	41	65
	15	15	30	40	90	90	50	85	70	45	15	55
Schmücke	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Brotjacklriegel	–	–	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–	–	–
Košetice	41	34	32	50	–	–	–	57	39	120	111	166
	15	28	20	45	–	–	–	50	15	110	60	125
Donon	24	19	32	38	37	97	76	87	68	42	24	17
	12	12	29	40	12	72	77	93	47	36	27	12

Appendix B

Time series of VOC measured in 2000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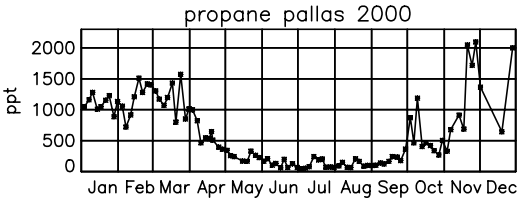
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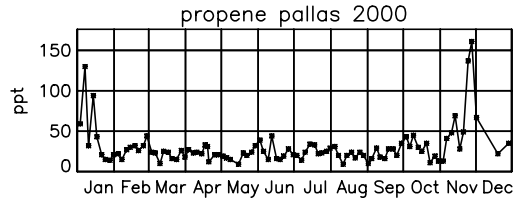
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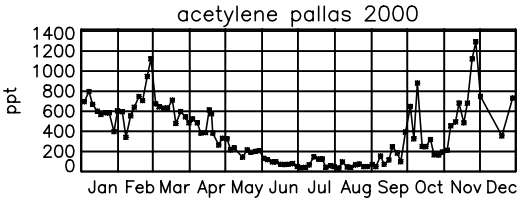
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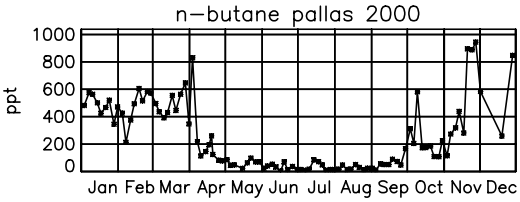
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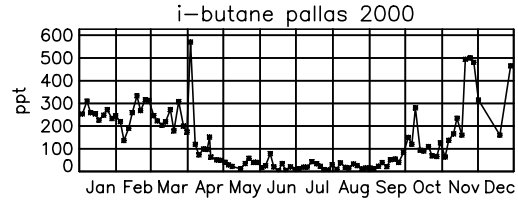
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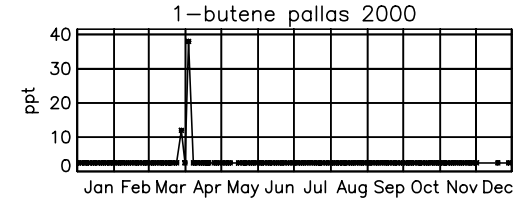
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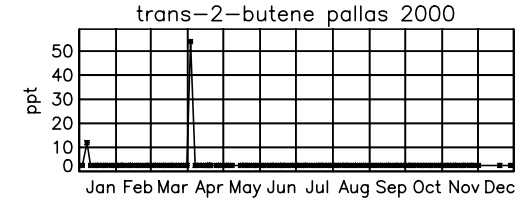
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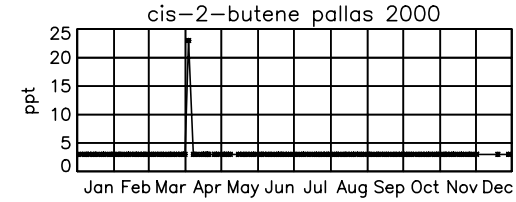
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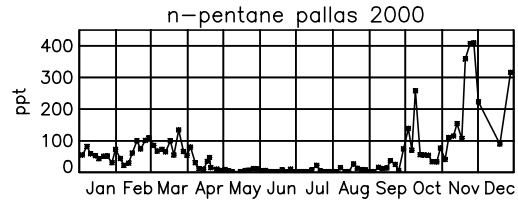
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ppt
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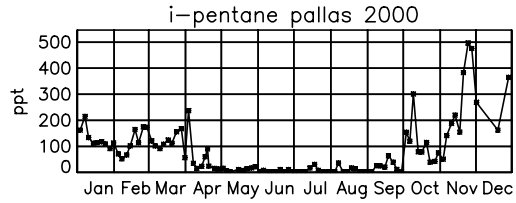
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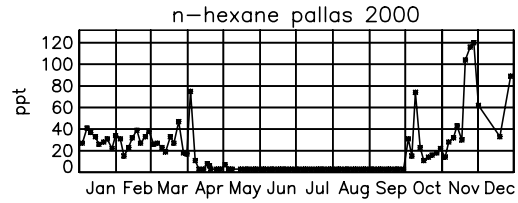
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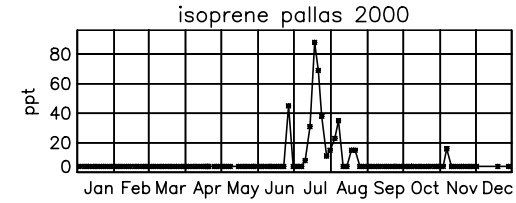
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ppt
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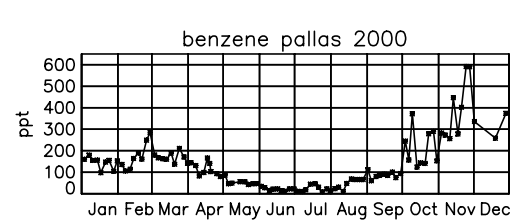
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ppt
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100
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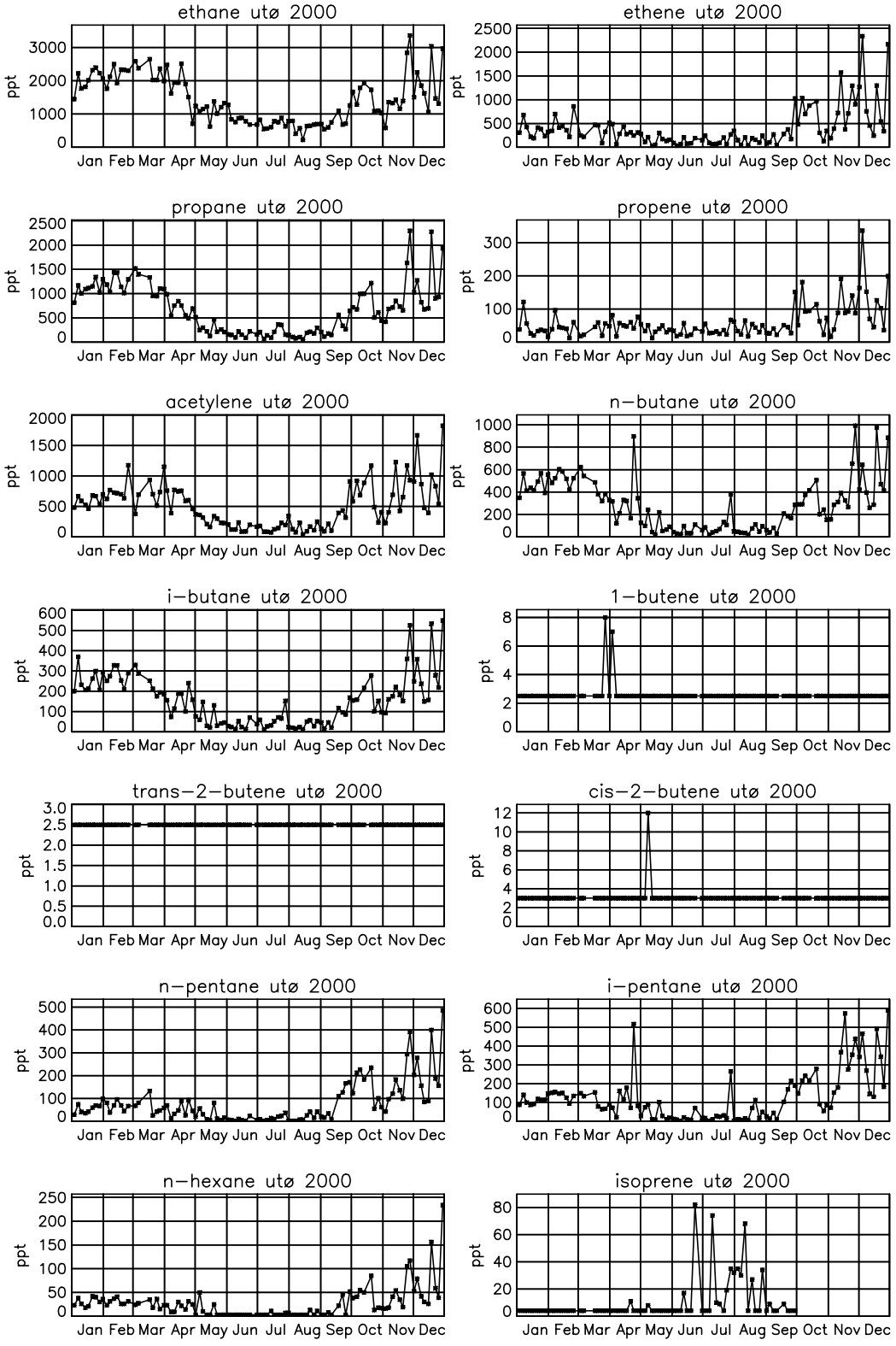
400
500
600
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethane utø 2000
ethene utø 2000
propane utø 2000
propene utø 2000
acetylene utø 2000
n−butane utø 2000
i−butane utø 2000
1−butene utø 2000
trans−2−butene utø 2000
cis−2−butene utø 2000
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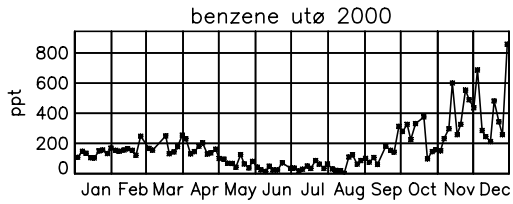
i−pentane utø 2000
n−hexane utø 2000
isoprene utø 2000
ppt
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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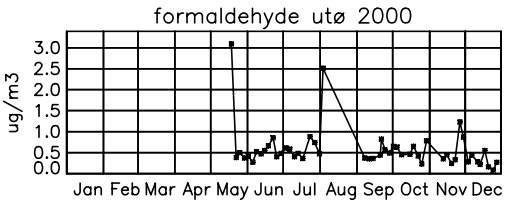
ppt
800
600
400
200
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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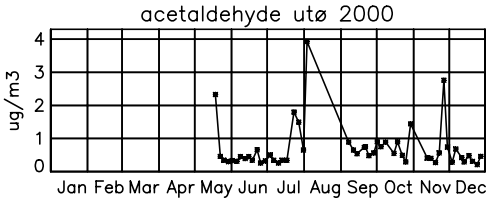
2.5
2.0
1.5
1.0
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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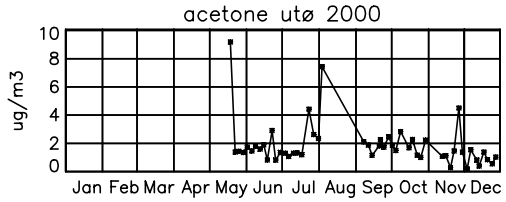
ug/m3
4
3
2
1
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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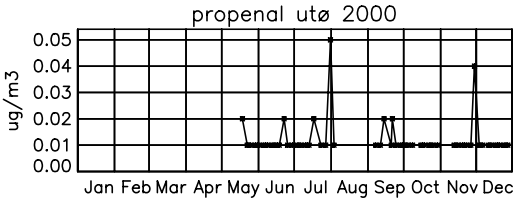
10
8
6
4
2
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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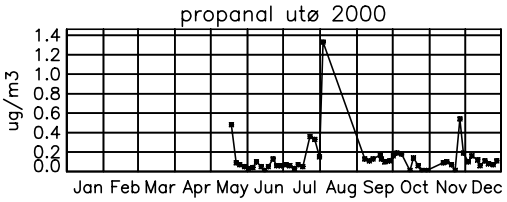
ug/m3
0.05
0.04
0.03
0.02
0.01
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propanal utø 2000
ug/m3
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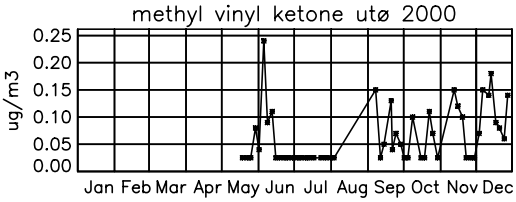
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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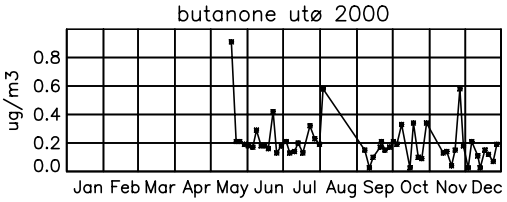
0.20
0.15
0.10
0.05
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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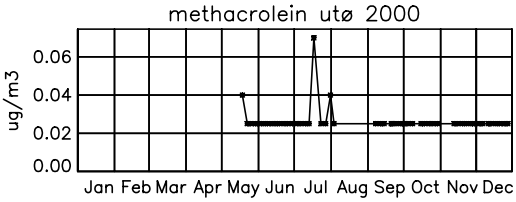
0.6
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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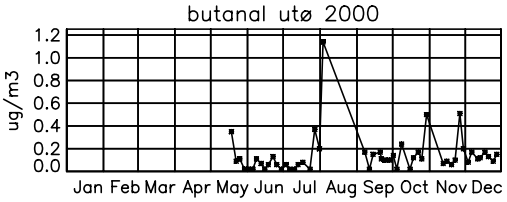
0.04
0.02
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


butanal utø 2000
ug/m3
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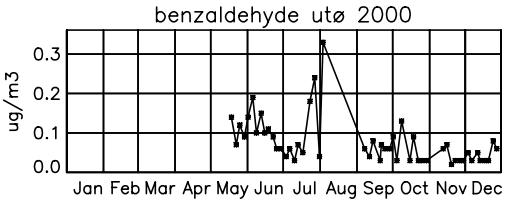
1.0
0.8
0.6
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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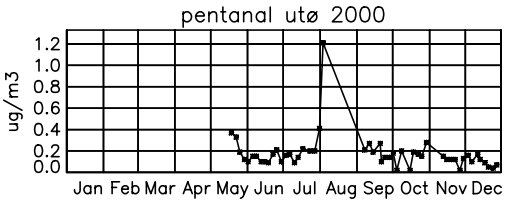
0.2
0.1
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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1.0
0.8
0.6
0.4
0.2
0.0
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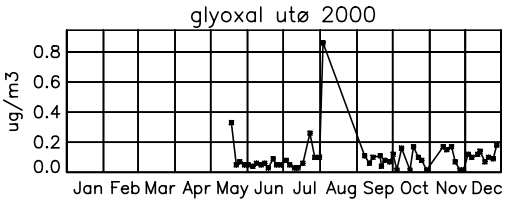
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ug/m3
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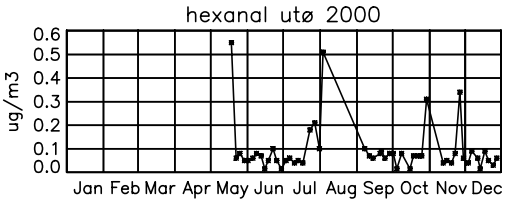
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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0.5
0.4
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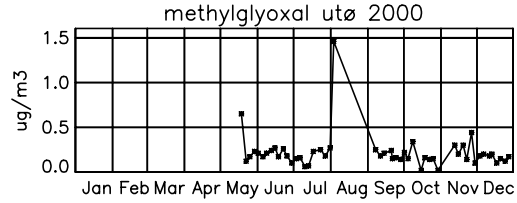
0.2
0.1
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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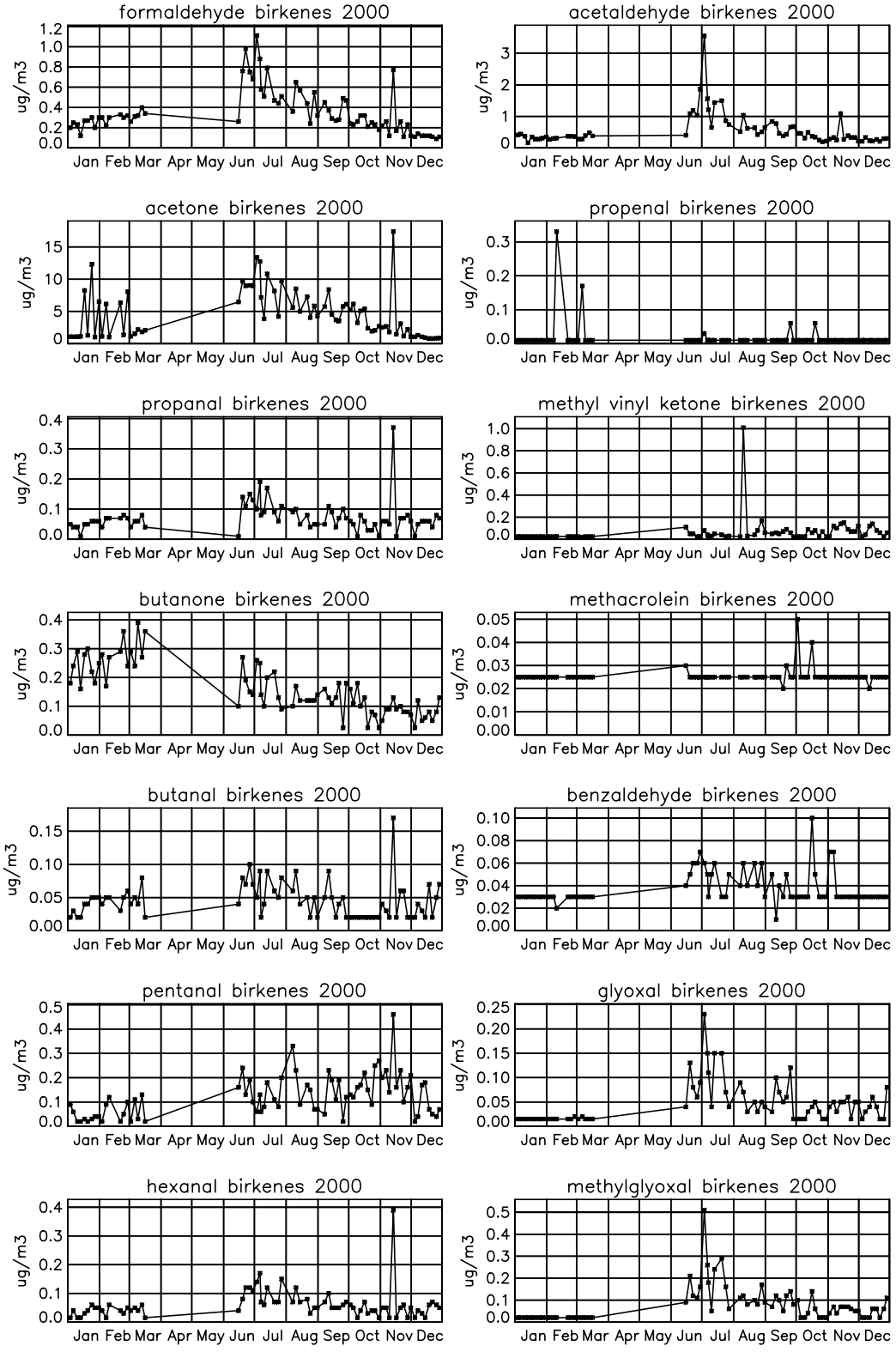
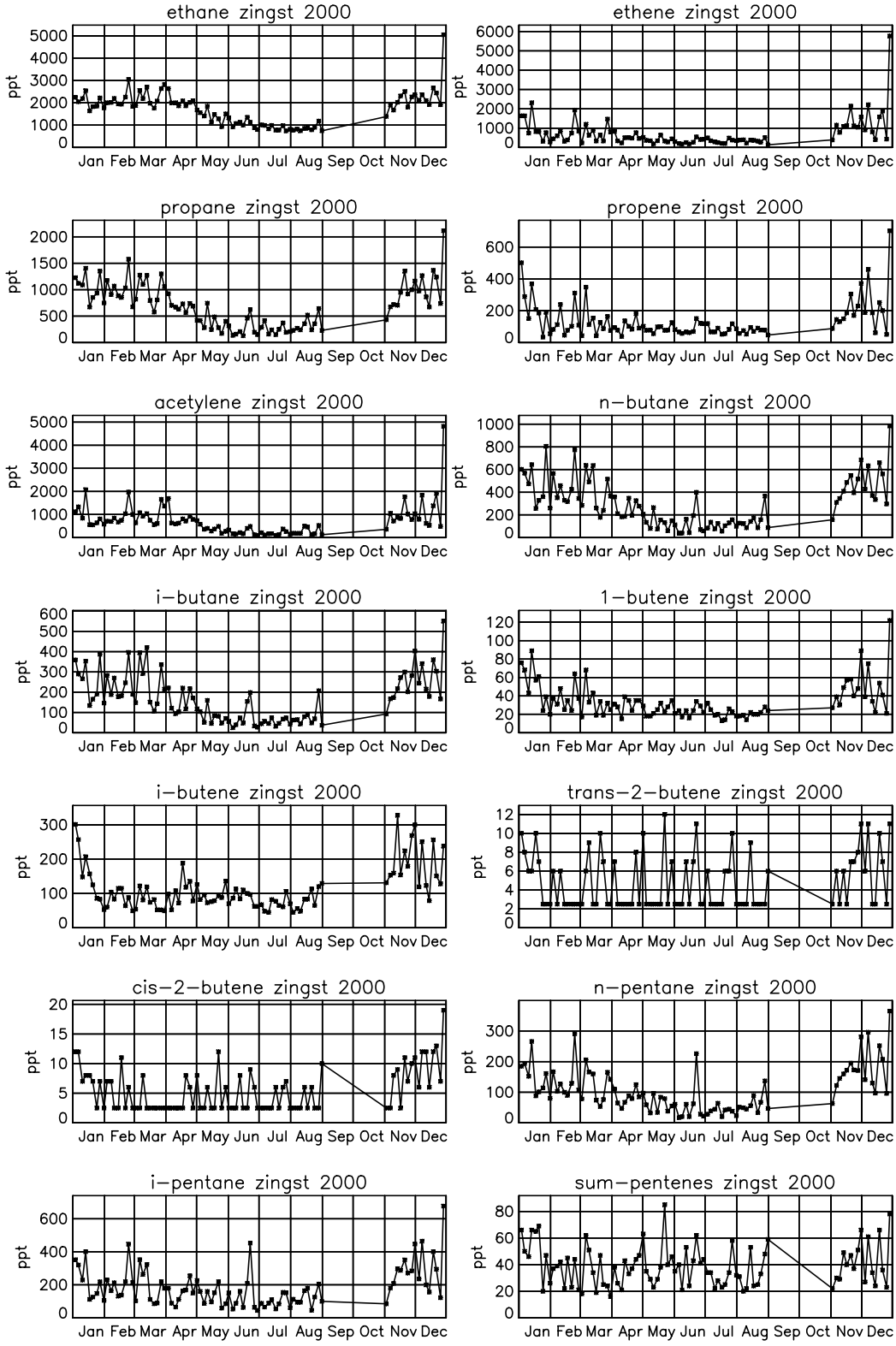
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


formaldehyde birkenes 2000
acetaldehyde birkenes 2000
acetone birkenes 2000
propenal birkenes 2000
propanal birkenes 2000
methyl vinyl ketone birkenes 2000
butanone birkenes 2000
methacrolein birkenes 2000
butanal birkenes 2000
benzaldehyde birkenes 2000
pentanal birkenes 2000
glyoxal birkenes 2000
hexanal birkenes 2000
methylglyoxal birkenes 2000
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethane zingst 2000
ethene zingst 2000
propane zingst 2000
propene zingst 2000
acetylene zingst 2000
n−butane zingst 2000
i−butane zingst 2000
1−butene zingst 2000
i−butene zingst 2000
trans−2−butene zingst 2000
cis−2−butene zingst 2000
n−pentane zingst 2000
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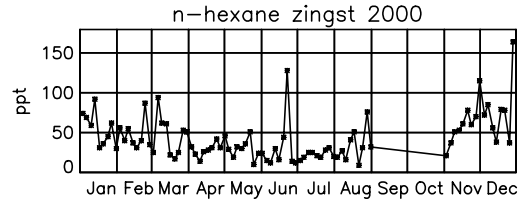
sum−pentenes zingst 2000
ppt
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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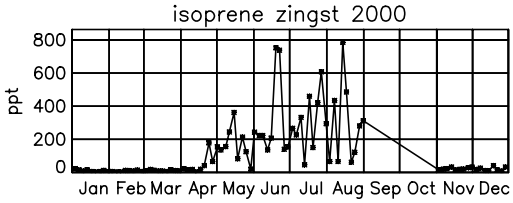
ppt
0
50
100
150
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ppt
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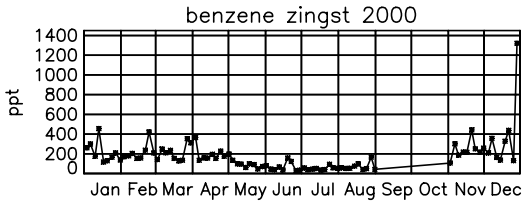
400
600
800
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ppt
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200
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600
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1000
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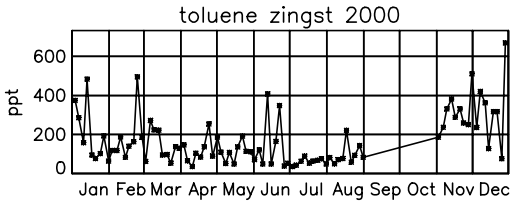
1400
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ppt
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600
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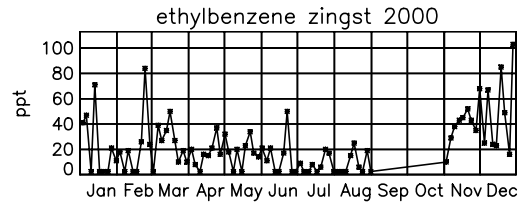
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ppt
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60
80
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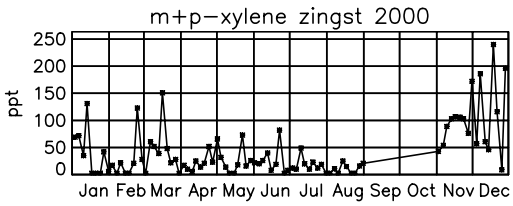
m+p−xylene zingst 2000
ppt
0
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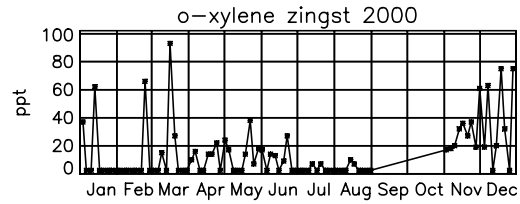
100
150
200
250
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


o−xylene zingst 2000
ppt
0
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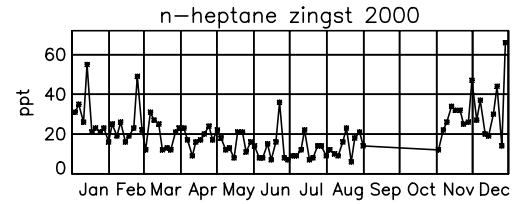
60
80
100
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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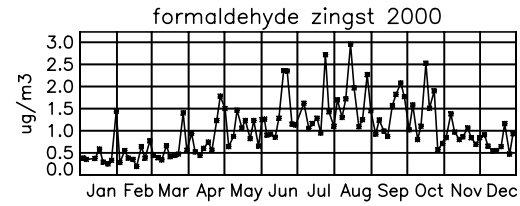
ppt
0
20
40
60
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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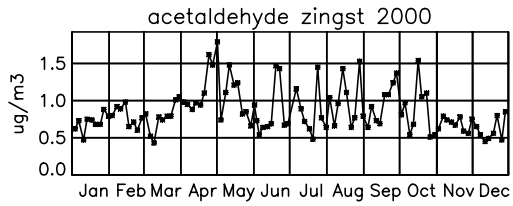
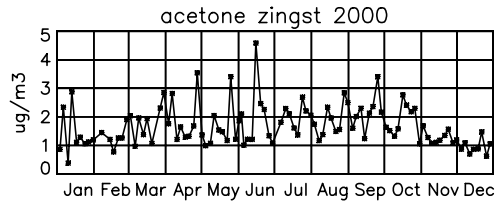
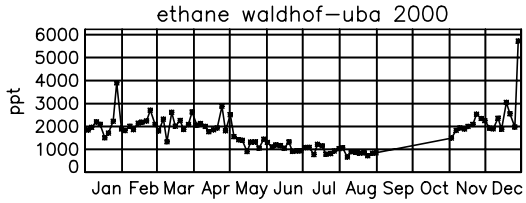
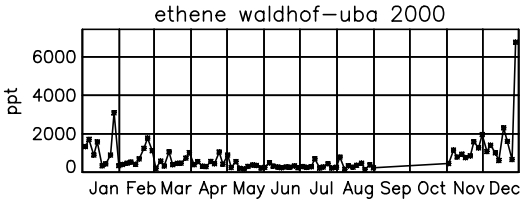
0.5
1.0
1.5
2.0
2.5
3.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


acetaldehyde zingst 2000
ug/m3
0.0
0.5
1.0
1.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


acetone zingst 2000
ug/m3
0
1
2
3
4
5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethane waldhof−uba 2000
ppt
0
1000
2000
3000
4000
5000
6000
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethene waldhof−uba 2000
ppt
0
2000
4000
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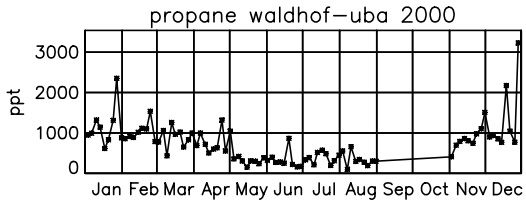
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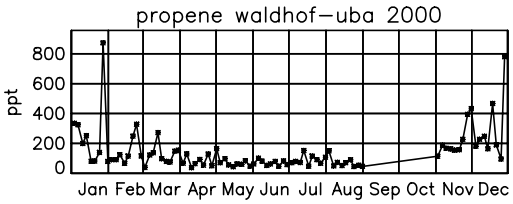
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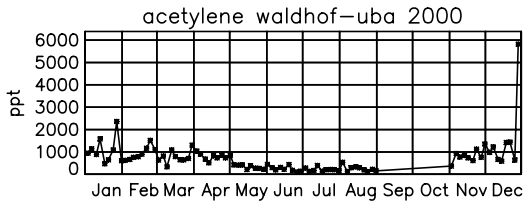
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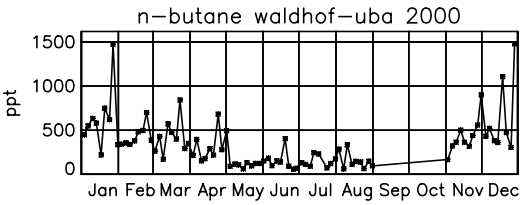
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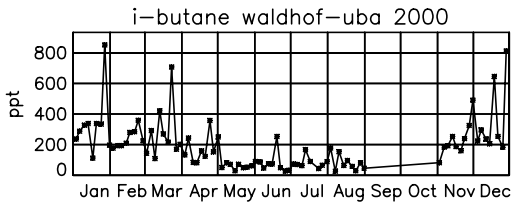
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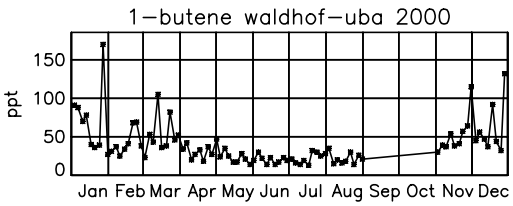
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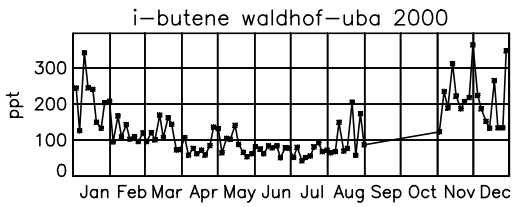
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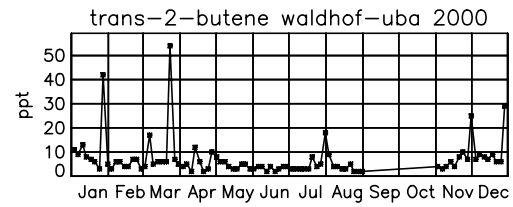
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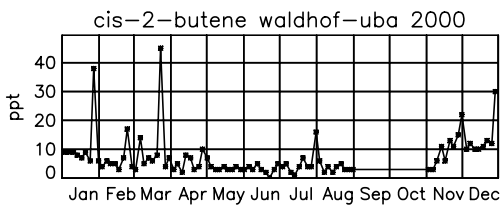
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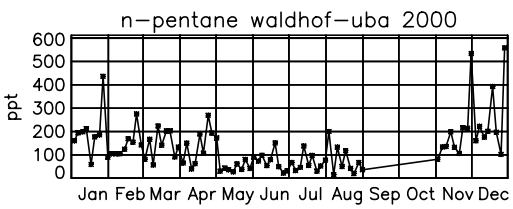
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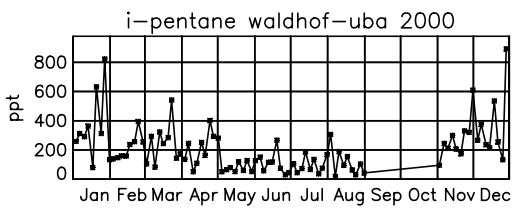
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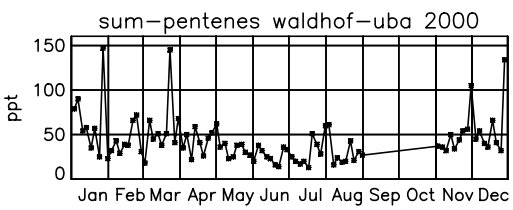
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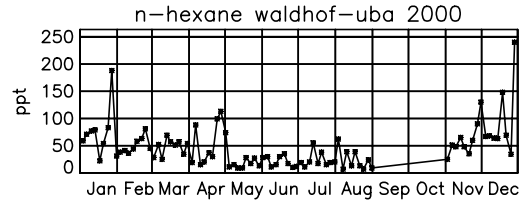
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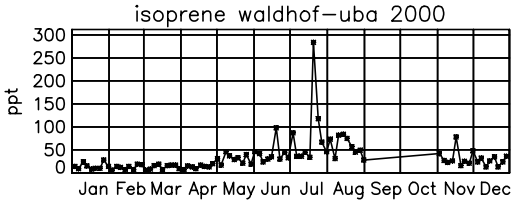
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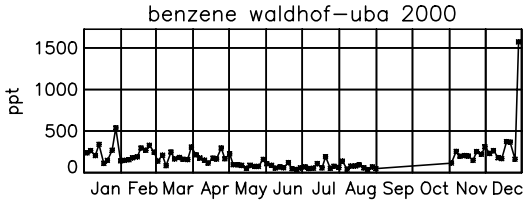
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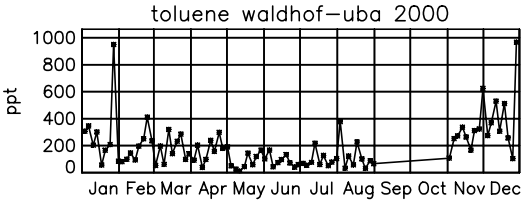
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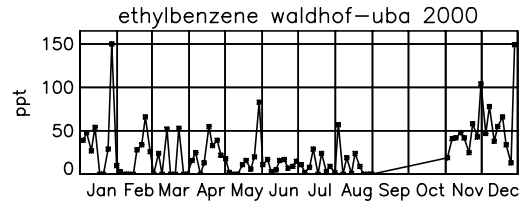
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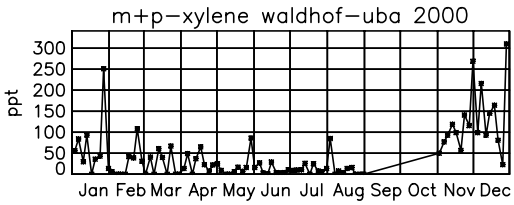
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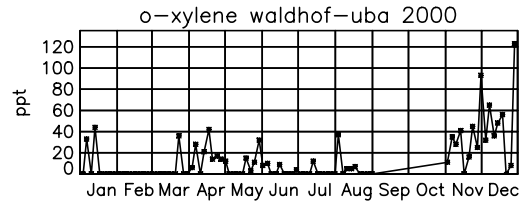
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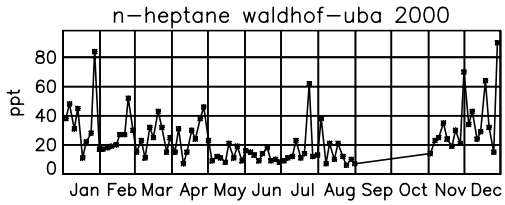
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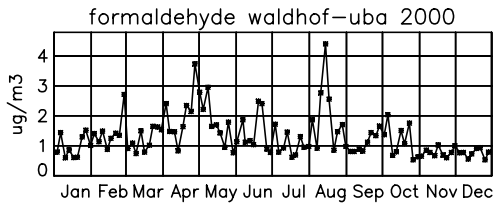
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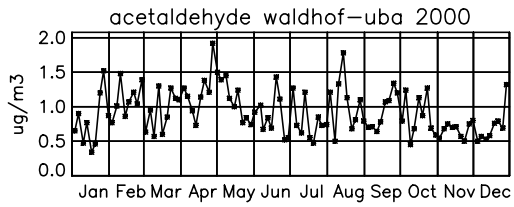
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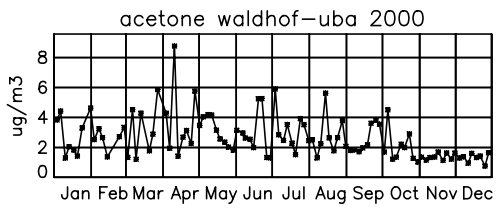
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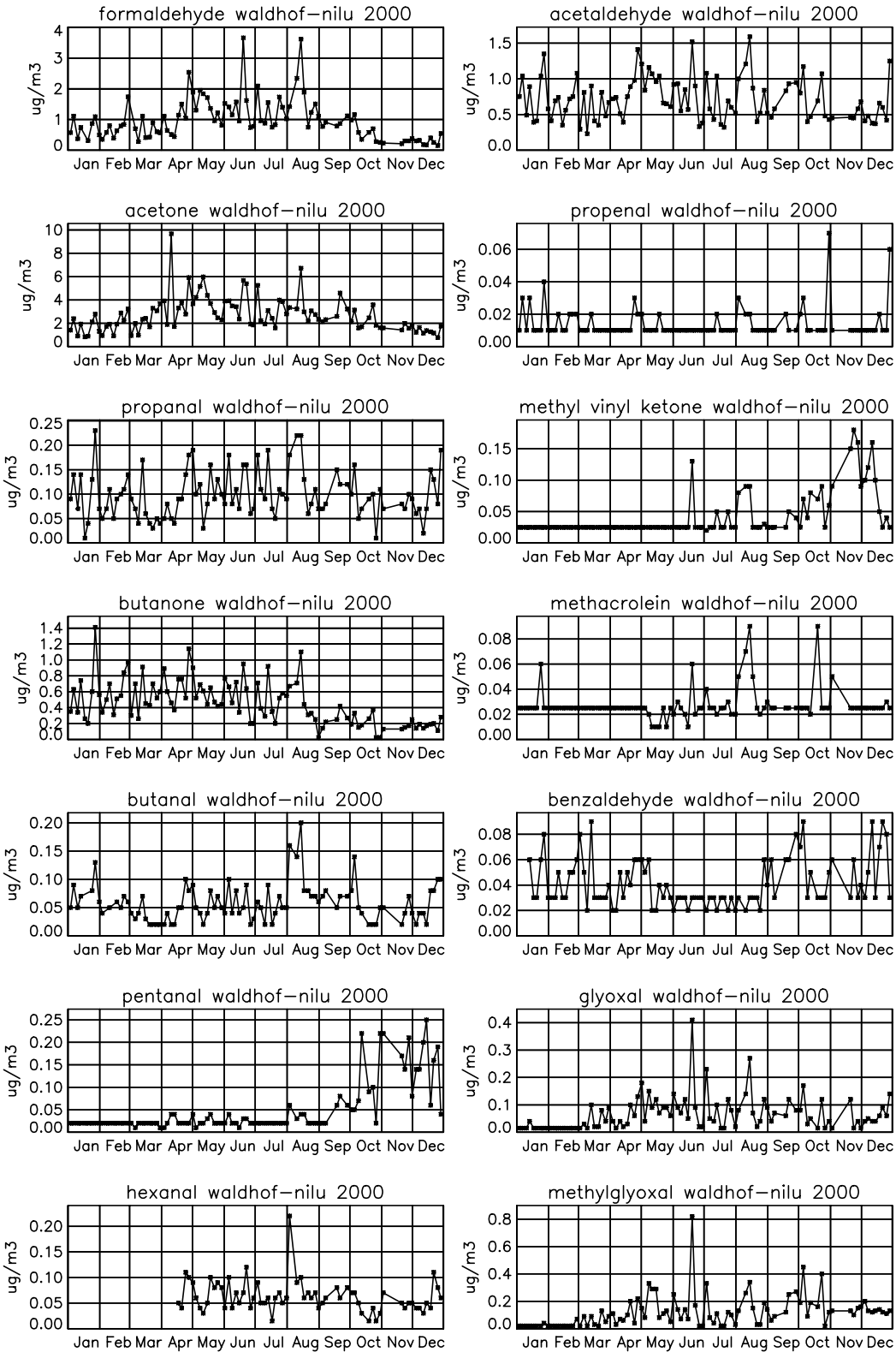
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benzaldehyde waldhof−nilu 2000
pentanal waldhof−nilu 2000
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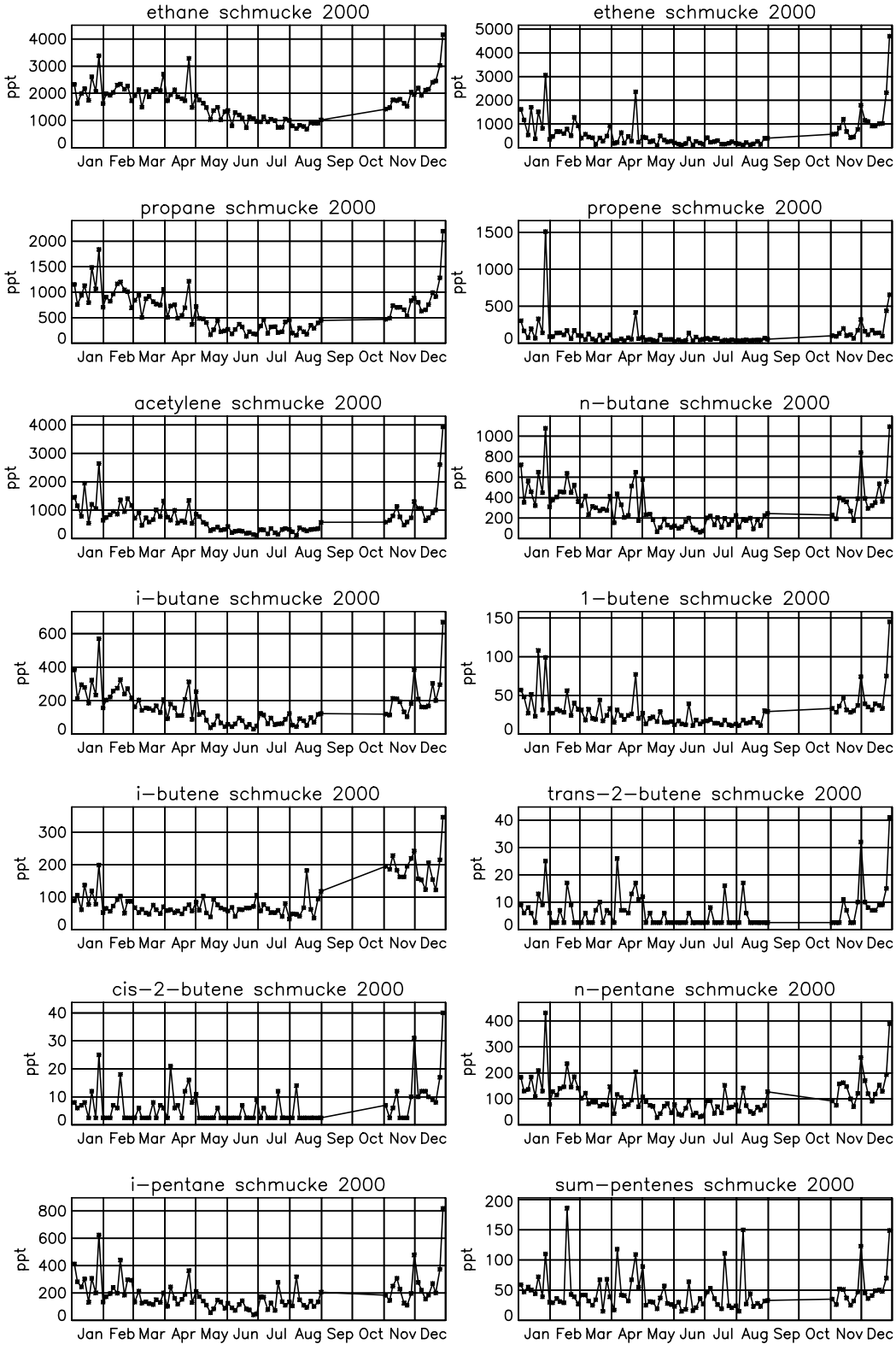
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i−butene schmucke 2000
trans−2−butene schmucke 2000
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ppt
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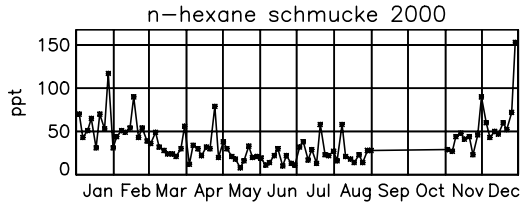
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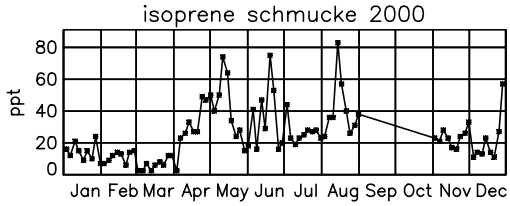
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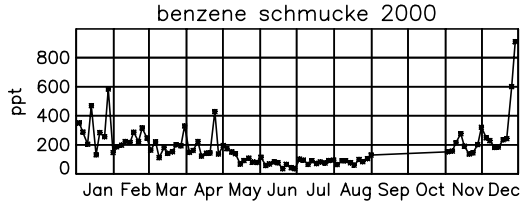
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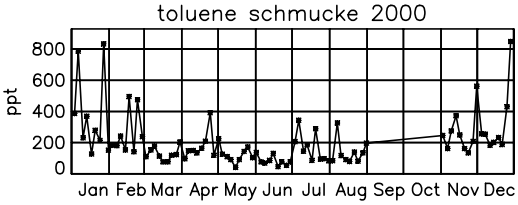
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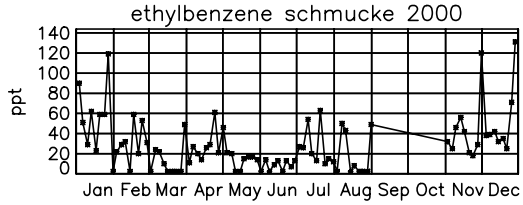
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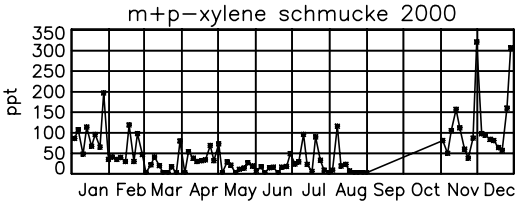
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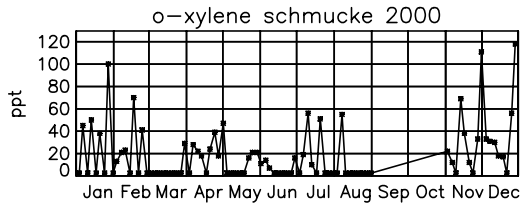
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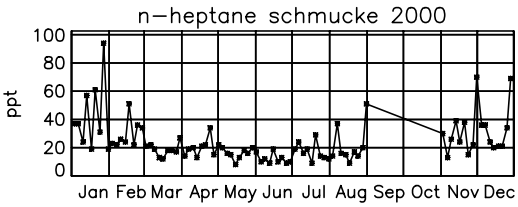
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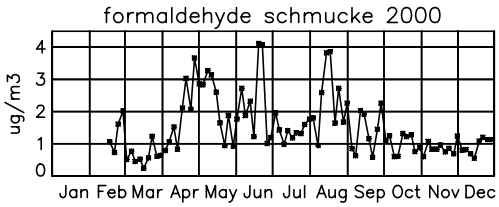
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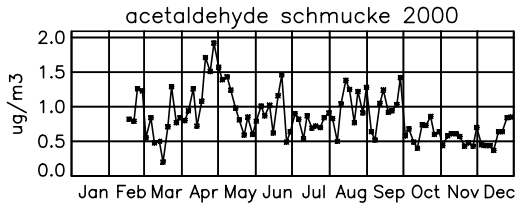
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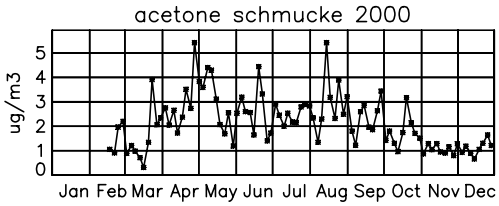
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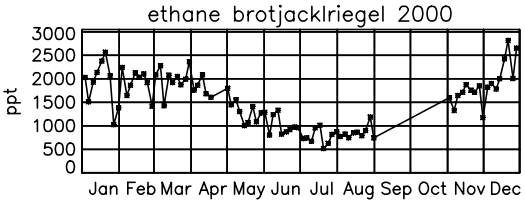
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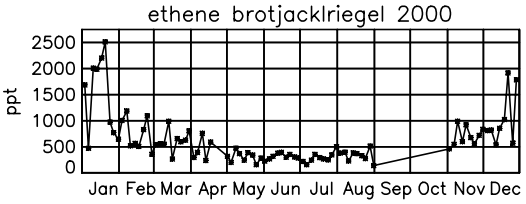
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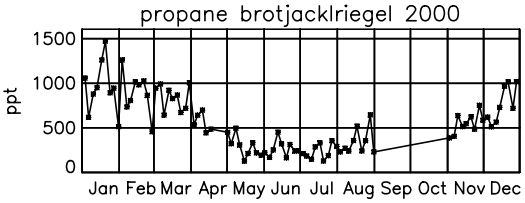
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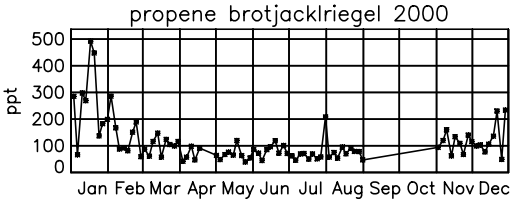
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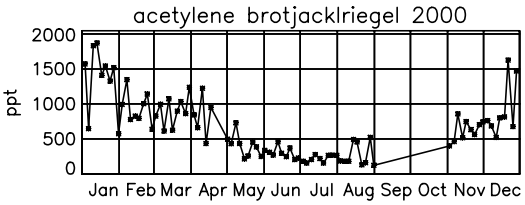
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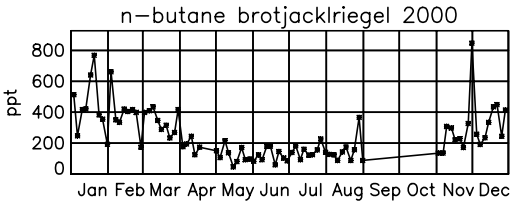
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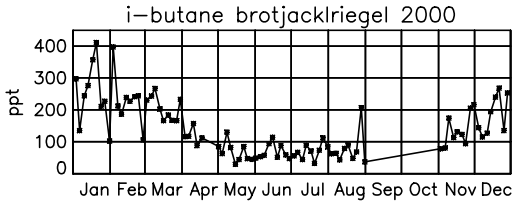
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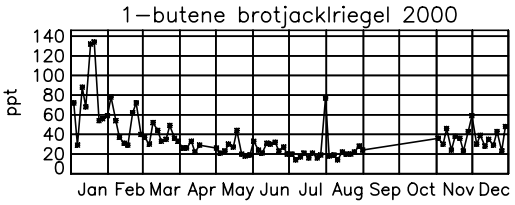
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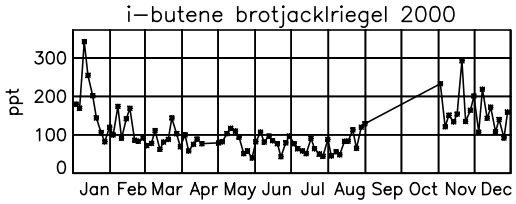
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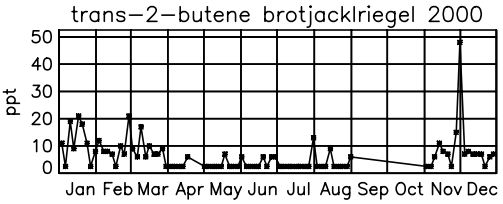
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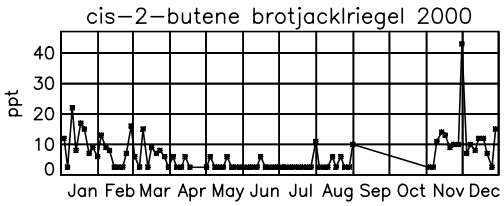
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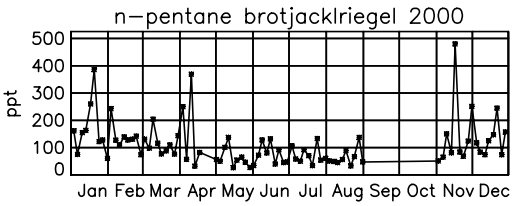
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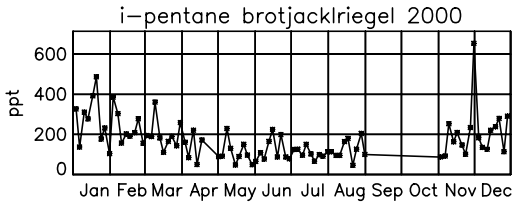
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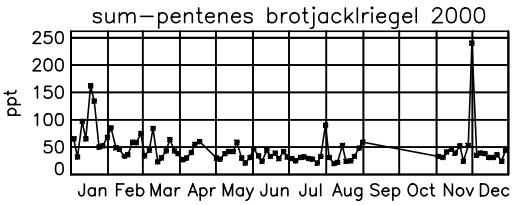
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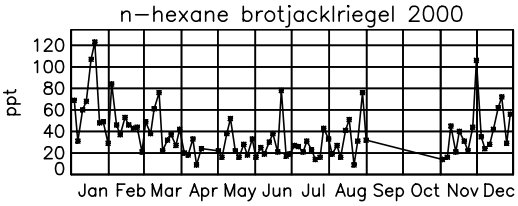
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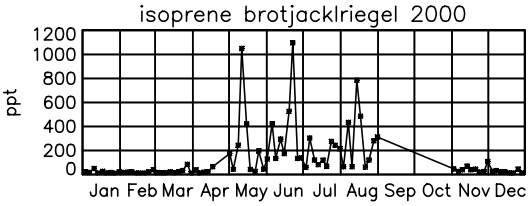
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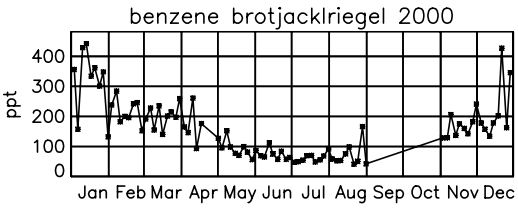
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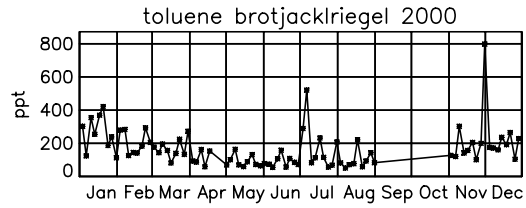
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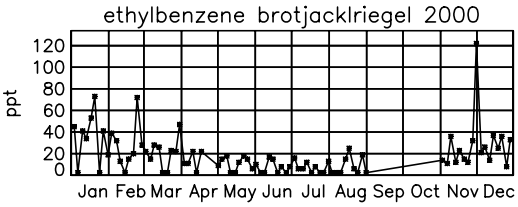
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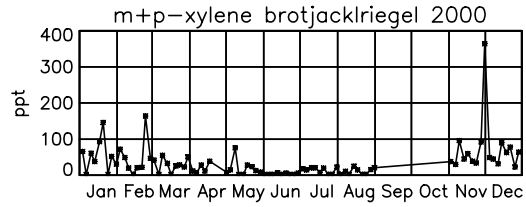
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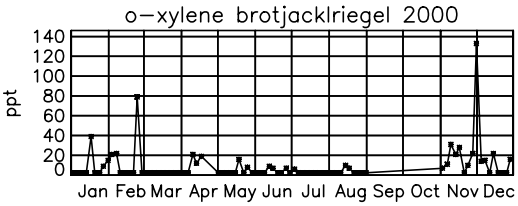
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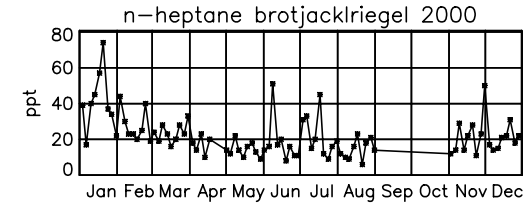
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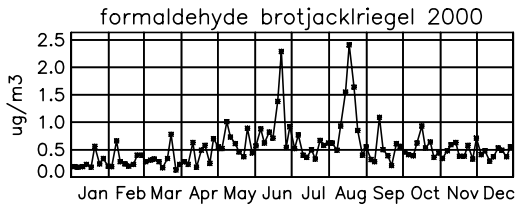
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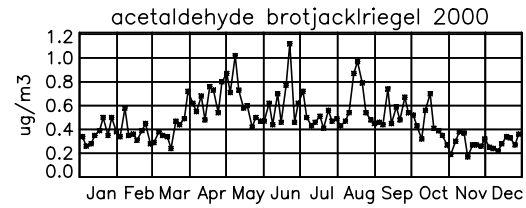
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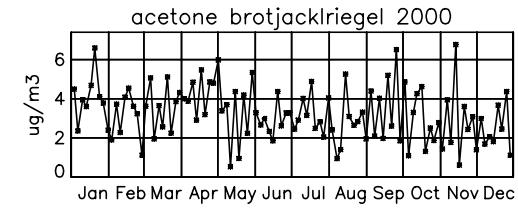
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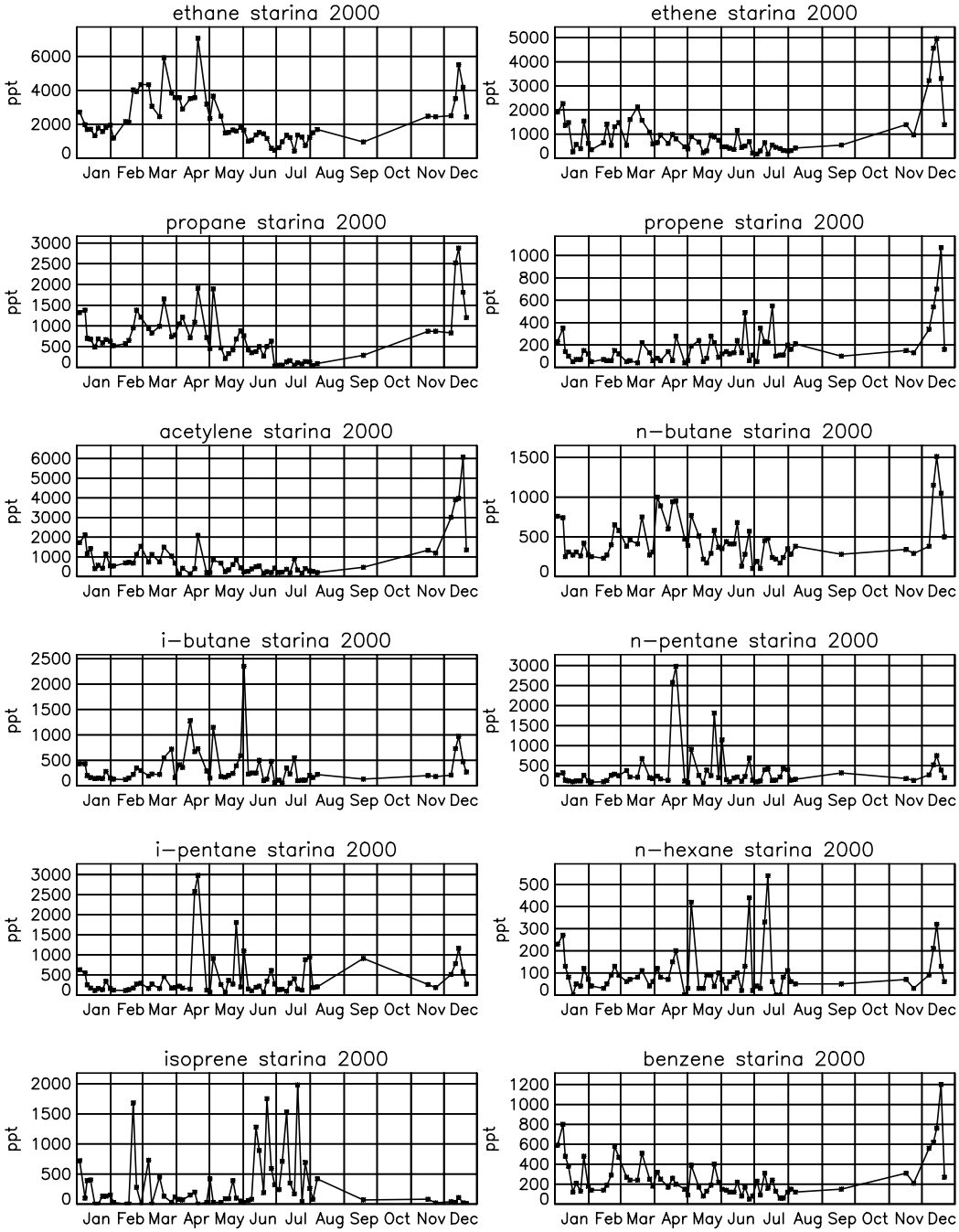
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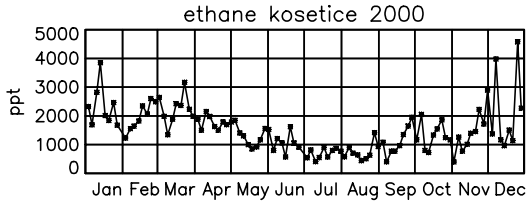
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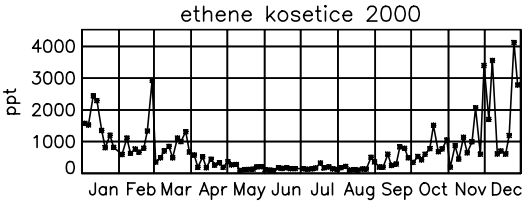
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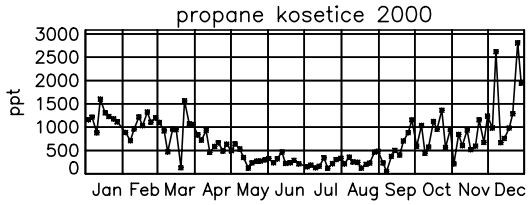
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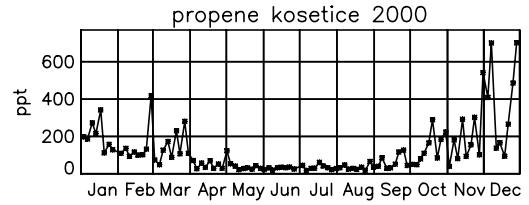
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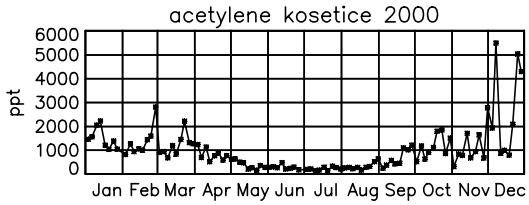
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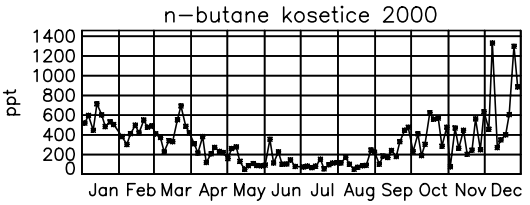
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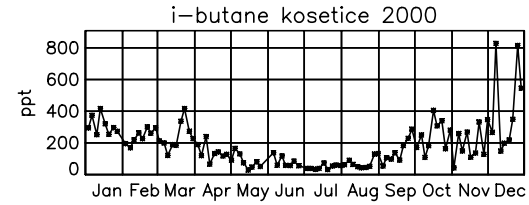
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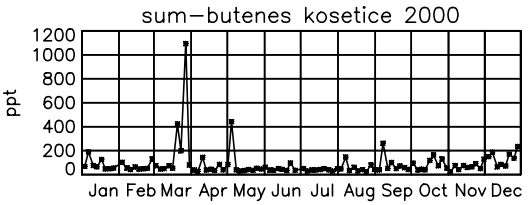
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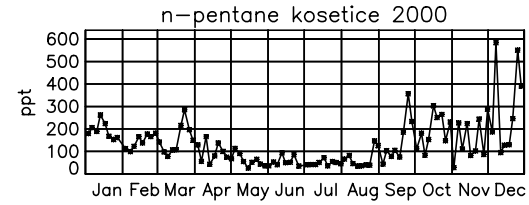
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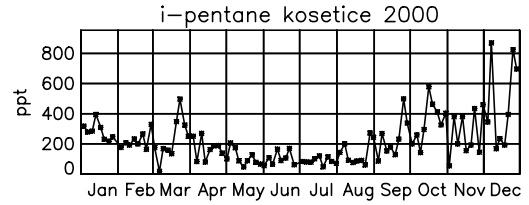
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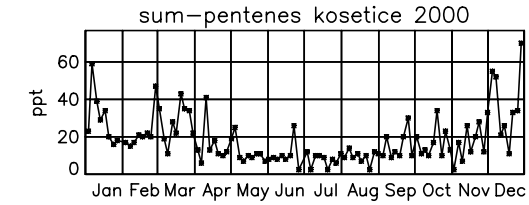
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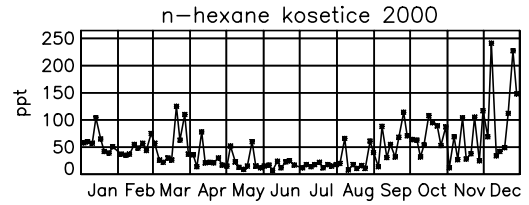
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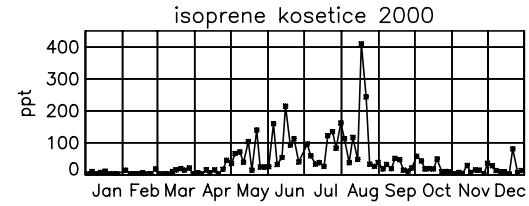
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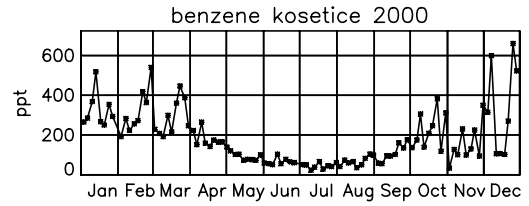
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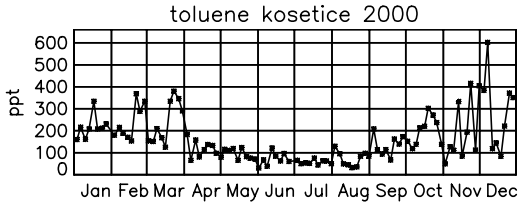
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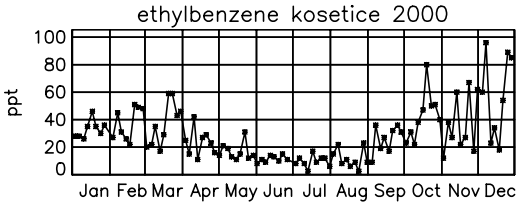
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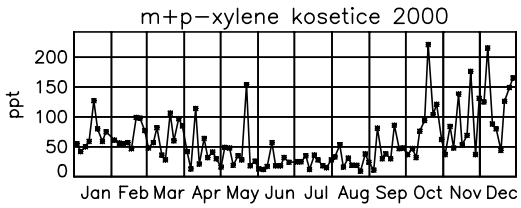
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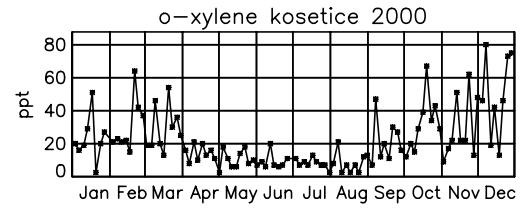
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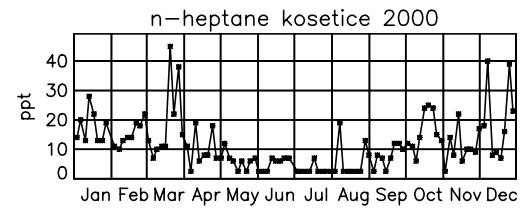
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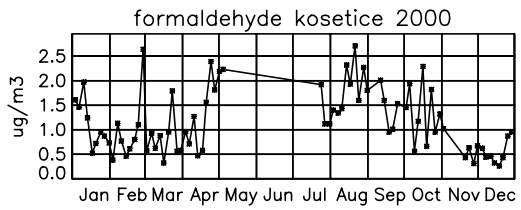
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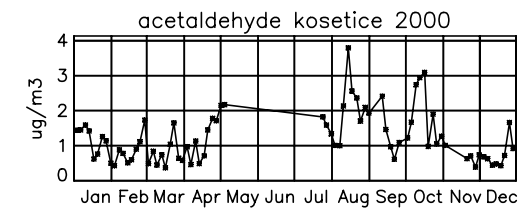
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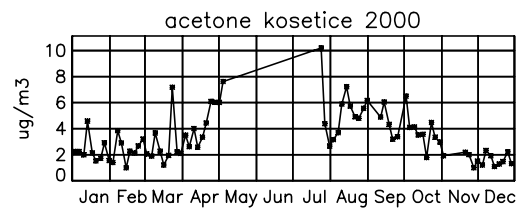
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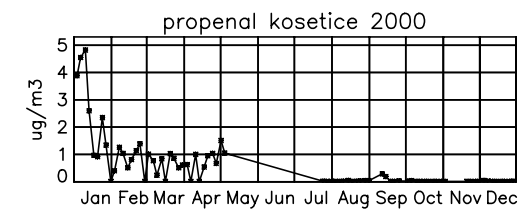
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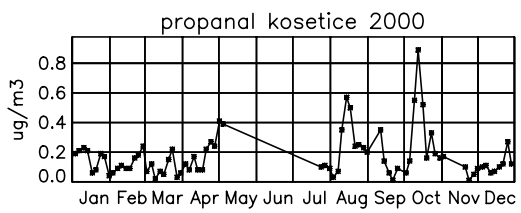
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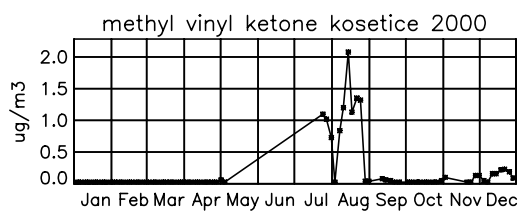
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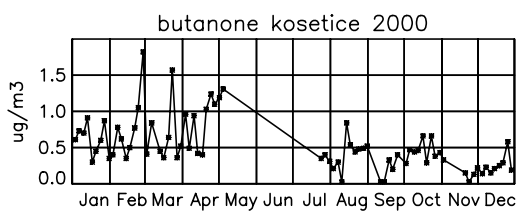
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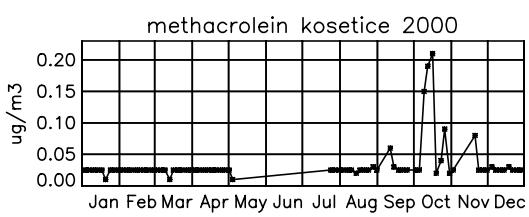
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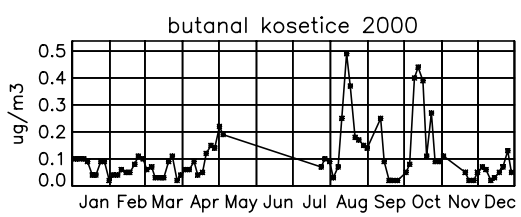
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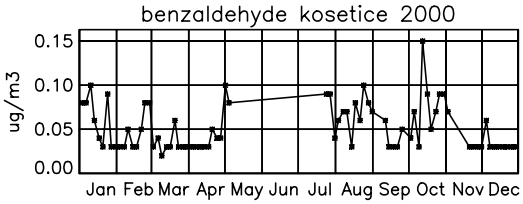
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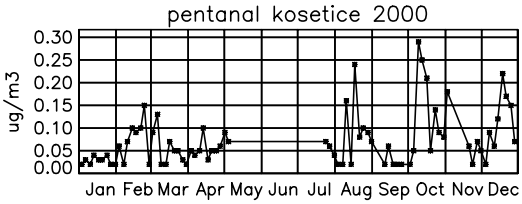
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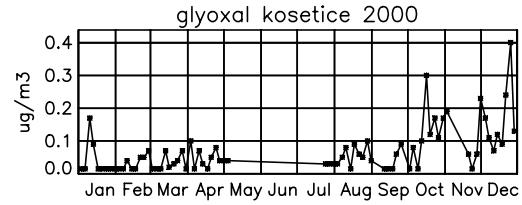
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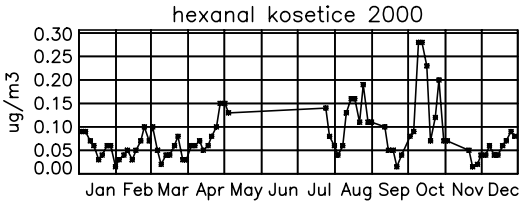
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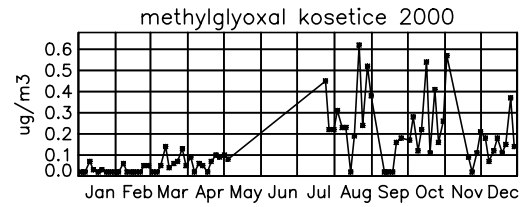
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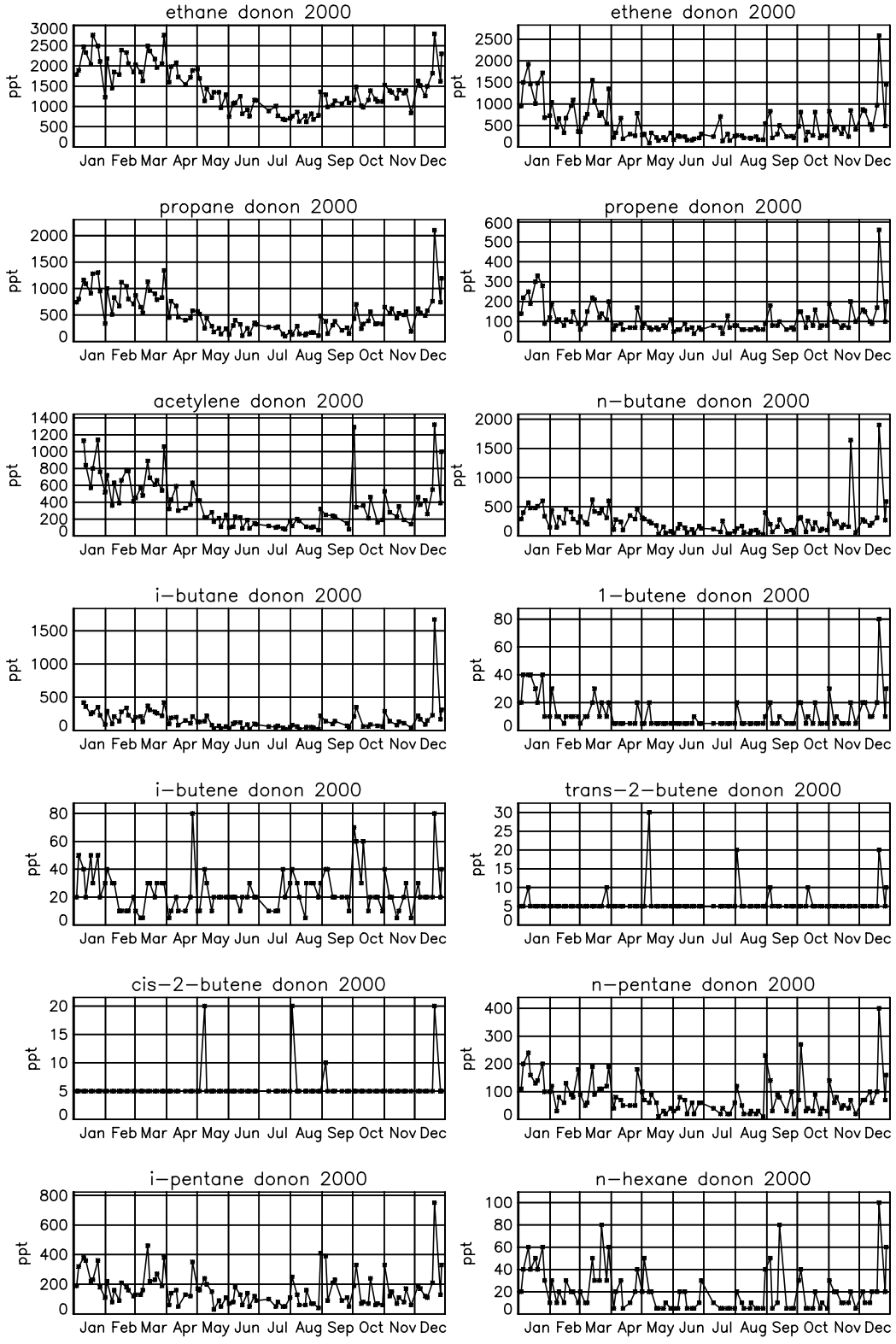
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propene donon 2000
acetylene donon 2000
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i−butene donon 2000
trans−2−butene donon 2000
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ppt
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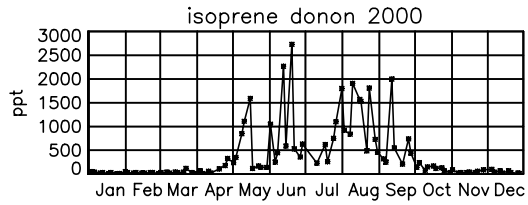
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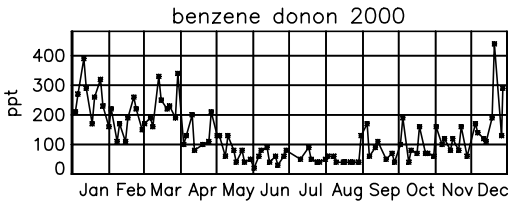
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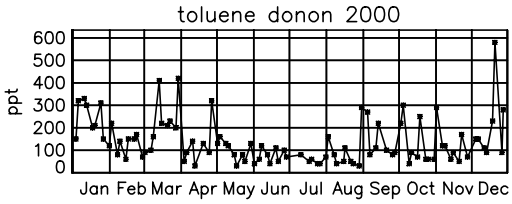
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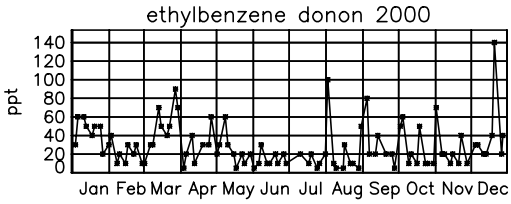
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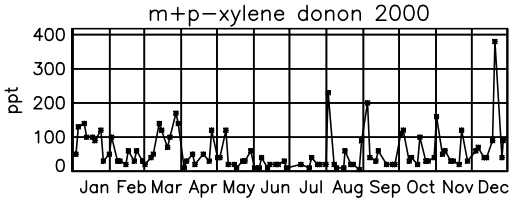
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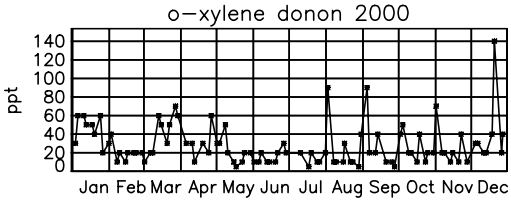
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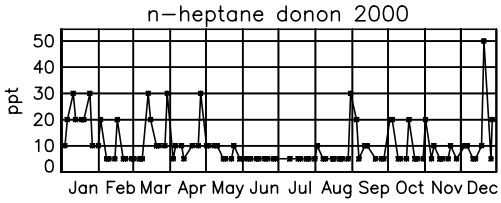
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ppt
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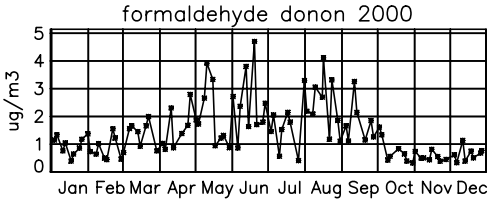
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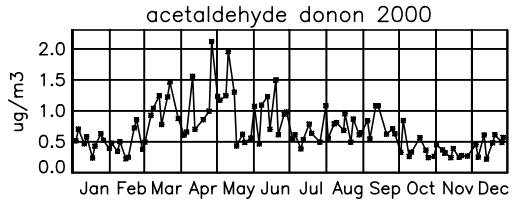
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0.5
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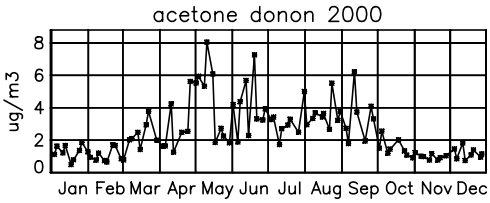
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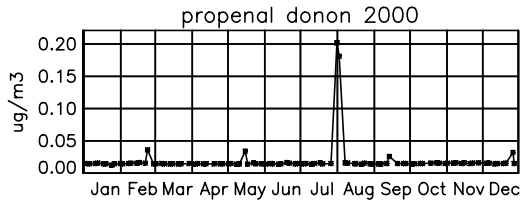
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0.10
0.05
0.00
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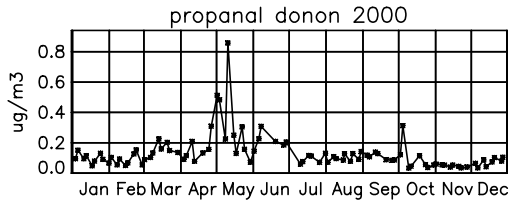
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ug/m3
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0.6
0.4
0.2
0.0
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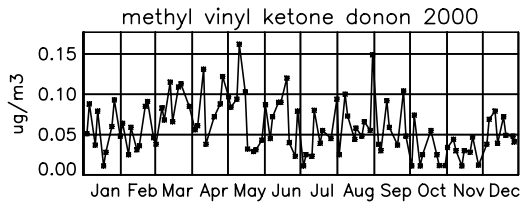
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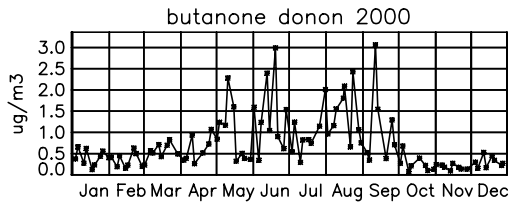
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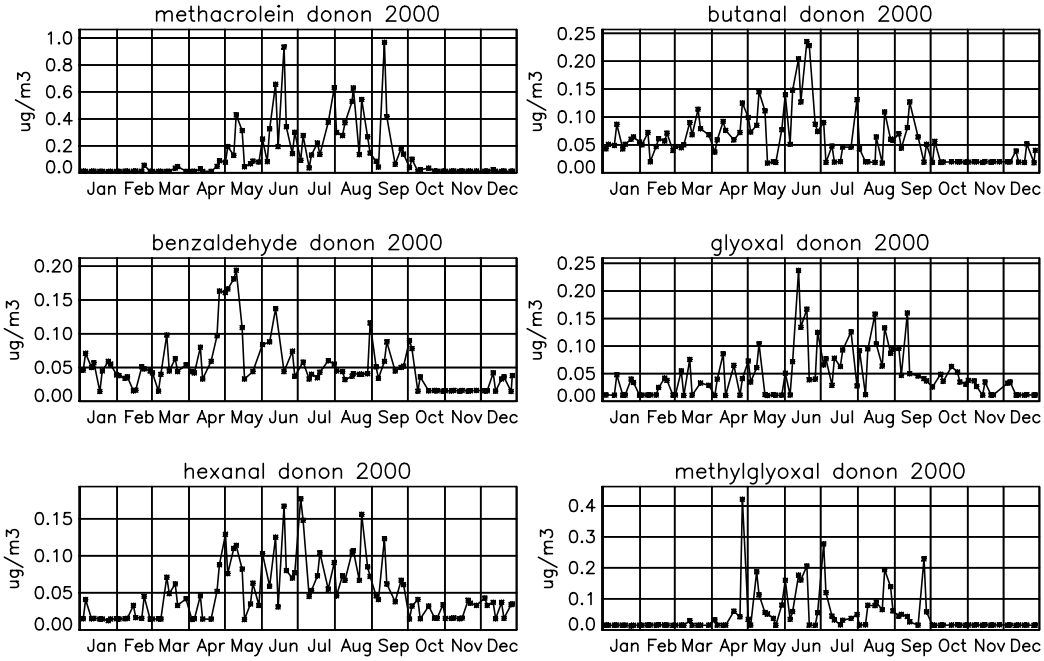
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butanal donon 2000
ug/m3
0.25
0.20
0.15
0.10
0.05
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
benzaldehyde donon 2000
ug/m3
0.20
0.15
0.10
0.05
0.00
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glyoxal donon 2000
ug/m3
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0.10
0.05
0.00
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hexanal donon 2000
ug/m3
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0.05
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
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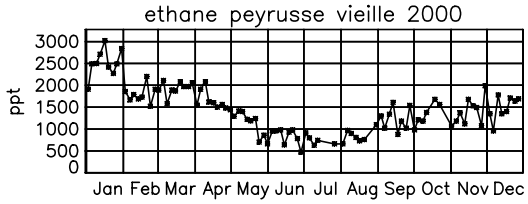
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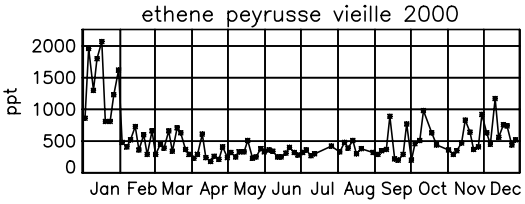
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ppt
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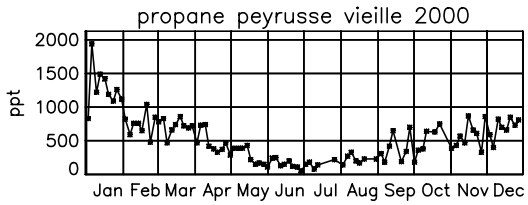
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ppt
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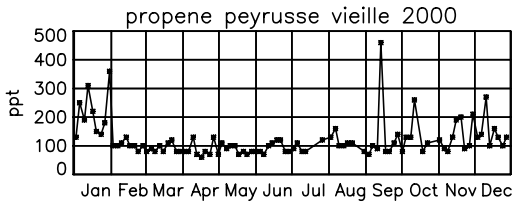
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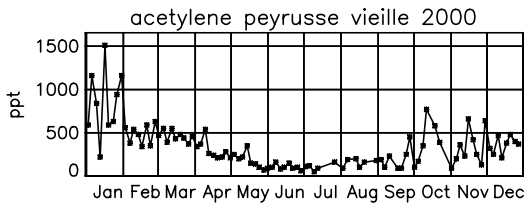
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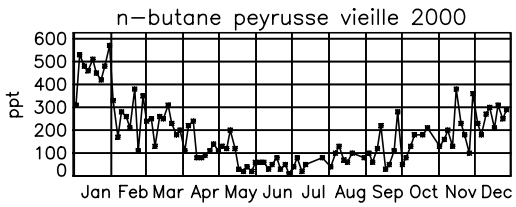
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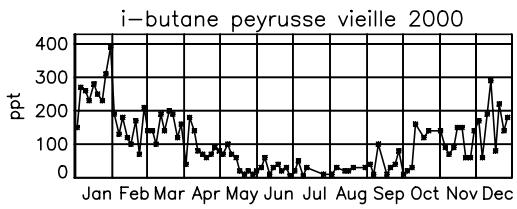
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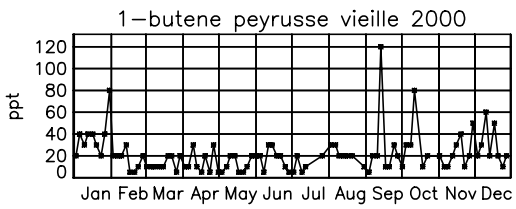
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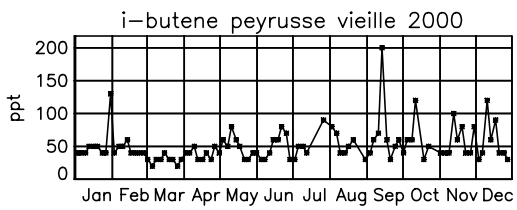
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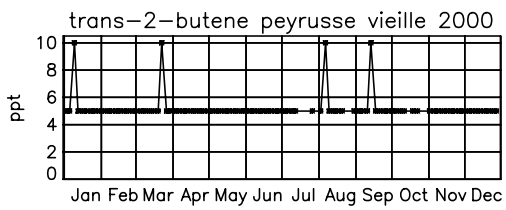
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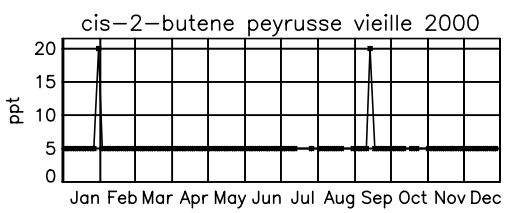
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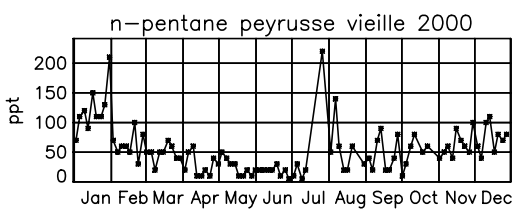
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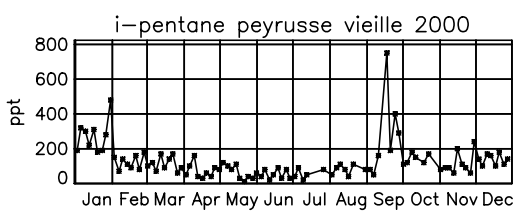
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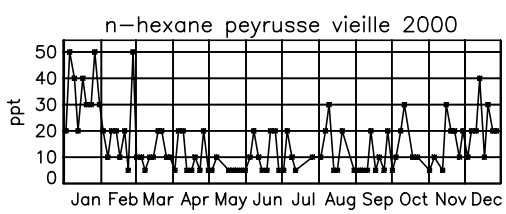
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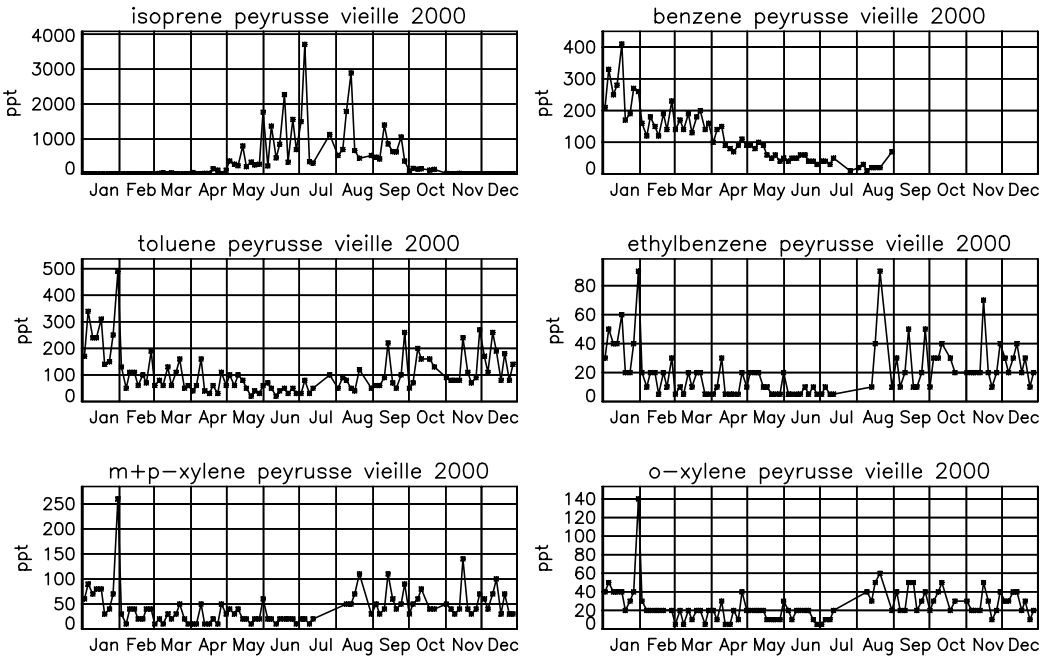
isoprene peyrusse vieille 2000
benzene peyrusse vieille 2000
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ppt
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec