

EMEP

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

VOC measurements 2011

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Summary

This report presents measurements of VOC carried out during 2011 at EMEP monitoring sites. VOC measurements are reported for 11 sites, and two of these with carbonyls. Except for two sites with continuous monitoring of hydrocarbons (Rigi and Hohenpeissenberg) all the VOC measurements are made by grab samples of light hydrocarbons in canisters and by 8-hours samples of carbonyls by DNPH adsorption tubes.

Compared to the previous years of monitoring, elevated and more variable levels of VOC in summer were seen at several of the German sites, particularly noticeable for ethene, ethyne, n-butane and toluene. The winter median concentration levels were also highest at sites in Germany. The elevated levels at these German sites as well as a number of seemingly unreasonably low levels of ethane at Košetice call for a closer investigation at the applied methods for sampling and analytical methods. In general, however, fairly uniform mean concentration levels of alkanes were seen, indicating that the VOCs become well mixed in the dark season without effective chemical loss mechanisms.

Comparison with EMEP model results for two French sites show fairly good agreement of isoprene given the large and small-scale variability in this compound. For the carbonyls a very good agreement is seen for formaldehyde, whereas the model underpredicted the levels of acetaldehyde and acetone, particularly in summer.

The long-term trends in hydrocarbons were estimated by applying a non-linear least squares fit to a seasonal trend equation. The results indicated downward trends of the order of 10-50 % for the decadal period 2001-2011 for several compounds/stations. The results for some of the German sites were an exception to this however, with marked increases in the levels of many compounds, apparently due to elevated levels the last few years.

VOC measurements 2011

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 Gothenburg protocol to Abate Acidification, Eutrophication and Ground-level Ozone was adopted by the Executive Body of UN-ECE, and on the 17th May 2005 the Protocol entered into force. 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. According to the Protocol, 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.

In 2012 a revised Gothenburg protocol was adopted. A main difference from the previous protocol is that the emission ceilings now are given as percentage reductions from 2005 to 2020 and thereafter. Furthermore, PM2.5 and BC (black carbon) is now included in the protocol. According to the revised protocol, the VOC emissions from the parties to the convention must be cut by 28 % as an average for all the parties in 2020 compared to the 2005 emissions, with national commitments ranging from 8 % (the Netherlands) to 54 % (Greece).

The EMEP VOC monitoring programme was initiated at the EMEP Workshop on Measurements of Hydrocarbons/VOC in Lindau, 1989 (EMEP/CCC, 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/CCC, 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/CCC, 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	
Aldehydes	Trimethylbenzenes	
	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 (CZ03), Waldhof (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 was in a range $\pm 25\%$ from the median. The exercise showed that the majority of the participating laboratories had the required analytical technique to correctly analyse a wide range of NMHC within an accuracy of $\pm 10\text{--}15\%$. 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 C₂–C₅ hydrocarbons).

In the EU FP5 project AMOHA (Accurate Measurements of Hydrocarbons in the Atmosphere) a large number of laboratories in Europe participated in parallel sampling and analyses of hydrocarbons in ambient air (Slemer et al., 2002). A major part of the project was to organize four annual intercomparisons starting in 1997 and ending in 2000. The results showed that except for a few laboratories the agreement was within $\pm 25\%$ of the median for the lighter alkanes. For some aromatics and unsaturated hydrocarbons as well as the C₆-C₇ alkanes a large spread in the values were seen, indicating measurement difficulties with these compounds. The spread in the results were, however, much less for laboratories using a NPL standard for calibration (Aas et al., 2001). Thus, it may be concluded that a large part of the differences seen among the laboratories reflected the use of different calibration gases. When using the same NPL standard the results from this intercomparison were very satisfactory.

The EMEP VOC 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, 2007). An EMEP expert meeting on VOC measurements was organised in Berlin, 1994 (EMEP/CCC, 1995a), 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; Borbon et al., 2004; Hakola et al., 2006).

An initiative has been taken to increase the cooperation and exchange of VOC data between GAW (Global Atmospheric Watch) and EMEP. At the EMEP TFMM workshop in Oslo in November 2004, on the implementation of the EMEP monitoring strategy, a closer harmonisation between the VOC monitoring in EMEP and GAW was discussed. Minutes and conclusions from the workshop are given elsewhere (EMEP/CCC, 2005). Harmonisation of data quality objectives (DQOs) and using a common audit questionnaire were recommended, and it is also a wish to arrange common GAW/EMEP training course and to further increase the exchange of VOC monitoring data between EMEP, GAW and WDCGG (World Data Centre of Greenhouse Gases).

In 2006 a WMO/GAW workshop on global measurements of VOCs (WMO, 2007) proposed a list of species to be measured based on current and future possibilities and needs of GAW. The proposed species are: ethane, propane, acetylene, isoprene, formaldehyde, terpenes, acetonitrile, methanol, ethanol, acetone, DMS, benzene, toluene, iso- and n-butane, iso- and n-pentane. Most of these compounds are already part of the EMEP VOC programme with some exceptions. The alcohols (methanol and ethanol) are likely to become more important in the future due to increased use of biofuels in vehicles. Furthermore, terpenes are important as precursors for secondary organic aerosols. These compounds would be of interest to include in the EMEP monitoring as well, but require other sampling methods and instrumentations than presently used for the hydrocarbons and carbonyls.

In the new EMEP Monitoring Strategy for 2010-2019 (ECE/EB.AIR/GE.1/2009/15), which hydrocarbons and carbonyls to measure have not been specified, but it is clearly stated that it is necessary to harmonise with the WMO GAW programme.

In April 2011 the project ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network) was initiated. This is a European Project (EC 7th Framework Programme) aiming at integrating European ground-based stations equipped with advanced atmospheric probing instrumentation for aerosols, clouds, and short-lived gas-phase species. ACTRIS will have the essential role to support building of new knowledge as well as policy issues on climate change, air quality, and long-range transport of pollutants. WP4 in ACTRIS is dedicated to the measurements of VOCs and nitrogen oxides. The aim is to integrate and harmonise such trace gas measurements in Europe. The development of standardised measurement protocols (SOPs) and common European calibration scales for VOCs is a central issue. Furthermore, ACTRIS will foster the dissemination of the methods and quality assured data to scientific groups related to the analysis and modelling of air pollutants in Europe and to support EC directives relevant for air pollution and the CLRTAP abatement strategies.

2. Status of the measurement programme in 2011

2.1 The station network

The location of the monitoring sites for VOC presented in this report is shown in Figure 1 and an overview of the measurement programme and the responsible laboratories in 2011 is given in Table 2. Data from 11 measurement sites are presented in this year's report and only two of them with carbonyl data, both of them in France.

VOC data from a number of other sites were received but are not presented here either because the data were too sparse and scattered in time, or because the values indicated significant local influences or because the data were considered to be at a preliminary and unsettled level qualitatively.

Table 3 gives the number of valid (daily) samples of hydrocarbons and carbonyls (after inspection and removal of outliers). According to EMEP's recommenda-

tions, the samples should be taken at least twice a week, implying that 104 samples per year correspond to 100% data cover.

A 90% data completeness, i.e. 94 samples per year, of daily values is given as data quality objective according to the EMEP manual (EMEP/CCC, 1995b). The data capture for hydrocarbons was lower than this for many sites in 2011. Hohenpeissenberg and Rigi have continuous sampling and thus a much higher data capture than the other sites. Carbonyls are only measured once per week in France giving a data capture of the order of 50%.

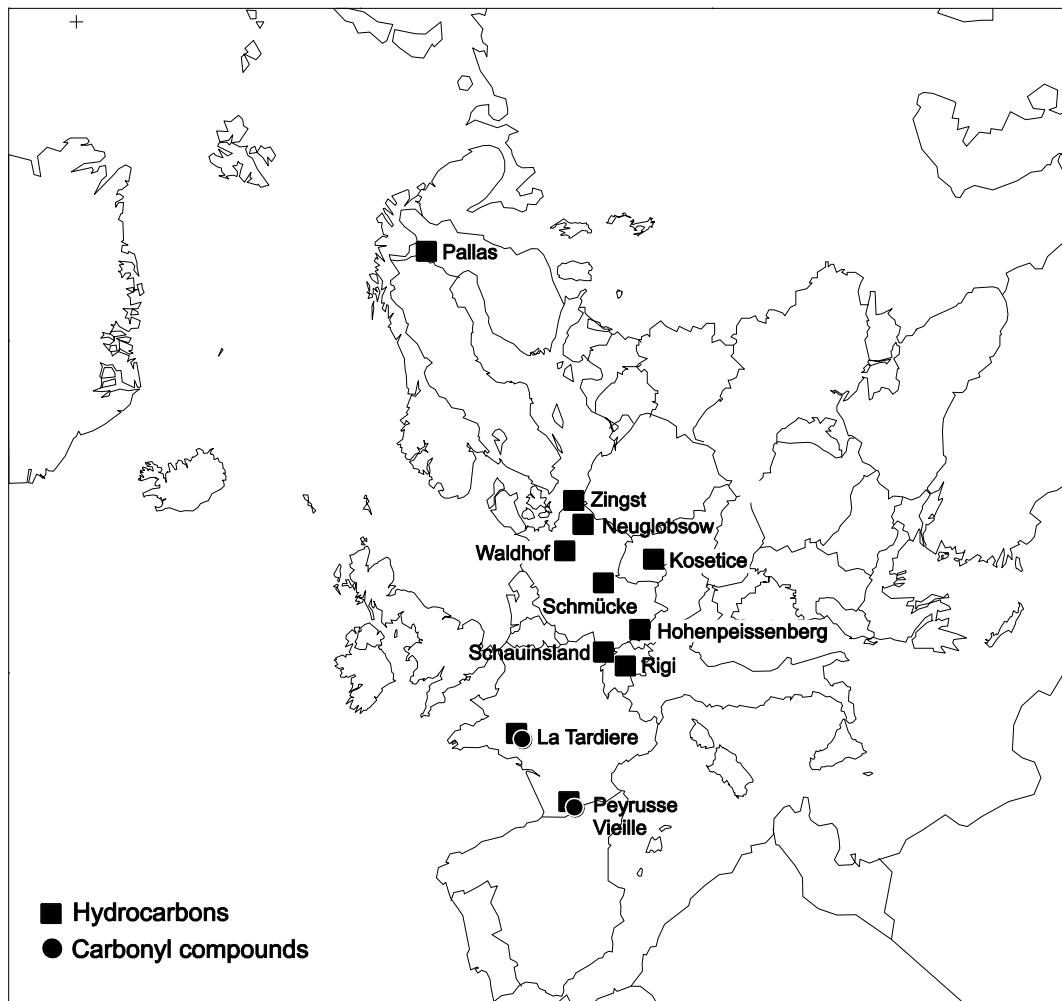


Figure 1: Monitoring sites for VOC in 2011.

Table 2: VOC monitoring sites with data for 2011 included in this report. 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.

Station	Code	HC ¹⁾	Lab. ²⁾	Carb ¹⁾	Comment
Pallas	FI96	Reg.	FMI	-	flask samples 2/week
Waldhof	DE02	Reg.	UBA	-	"
Schauinsland	DE03	Reg.	UBA	-	"
Neuglobsow	DE07	Reg.	UBA	-	"
Schmücke	DE08	Reg.	UBA	-	"
Zingst	DE09	Reg.	UBA	-	"
Hohenpeissenberg	DE43	Daily	DWD	-	2/day (noon, midnight) Jan-Aug
Košetice	CZ03	Reg.	CHMI	-	flask samples 2/week
Rigi	CH05	Cont.	EMPA	-	hourly data
Peyrusse Vieille	FR13	Reg.	EMD	Reg.	flask samples NMHC 2/week
La Tardière	FR15	Reg.	EMD	Reg.	DNPH samples OVOC 1/week
					flask samples NMHC 2/week
					DNPH samples OVOC 1/week

1) Reg. = regularly, Scat. = scattered, n.m. = not measured., n.a. = not yet analysed, cont. = Continuous

2) CHMI = Czech Hydrometeorological Institute

DWD = Deutscher Wetterdienst

EMD = Ecole des Mines de Douai (France)

EMPA = Swiss Federal Lab. for Materials Testing and Research

FMI = Finnish Meteorological Institute

UBA = Umweltbundesamt (Germany)

Table 3: The number of valid samples of hydrocarbons (HC) and carbonyls (Carb) in 2011.

Station	Number of samples HC ²⁾	Number of samples Carb ³⁾
Pallas	71	-
Waldhof	92	-
Schauinsland	85	-
Neuglobsow	89	-
Schmücke	91	-
Zingst	94	-
Hohenpeissenberg ¹⁾	234	-
Košetice	102	-
Rigi ¹⁾	336	-
Peyrusse Vieille	64	49
La Tardière	102	48

¹⁾ Refers to days with monitoring data

²⁾ Refers to ethane (may differ for other HCs)

³⁾ Refers to formaldehyde (may differ for other carbonyls)

2.2 Analytical procedures and quality control

The procedures for sampling and chemical analyses in 2011 were similar to previous years, and are not discussed in this report. The technical procedures for the sampling and analysis of hydrocarbons by FMI at the Finnish station, as well as a site description and data interpretation, are given by Laurila and Hakola (1996). A presentation of the sampling and analyses performed by the laboratories at EMD (France), EMPA (Switzerland), CHMI (Czech Republic), MMA (Spain)

and UBA (Germany) has been given in previous annual reports and by Solberg et al. (1996) and is not repeated here. A new GC and new analytical methods were introduced by UBA for the German sites in 2006 leading to certain systematic changes. In general, the new method was more sensitive to C₇ and higher VOCs. The instrumentation and methods applied by DWD at Hohenpeissenberg have been successfully tested in two international intercomparison experiments (NOMHICE, AMOHA) and have been documented by Plass-Dülmer et al. (2002).

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, 1995b) 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 chemically stable in the prepared sample solution. Furthermore, LC/MS studies indicate possibilities of chromatographic interference in the C₄ carbonyl compound range. Thus, a revision of the monitoring procedures for these carbonyls is needed.

3. VOC concentrations in 2011

3.1 General

Monthly mean and median concentrations of the individual hydrocarbons and carbonyls for 2011 are tabulated in Appendix A. The monthly statistics were not calculated for sample numbers less than four. Time series of all compounds during 2011 are given in Appendix B. Note that the monthly means and medians in Appendix A were based on all data whereas the time series in Appendix B were based on daytime values for the sites with continuous monitoring (Rigi and Hohenpeissenberg). Based on previous experience there is not much difference in the anthropogenic HC concentrations at noon and at midnight at Hohenpeissenberg (pers. comm., Christian Plass-Dülmer). For isoprene the difference is substantial as this is a reactive biogenic compound, emitted during daytime, with low concentrations during night.

A comparison of the seasonal mean and percentile concentrations of hydrocarbons in winter (Jan., Feb., Nov., Dec.) and carbonyls in summer (May, Jun., Jul., Aug.) measured at the different stations is given in Figure 2 and Figure 3. The stations are arranged from north to south. Considering that the sites span a wide area from Southern Europe to the most northern part of the continent, the hydrocarbon winter mean levels are fairly uniform. In general, the lowest levels of the hydrocarbons are seen at Pallas in Finland, Schauinsland in Germany and La Tardière in France.

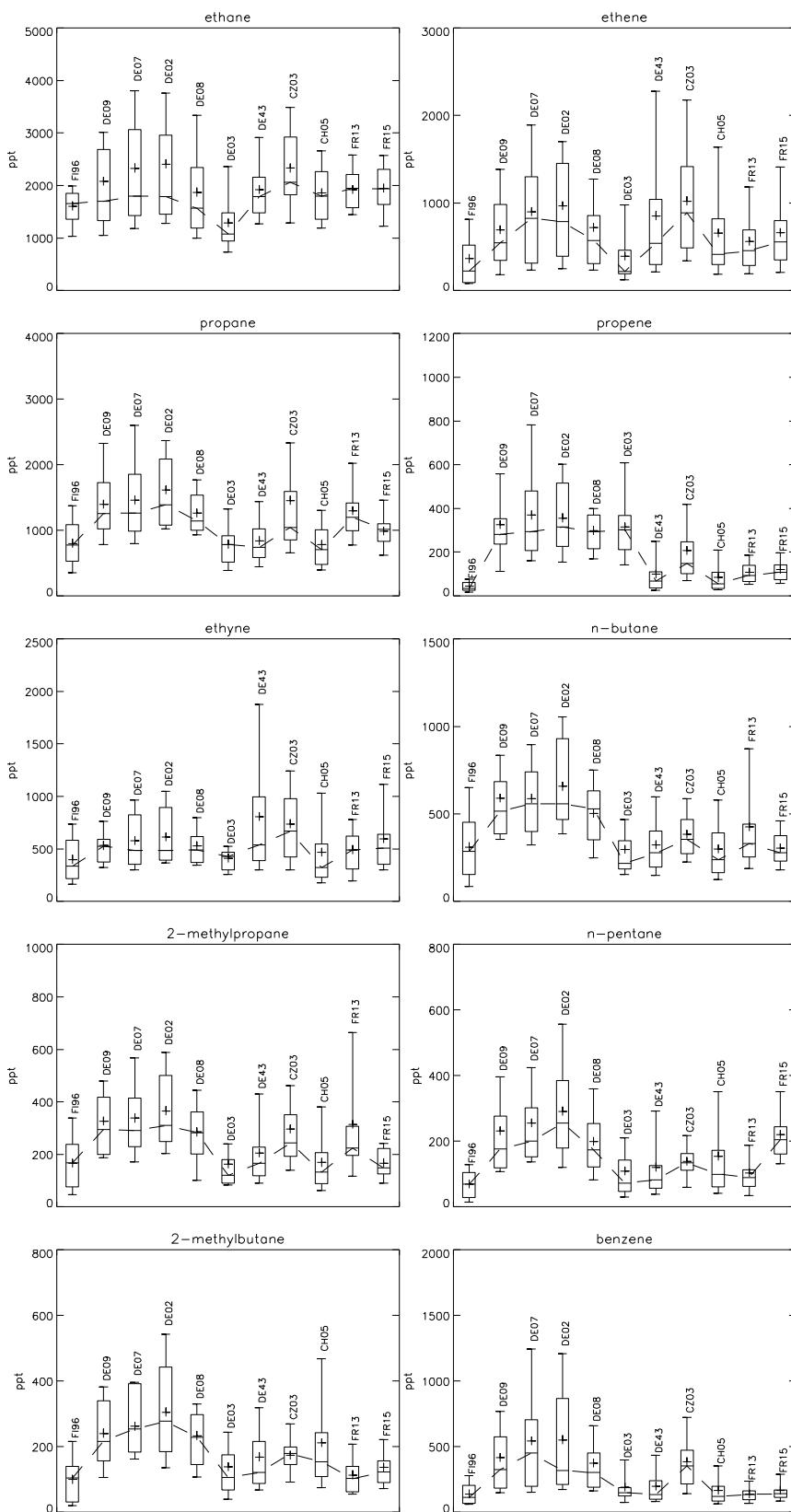


Figure 2: Box- and whisker-diagrams for hydrocarbons during winter 2011 (Jan., Feb., Nov., Dec.). The markers indicate the 10-, 25-, 50-, 75- and 90-percentiles. Mean values are indicated by a cross. The dashed line connects the median values for clarity.

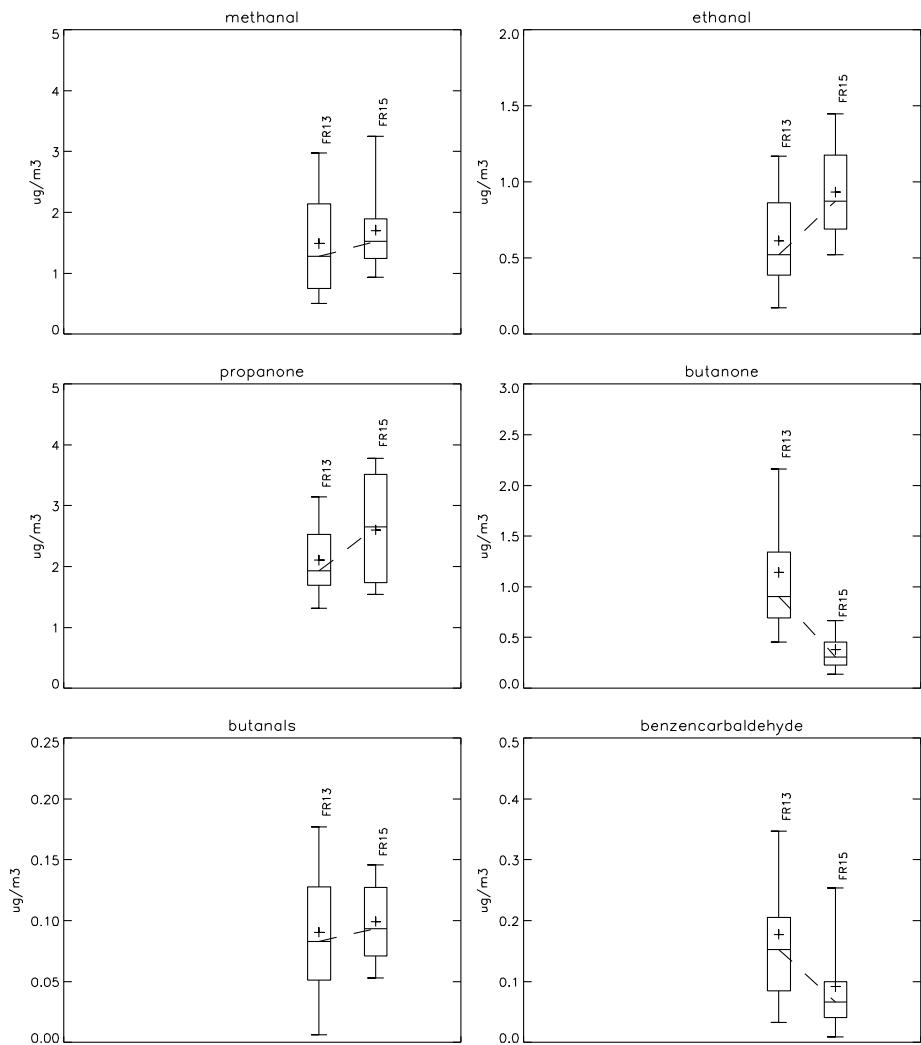


Figure 3: Box- and whisker-diagrams for carbonyls during summer 2011 (May, June, July, August). The markers indicate the 10-, 25-, 50-, 75- and 90-percentiles. Mean values are indicated by a cross.

Certain results from the monitoring in 2011 need to be mentioned. Compared to previous years, surprisingly high summer levels of many compounds were reported at some of the German sites (Zingst, Neuglobsow, Waldhof and Schmücke). This regards in particular ethene, ethyne, n-butane and toluene. The data reveal higher variability and generally higher levels than expected in summer for these compounds, thus masking the expected seasonal cycle which is normally seen (winter maximum and summer minimum). The reason behind this is not clear.

Furthermore, at Košetice (CZ03), very low levels of ethane were frequently reported in summer 2011 (i.e. < 500 ppt). Due to the long atmospheric lifetime ethane is considered to have a hemispherical background level, even in summer, in practice a lower threshold level varying through the year. Concentration levels below this threshold is not to be expected. In 2011 the concentration was frequently reported to be below 400 ppt at Košetice, even below 100 ppt at a few occasions. In previous years values below 400 ppt were very rarely seen. Ethane

concentrations in central Europe below 400 ppt and even below 100 ppt seems unrealistic. The very low ethane values reported at Košetice and the surprisingly high levels of several VOCs at Zingst, Neuglobsow, Waldhof and Schmücke (compared to other stations and previous years) calls for a special effort to investigate possible artefacts induced by the sampling or analytical methods.

3.2 Regional distribution of VOC

Figure 4—Figure 13 show maps with the stations' median concentrations of 10 light hydrocarbons for the winter months January, February, November and December in 2011 taken together. These medians are based on the average of the daytime values at Rigi and Hohenpeissenberg.

Although the number of sites obviously is too low to give a picture of the regional background distribution of hydrocarbons in Europe, some characteristics are indicated by these results. Similar figures for three carbonyls for the summer months May-August 2011 are given in Figure 14—Figure 16.

For some compounds the data show higher regional differences than experienced previously. The winter median concentrations of propene, n-butane and i-butane (2-methylpropane) were highest at the German sites.

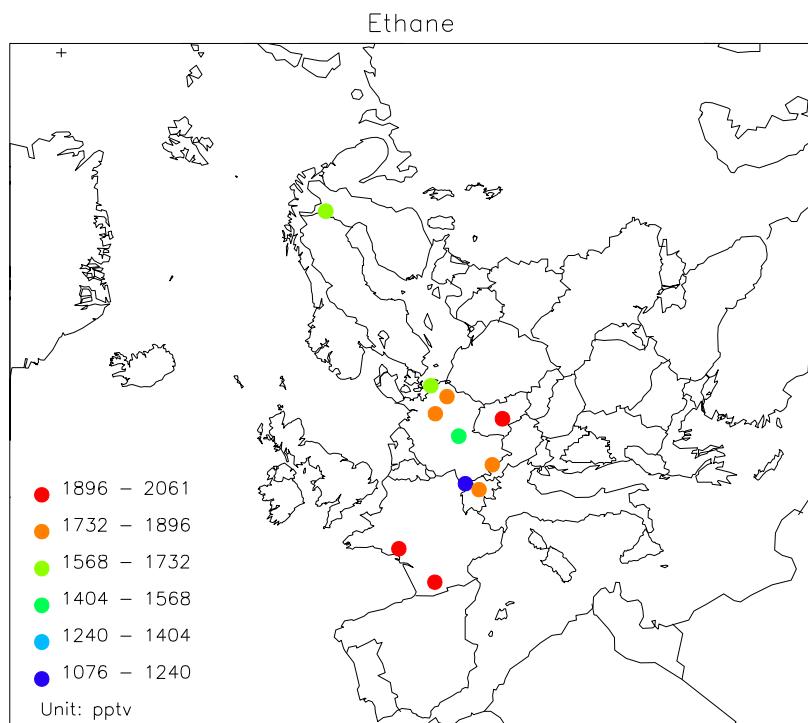


Figure 4: Median concentration of ethane at EMEP sites in the winter months November, December, January and February 2011 taken together.

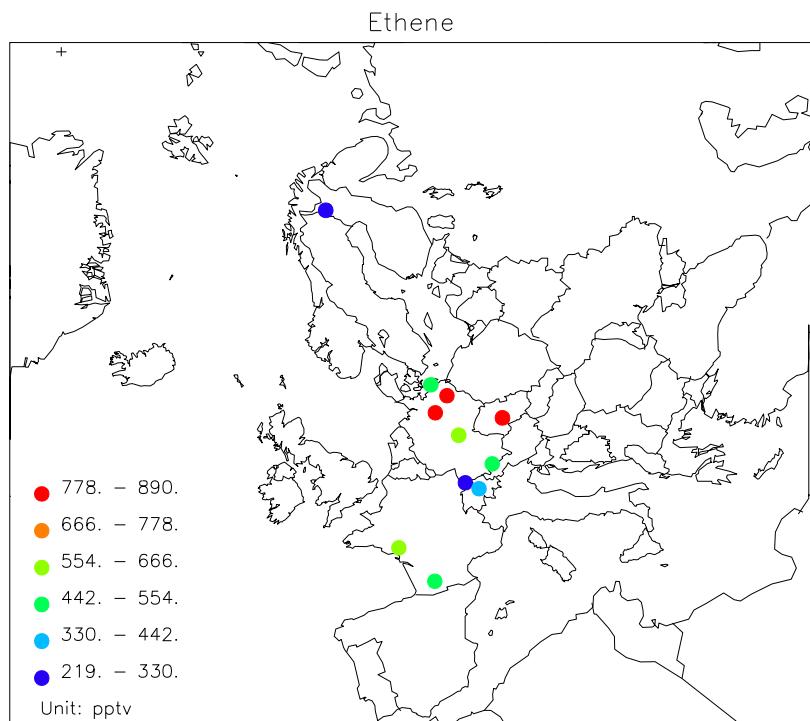


Figure 5: Median concentration of ethene at EMEP sites in the winter months November, December, January and February 2011 taken together.

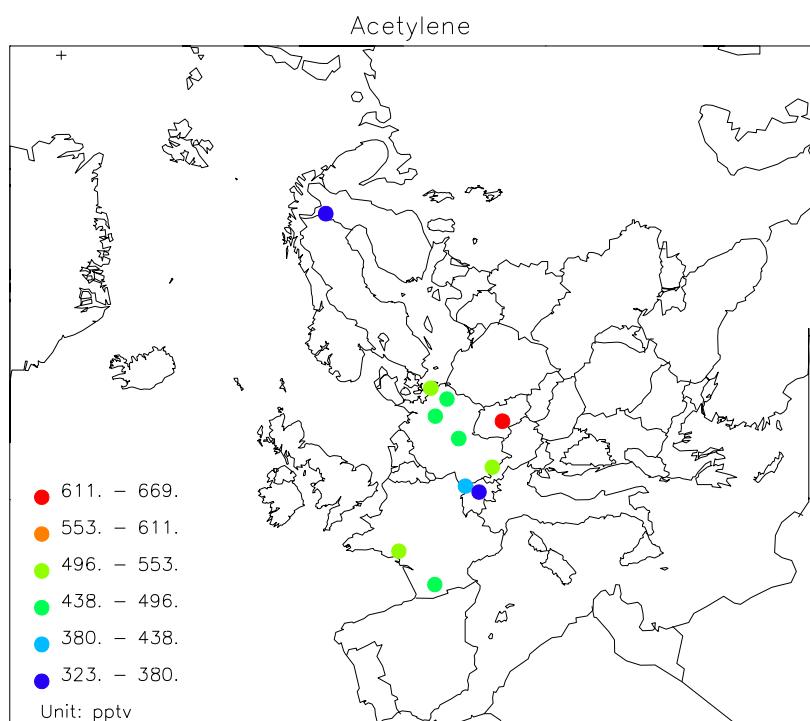


Figure 6: Median concentration of acetylene at EMEP sites in the winter months November, December, January and February 2011 taken together.

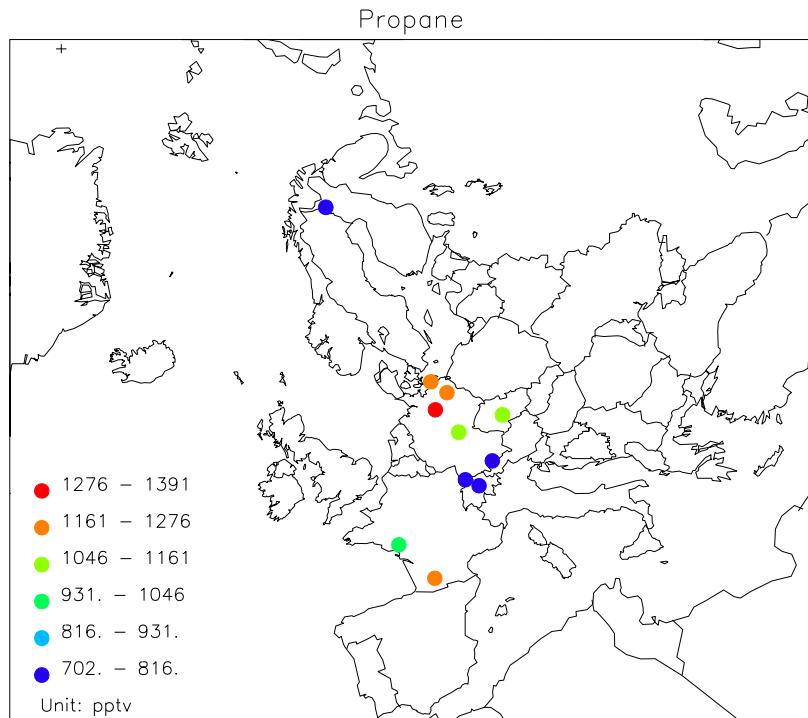


Figure 7: Median concentration of propane at EMEP sites in the winter months November, December, January and February 2011 taken together.

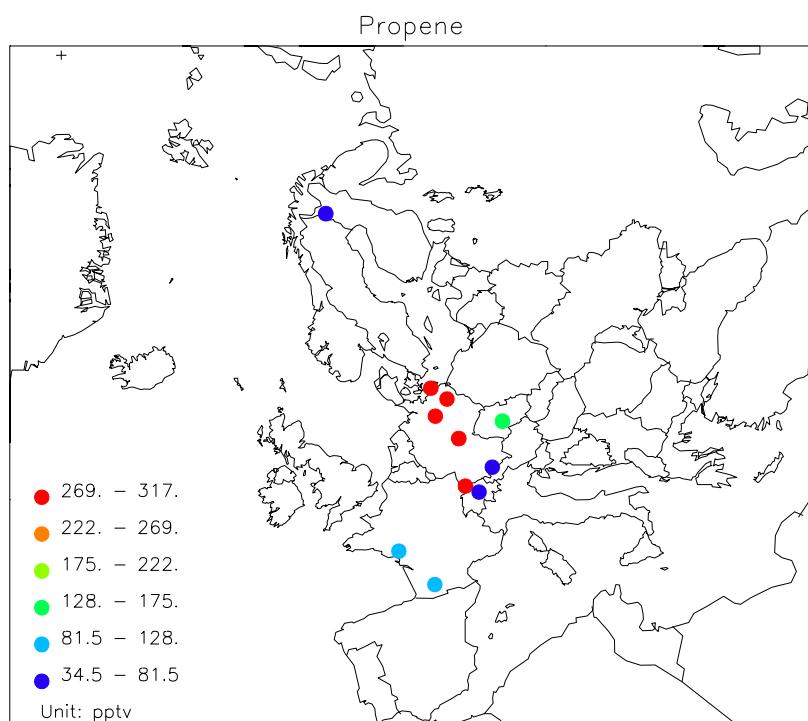


Figure 8: Median concentration of propene at EMEP sites in the winter months November, December, January and February 2011 taken together.

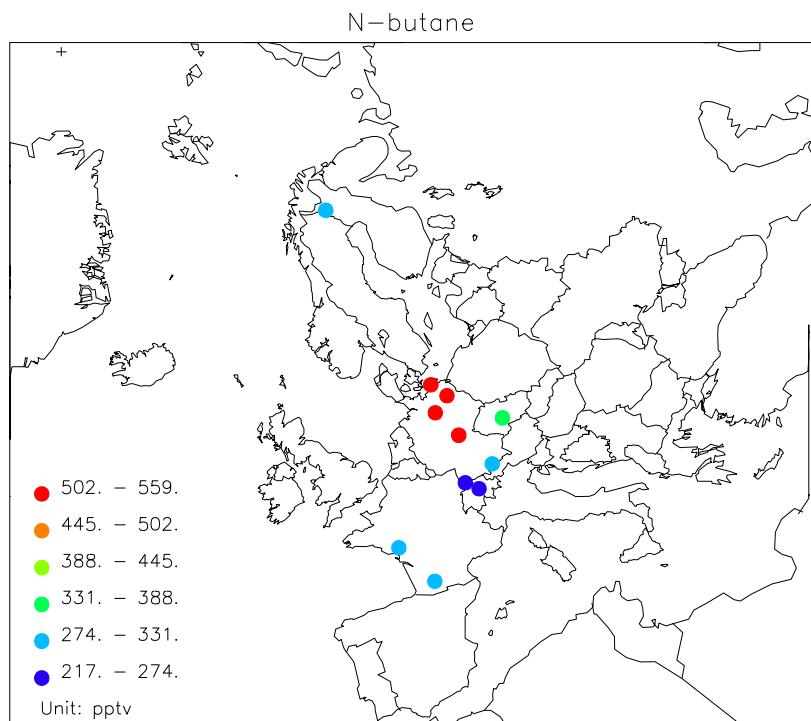


Figure 9: Median concentration of n-butane at EMEP sites in the winter months November, December, January and February 2011 taken together.

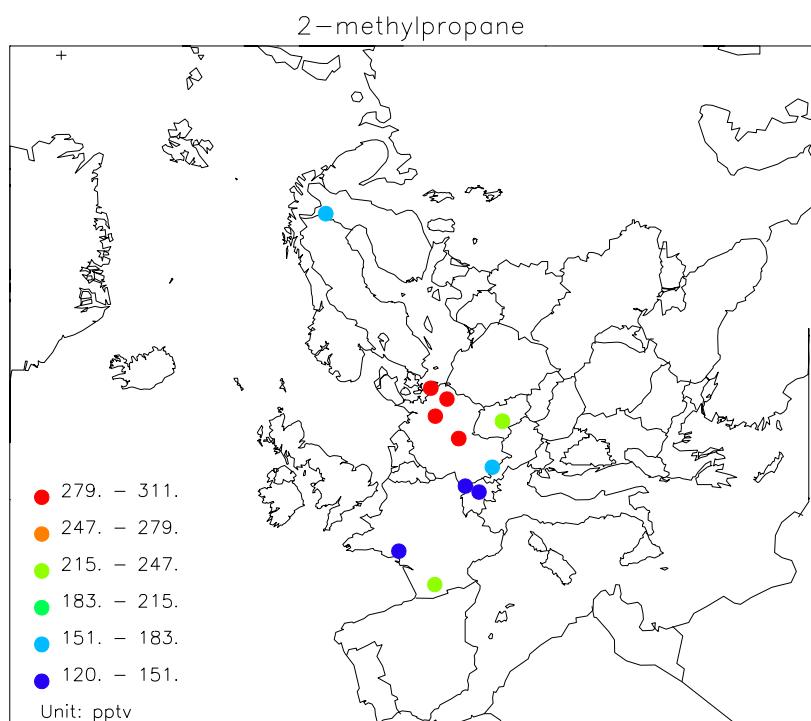


Figure 10: Median concentration of i-butane at EMEP sites in the winter months November, December, January and February 2011 taken together.

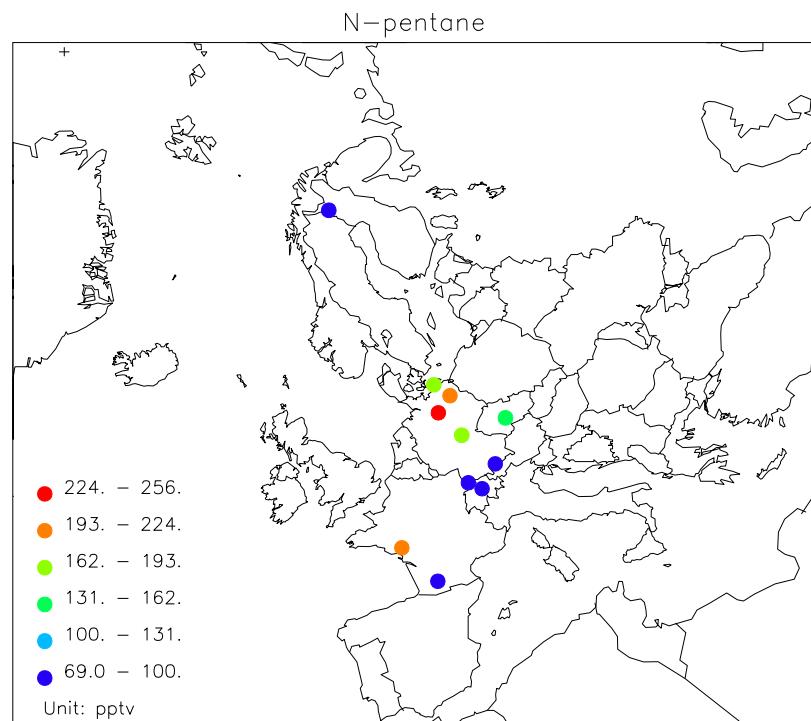


Figure 11: Median concentration of n-pentane at EMEP sites in the winter months November, December, January and February 2011 taken together.

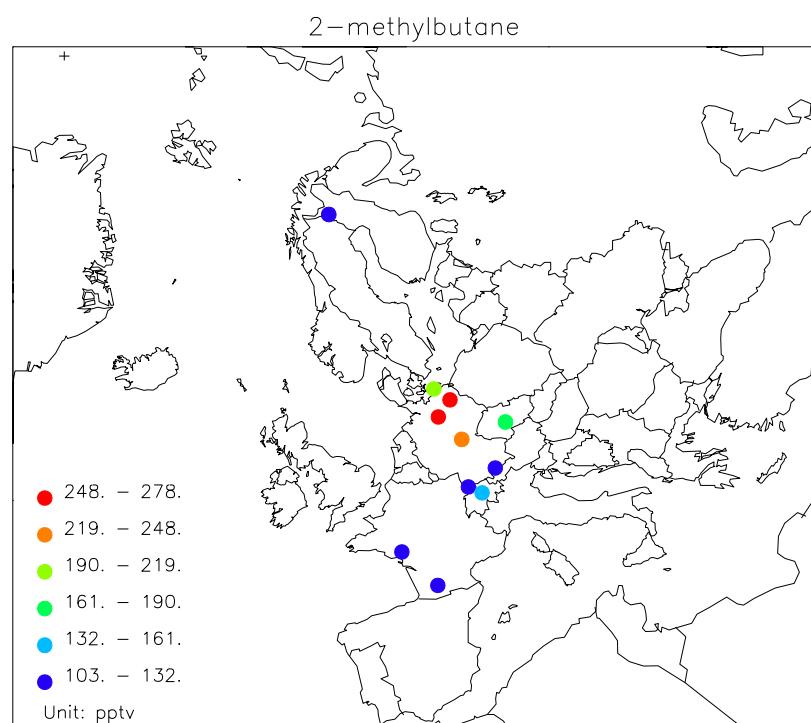


Figure 12: Median concentration of i-pentane at EMEP sites in the winter months November, December, January and February 2011 taken together.

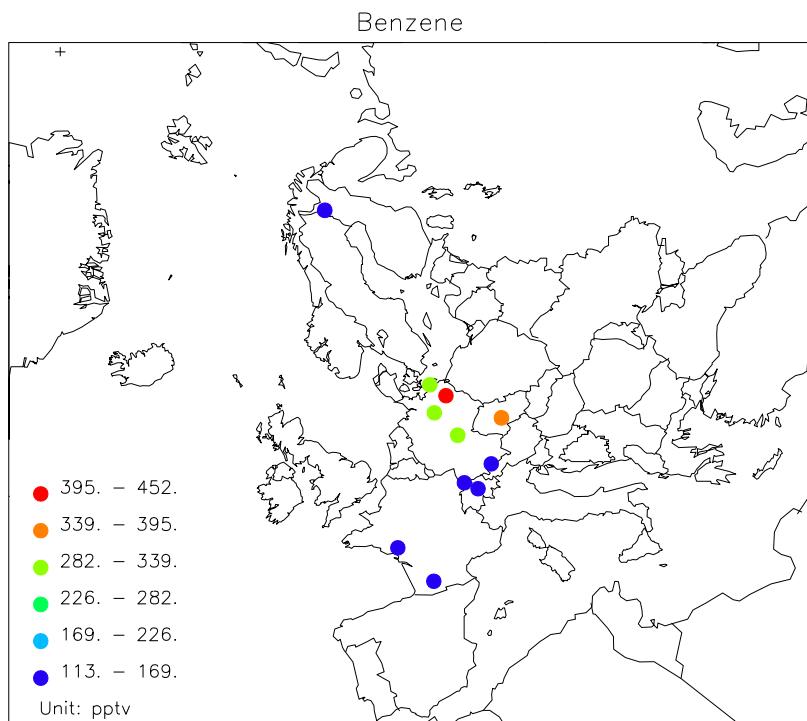


Figure 13: Median concentration of benzene at EMEP sites in the winter months November, December, January and February 2011 taken together.

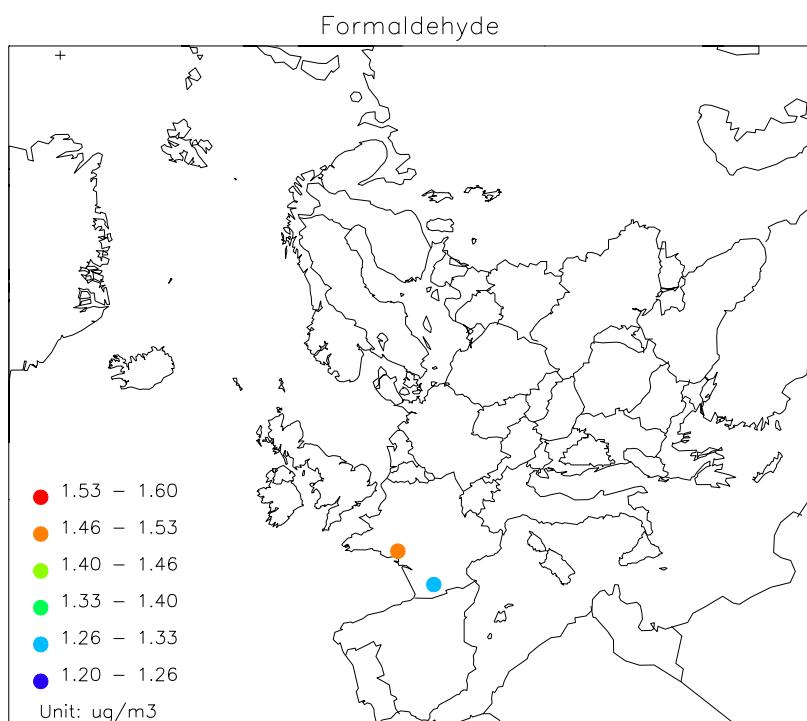


Figure 14: Median concentration of formaldehyde at EMEP sites in the summer months May, June, July and August 2011 taken together.

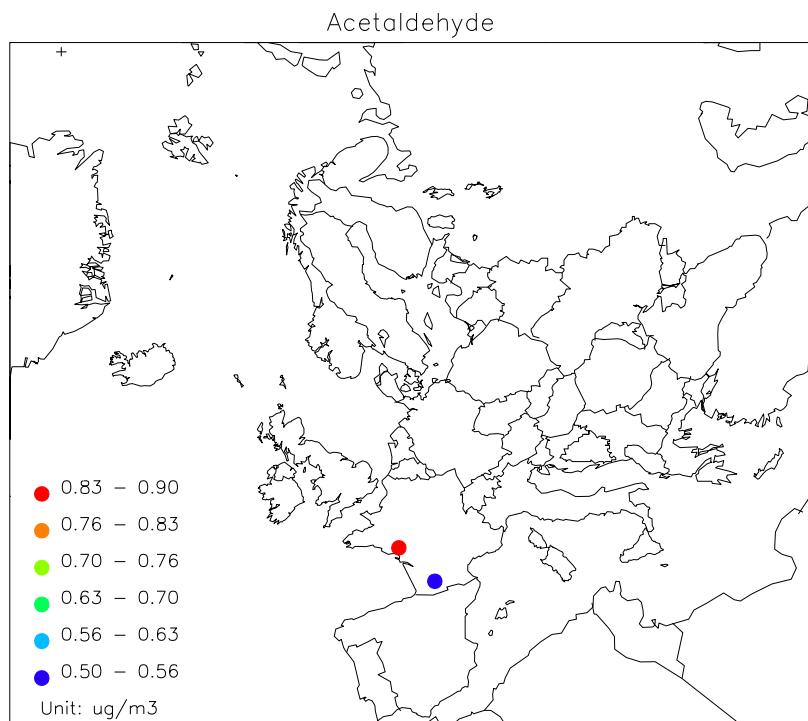


Figure 15: Median concentration of acetaldehyde at EMEP sites in the summer months May, June, July and August 2011 taken together.

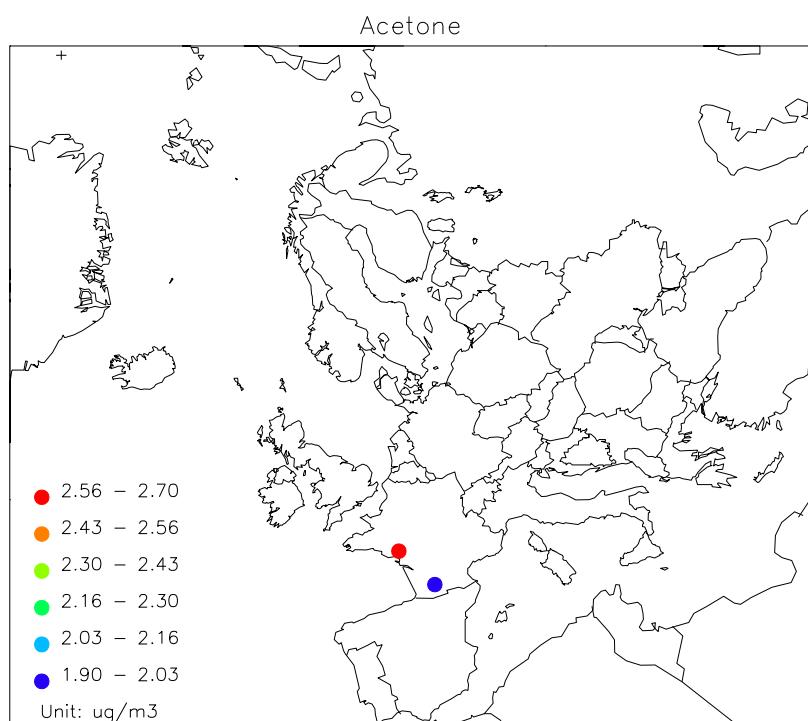


Figure 16: Median concentration of acetone at EMEP sites in the summer months May, June, July and August 2011 taken together.

4. Modelled VOCs

Preliminary results of a few modelled VOCs using the EMEP model with the CRI_v2_R5 chemistry (Simpson et al., 2012), together with observed values for 2011 are shown below. Results are presented for the two French sites only, as these are the only sites with carbonyl measurements. Due to the lumping of the anthropogenic hydrocarbons in the model's chemical scheme, they could not be compared directly with the observed individual species. Thus, we only show the results for the isoprene in addition to formaldehyde, acetaldehyde and acetone.

Due to the short atmospheric lifetime of isoprene and the large uncertainty and spatial inhomogeneity in biogenic emissions, a regional scale model with 50 km resolution should not be expected to agree very well with grab samples at rural stations. Given these limitations, Figure 17 shows that the model agrees fairly well with the isoprene measurements although a systematic bias (model underprediction) is seen. With respect to the carbonyls, the comparison shows a very nice agreement for formaldehyde at both stations whereas acetaldehyde and acetone is underpredicted by the model, particularly in summer.

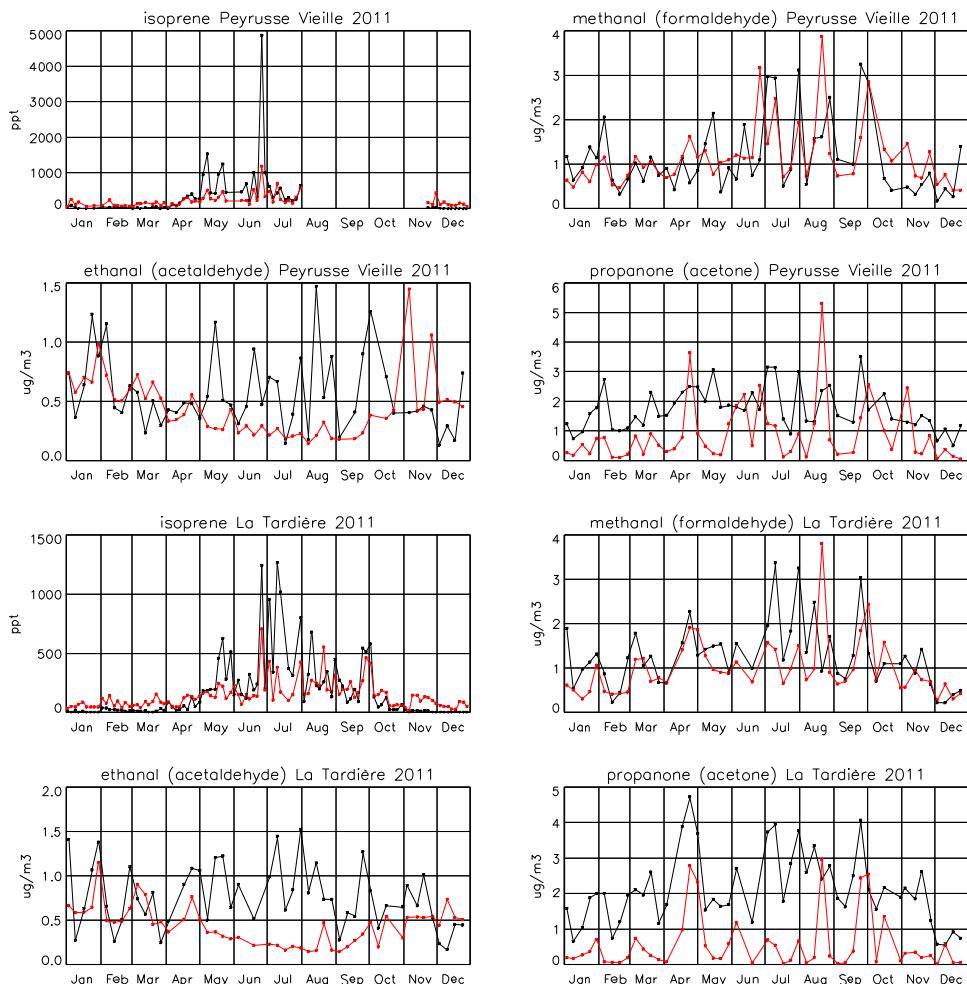


Figure 17: Measurements of isoprene, formaldehyde and acetaldehyde at Peyrusse Vieille (FR13) and La Tardière (FR15) in black together with results from the EMEP model (red).

5. Long-term trends in VOC

According to officially reported data, the emissions of VOC in Europe have dropped considerably the last decades. The total anthropogenic emission of VOC from the standard (old) EMEP domain went down from approx. 28 Tg in 1990 to 17 Tg in 2000 and 12 Tg in 2011 (EMEP, 2013). This corresponds to an approximate reduction of 30 % after 2000 and 57 % after 1990.

A simple 1:1 relationship between observed VOC concentrations at rural background sites and the overall European emission numbers are however, not to be expected. Interannual variations in atmospheric transport patterns, vertical mixing, photochemical oxidation as well as spatial differences in emission reductions complicates the analyses.

The measurements of selected VOCs at seven sites from 2001 to 2011 are shown in Figure 18–Figure 26. Based on these data seasonal trend curves were calculated by a non-linear least squares fit method and the fitted curves are given in red in the figures. The decadal trends and the corresponding confidence intervals are given in the plots when a statistically significant trend was estimated. We computed the fit to the following equation:

$$c(t) = [a_0 + a_1(\sin(2\pi(t-a_2)))]\exp[a_3(t-t_0)]$$

where

$c(t)$ = mixing ratio at time t measured in years

t_0 = 2001

and the coefficients a_0 , a_1 , a_2 , a_3 were determined by the non-linear least squares fit.

This expression represents a simple seasonal cycle with a mean level a_0 , amplitude a_1 and phase displacement a_2 superimposed on a long-term exponential change with the a_3 coefficient defining the rate of either a growth ($a_3 > 0$), a decline ($a_3 < 0$) or no trend ($a_3 = 0$).

To assure positive solutions the least squares fit was applied to log-transformed data, i.e. to $c^*(t) = \log(c(t))$, where $c(t)$ is the observed mixing ratio in ppt. This also corresponds to a weighting of the data, increasing the weight of the low-level concentrations. Without such a weighting, the least squares fit would be strongly determined by the highest concentration values mostly observed in winter.

The result of this exercise for the seven sites and nine species shown in Figure 18–Figure 26 is somewhat mixed. The estimated curves give a reasonable fit to the measurements on an overall basis. The estimated decadal trends differ, however, somewhat between the sites and compounds. Except for Waldhof and Schmücke a downward trend of the order of 10-50 % is calculated for many of the compounds/stations. These numbers, although highly uncertain, agree with the magnitude of the official emission data for the 2000-2011 period. For Waldhof and Schmücke the data indicate a strong increase in several compounds, most marked for benzene, toluene and propene. As mentioned above, the elevated levels of VOC at several German sites in 2011 (and partly 2010) is in contrast to the other sites and calls for a special attention with respect to sampling and analytical methods.

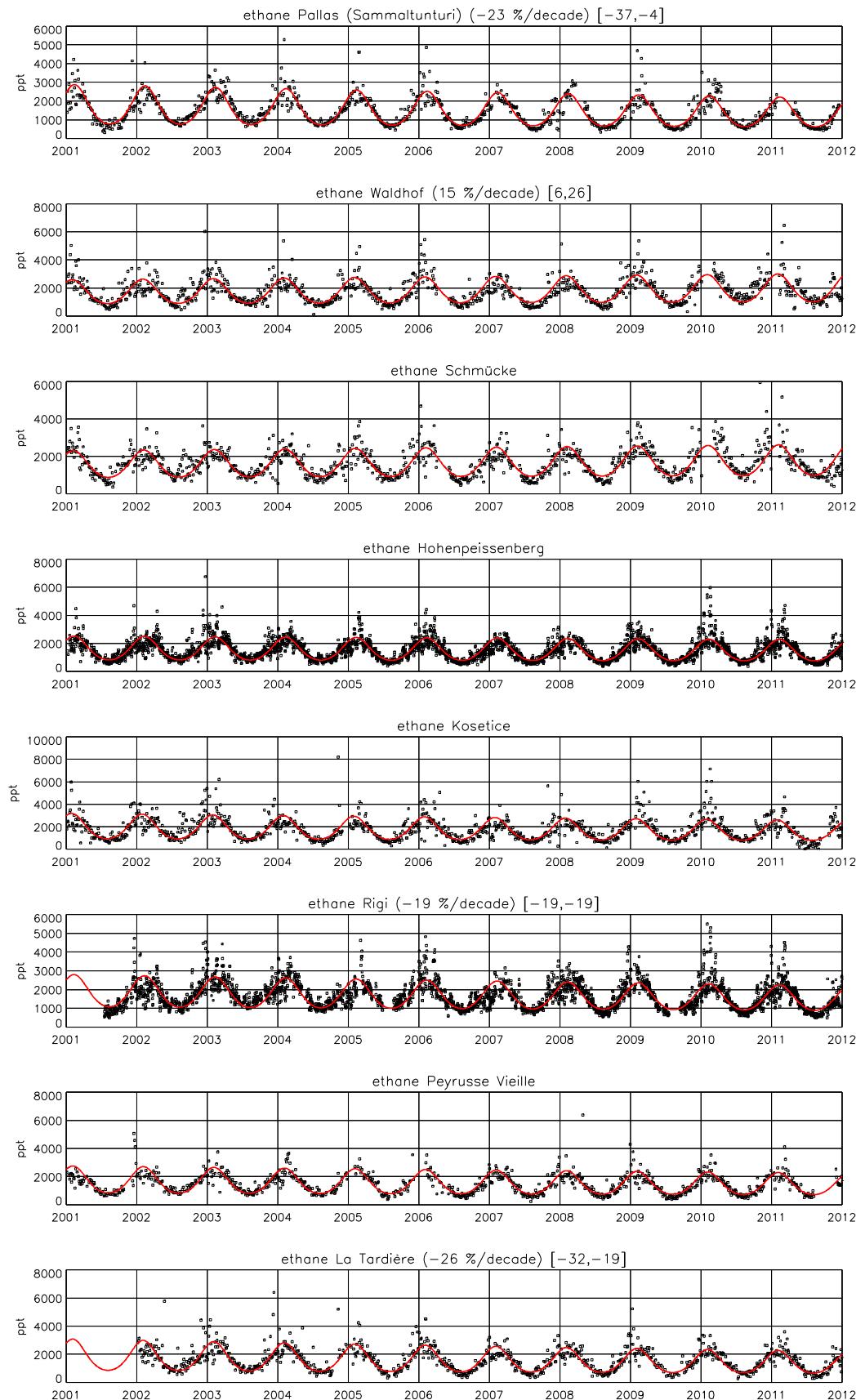


Figure 18: Measurements of ethane 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

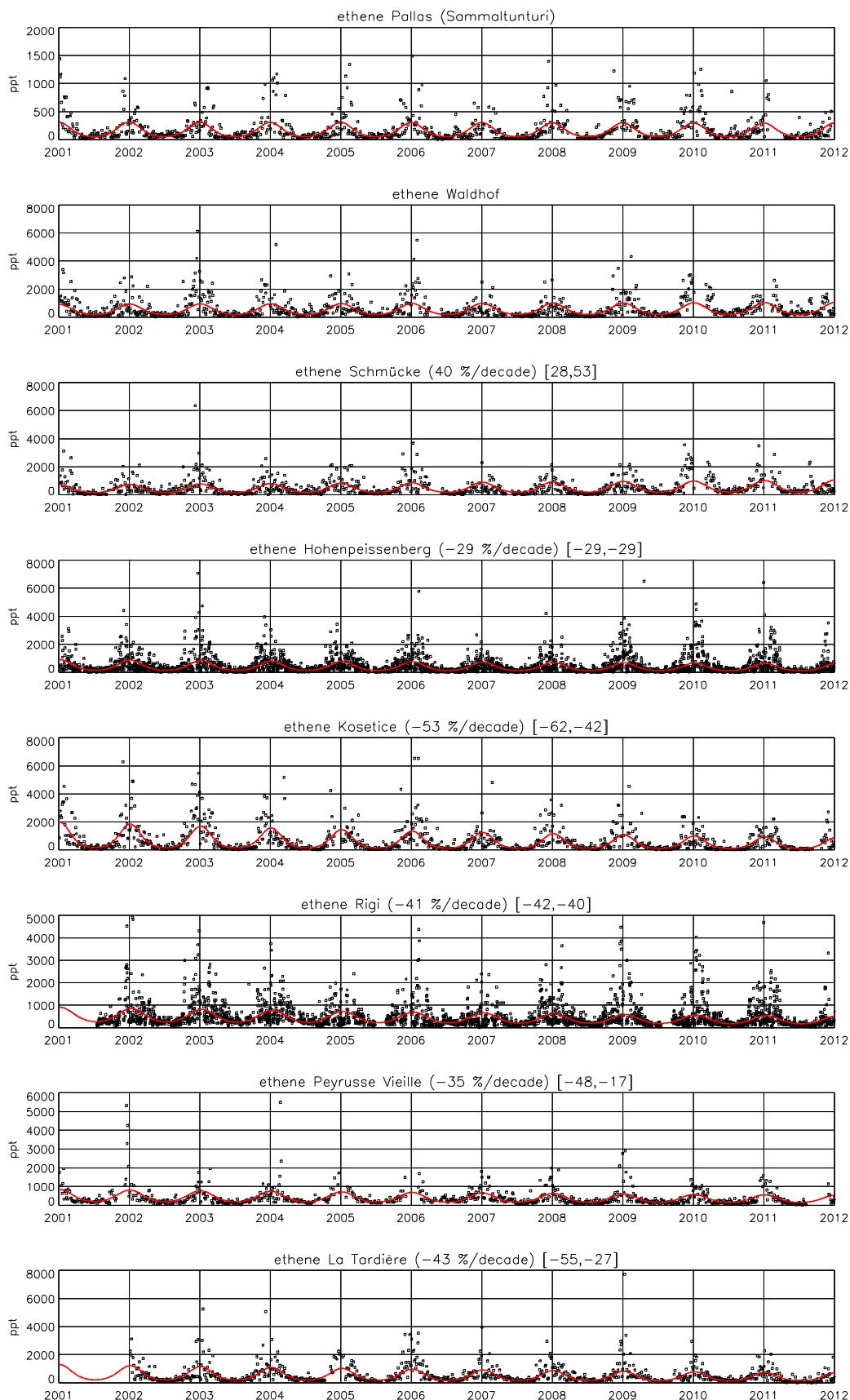


Figure 19: Measurements of ethene 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

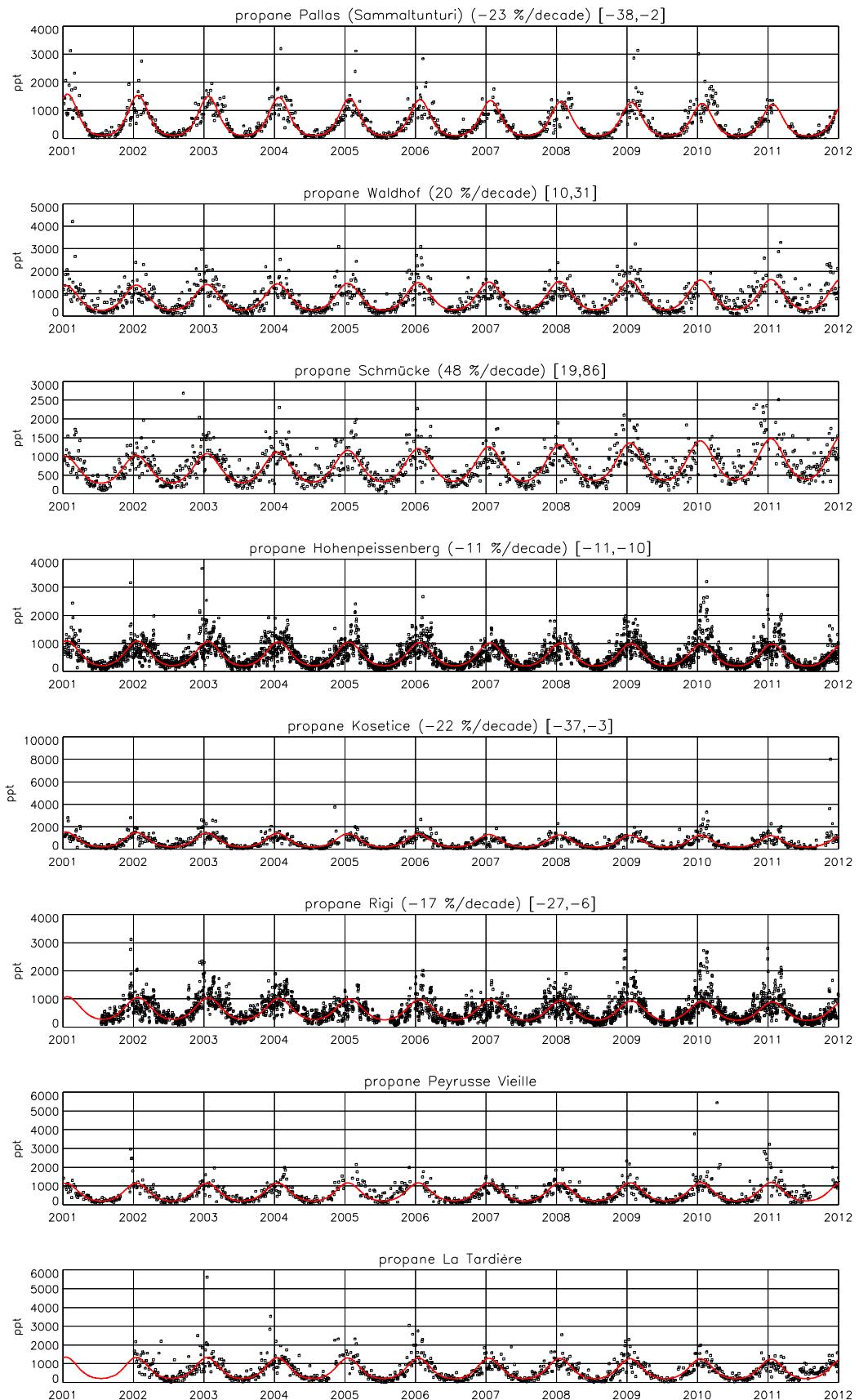


Figure 20. Measurements of propane 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

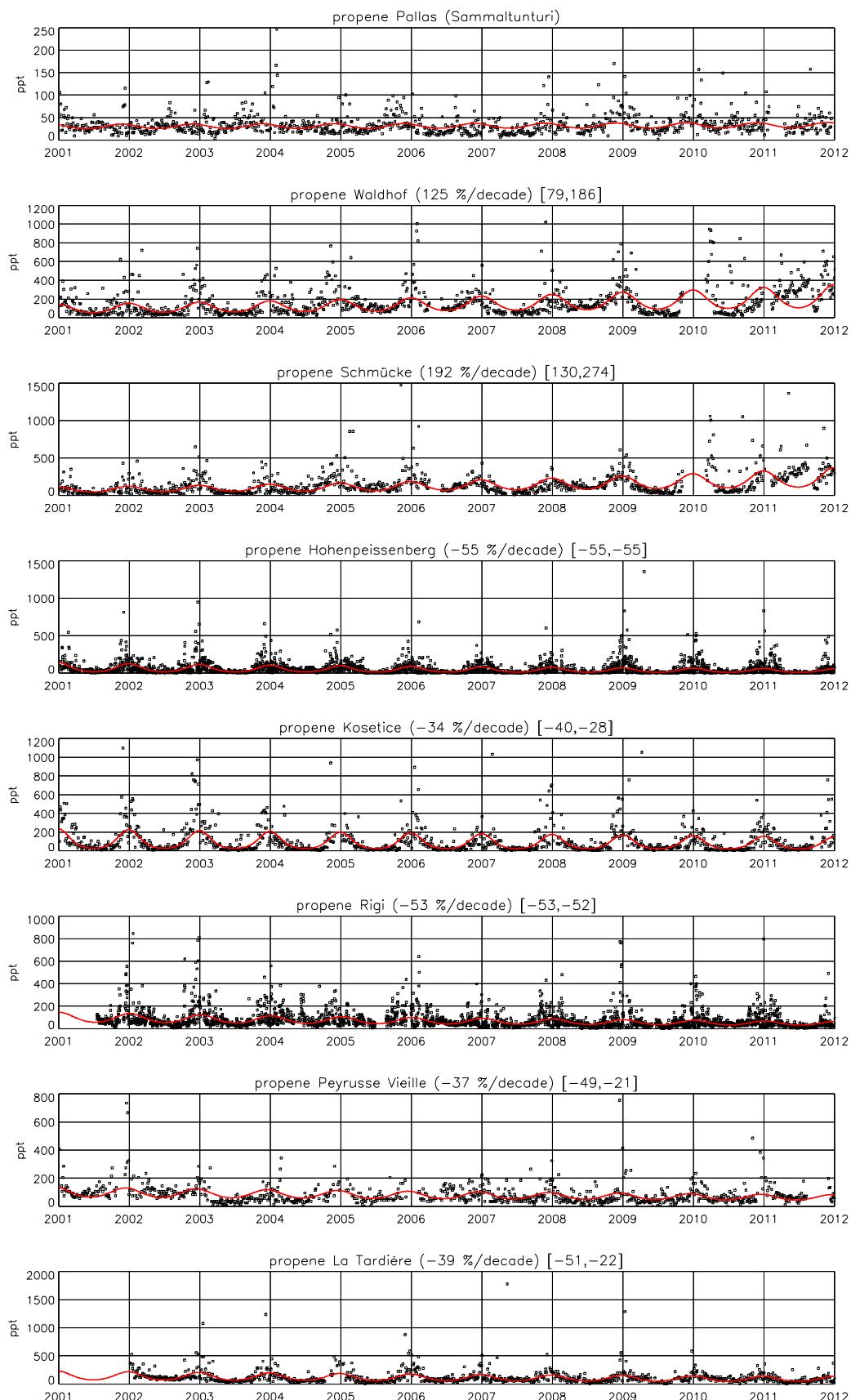


Figure 21: Measurements of propene 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

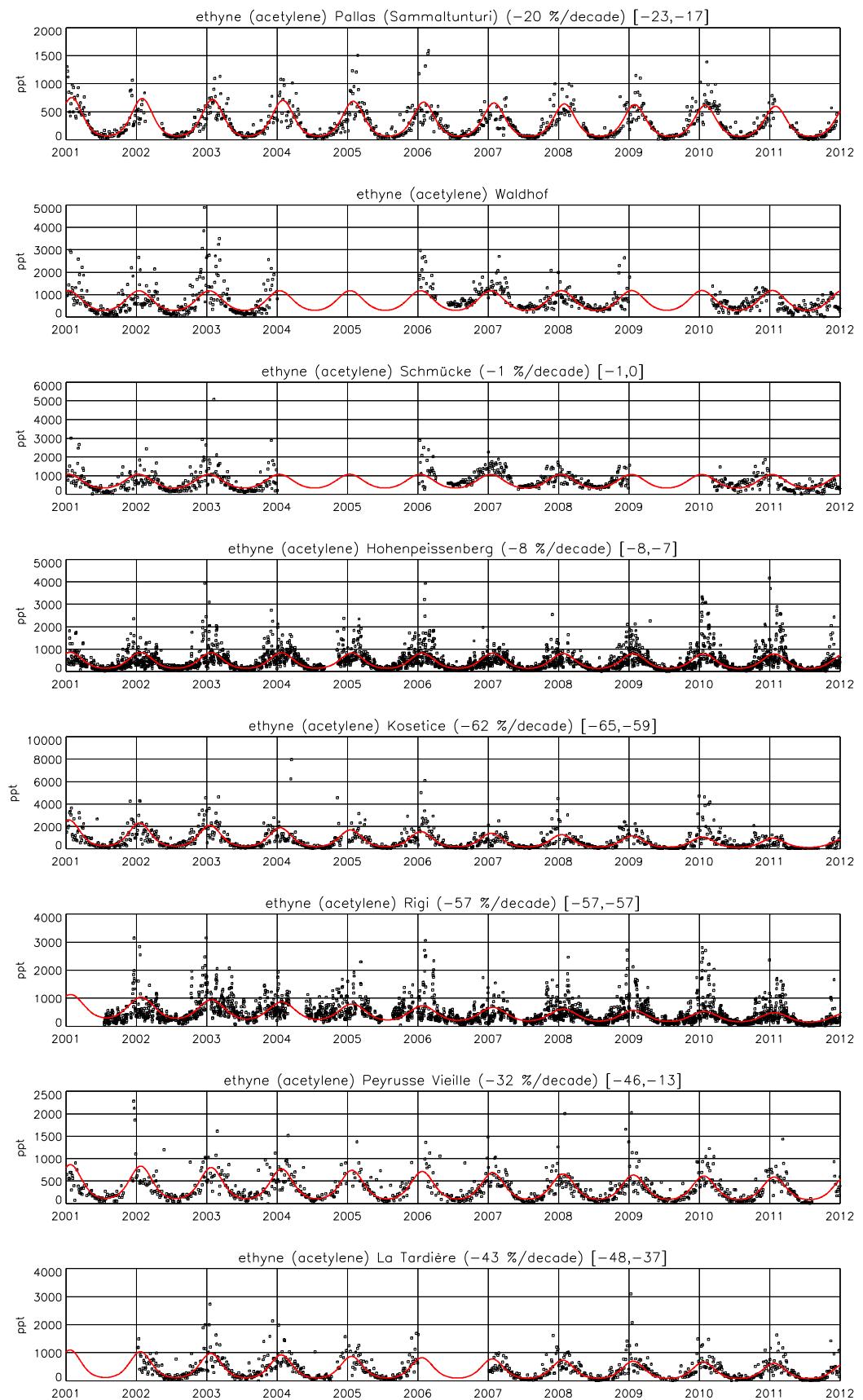


Figure 22: Measurements of ethyne 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

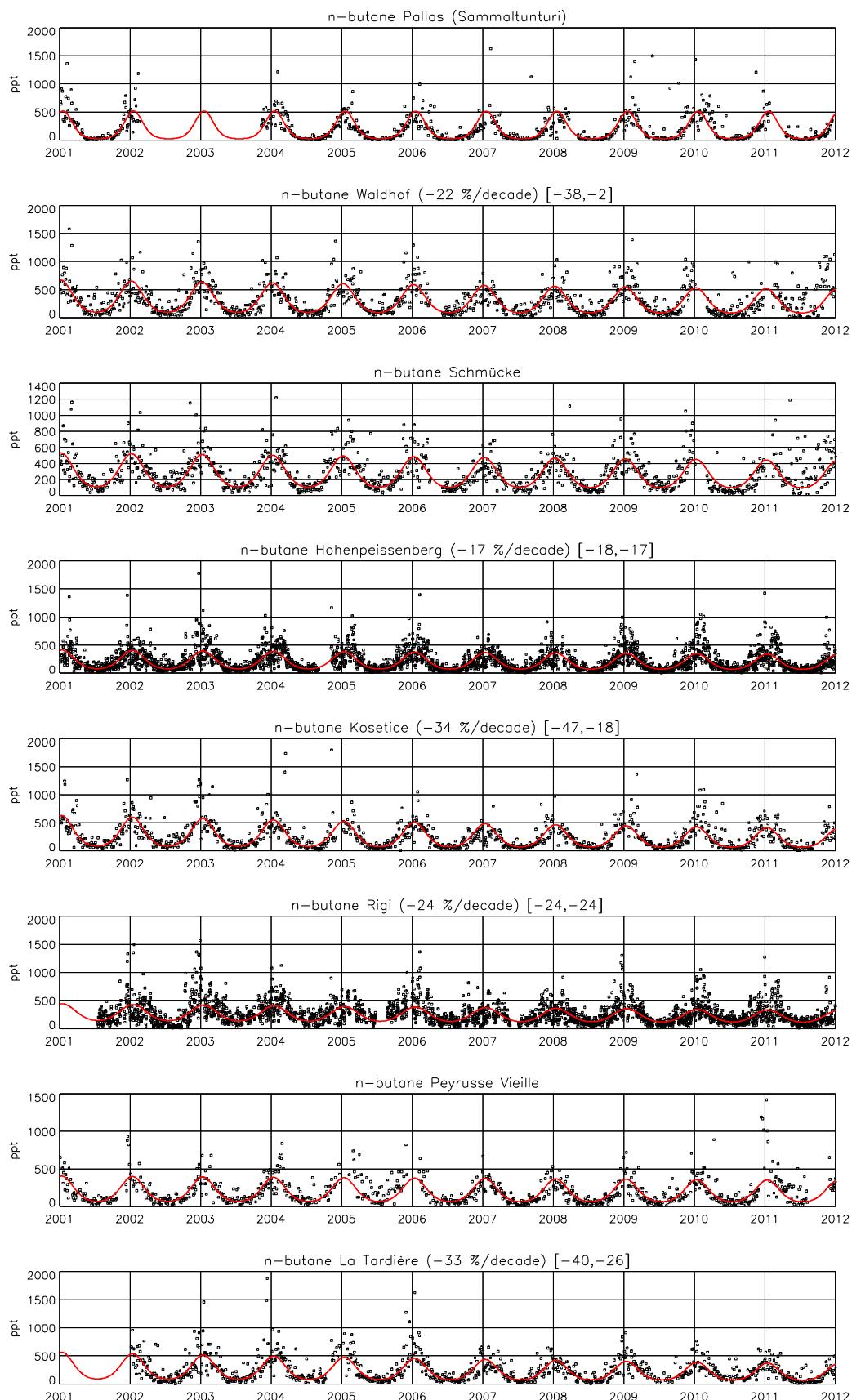


Figure 23: Measurements of n-butane 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

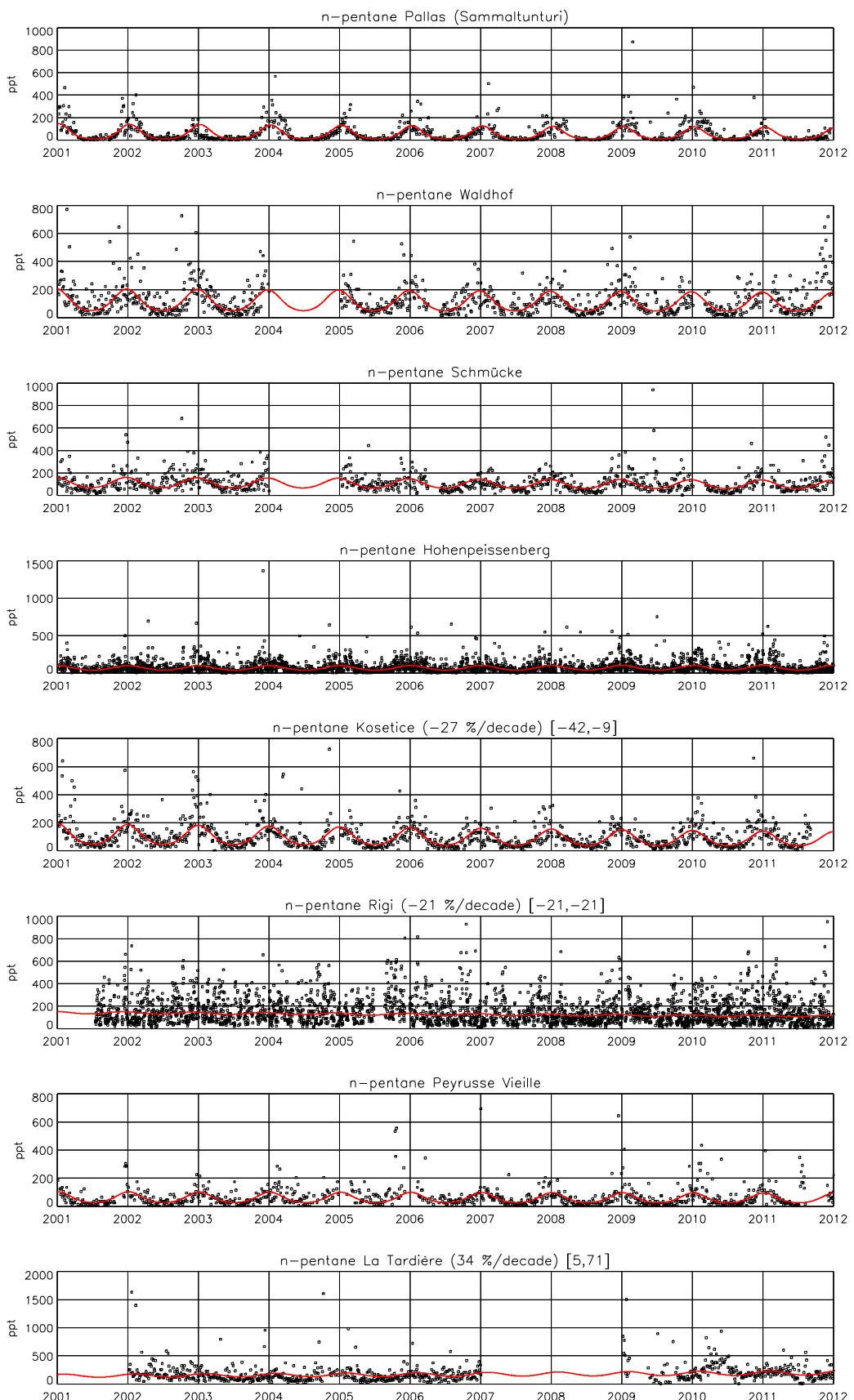


Figure 24: Measurements of n-pentane 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

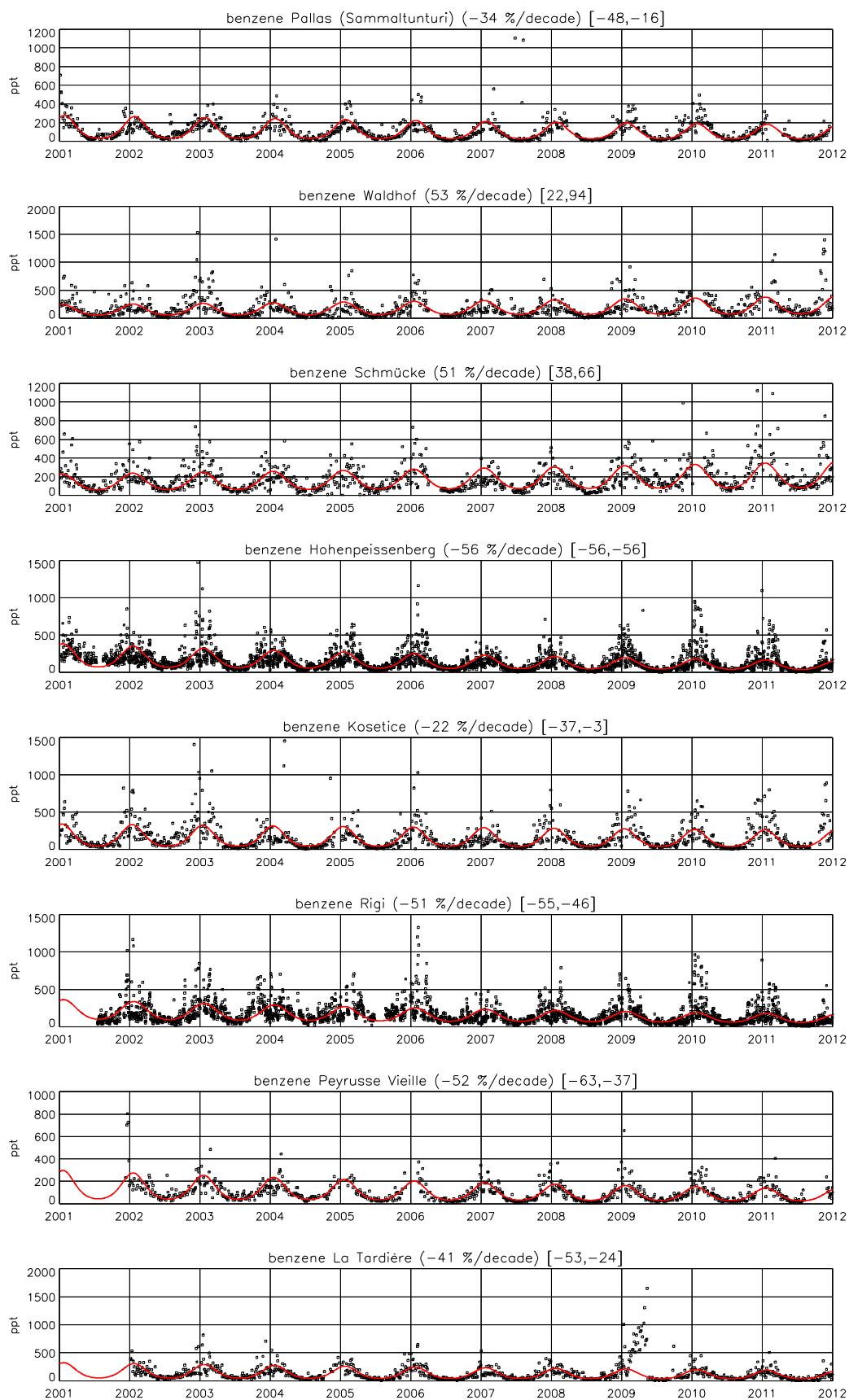


Figure 25: Measurements of benzene 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

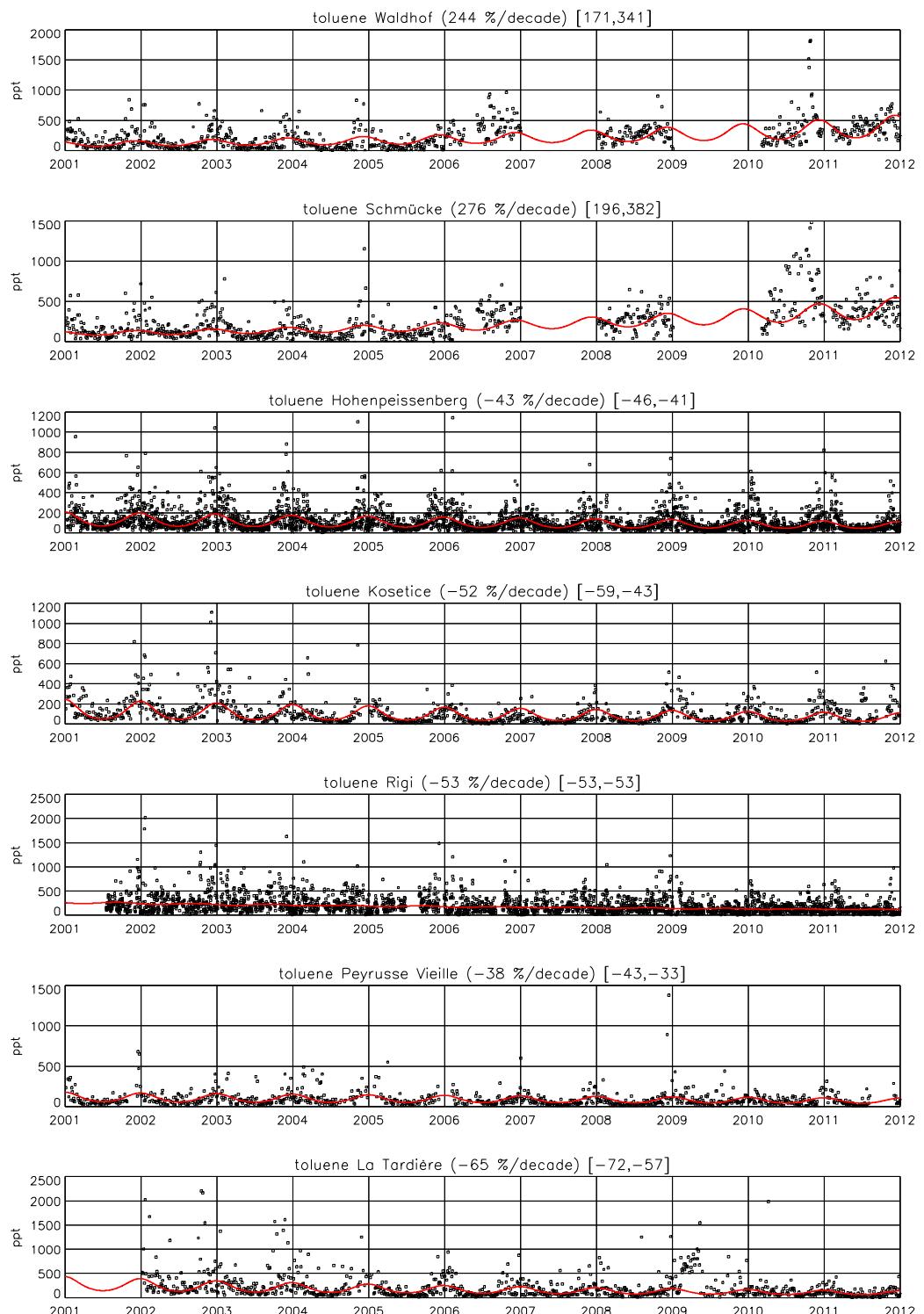


Figure 26: Measurements of toluene 2001-2011 together with the estimated trend curve in red (see text). Significant trends are given with confidence intervals.

Benzene is expected to have had a marked decline due to both the reduced benzene content of fuel and the general VOC emission reduction. By the least squares fit described above, we estimate a decadal reduction of 20-50 % during the period 2001-2011 for all sites except Waldhof and Schmücke. At these two

sites a strong increase (50 %) is estimated, apparently due to enhanced concentration levels the last few years. For acetylene, another tracer of traffic emissions, a decadal downward trend of 20-60 % is estimated at all sites except Waldhof and Schmücke.

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Appendix A

Monthly mean and median concentrations of hydrocarbons and carbonyls in 2011

**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	JAN 1974 1978	- -	- -	1545 1528	1269 1359	804 780	658 638	555 540	625 590	856 865	1317 1353	- -
Zingst	- -	3138 2729	2556 2144	1650 1784	2107 2019	1421 1454	1170 1170	1082 1087	1186 1152	1312 1236	1827 1827	1356 1366
Neuglobsow	- -	3513 3303	2475 2344	1286 1169	2288 2416	1486 1479	1332 1239	1167 1135	1099 1023	1411 1328	2180 2172	1292 1363
Waldhof	- -	3586 3326	2151 2146	1524 1652	2466 2529	1543 1708	1175 1173	1130 1121	1158 1108	1407 1439	2318 2702	1431 1393
Schmücke	- -	3179 2790	2200 1996	1711 1743	1909 1758	1513 1482	1185 1208	1011 976	1170 1053	1017 961	1212 1123	1439 1474
Schauinsland	- -	2200 2357	2470 2299	1156 1164	2196 2118	1303 1245	1206 1169	1064 972	1039 1055	799 816	1024 927	1078 1077
Hohenpeissenberg	2012 2059	2623 2595	2621 2295	1792 1837	1319 1253	926 834	811 784	642 619	756 719	1109 1114	1529 1412	1721 1702
Košetice	2096 2053	2783 2795	2506 2148	1640 1630	876 771	716 858	460 389	454 521	- -	1977 1836	2758 2808	1715 1854
Rigi	1905 1911	2219 2219	2637 2348	1763 1809	1428 1437	953 878	821 805	678 654	738 694	1168 1109	1476 1311	1809 1615
La Tardi��re	2051 2031	2296 2242	2411 2273	1697 1665	1400 1333	849 825	650 713	563 538	671 636	1032 1056	1865 1761	1616 1656
Peyrusse Vieille	2022 2080	2085 2003	2436 2332	1642 1601	1275 1267	708 687	727 704	- -	- -	- -	- -	1644 1540
	ETHENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	JAN 666 739	- -	- -	56 52	55 43	135 87	71 71	108 82	95 73	176 165	174 93	- -
Zingst	- -	988 951	763 683	298 209	269 279	405 379	374 356	1141 883	557 353	518 405	807 907	323 297
Neuglobsow	- -	1272 1196	805 666	227 233	366 366	342 345	423 400	700 730	421 423	482 452	1065 1051	358 282
Waldhof	- -	1397 1270	713 625	274 263	424 312	391 316	376 341	733 465	342 336	602 394	1252 1423	366 303
Schm��ck	- -	1359 1135	579 480	272 274	331 306	339 338	350 317	1089 869	470 328	405 407	514 562	400 403
Schauinsland	- -	651 625	821 708	207 206	369 351	440 487	400 393	- -	343 342	291 292	477 177	220 214
Hohenpeissenberg	922 741	1227 1050	737 503	230 213	161 129	143 110	143 119	120 93	174 150	423 353	756 526	426 345
Ko��etice	809 725	1419 1317	672 605	267 257	207 247	100 100	128 159	117 137	- -	587 695	1348 1054	541 410
Rigi	736 509	770 561	681 472	199 178	161 142	155 129	146 132	147 126	159 134	338 284	663 322	324 280
La Tardi��re	875 643	606 477	465 337	134 136	180 98	159 105	166 143	133 80	121 90	345 298	643 559	516 354
Peyrusse Vieille	953 1014	455 436	393 337	218 206	125 104	108 107	144 114	- -	- -	- -	- -	302 278

	PROPANE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	1192	-	-	461	238	88	89	87	218	264	515	-
	1185	-	-	452	255	79	81	75	195	279	530	-
Zingst	-	1602	1268	602	562	438	422	550	594	964	1611	1016
	-	1379	1271	574	446	364	434	538	530	846	1543	1020
Neuglobsow	-	1794	1100	548	627	484	488	540	494	882	1557	1022
	-	1830	995	551	589	372	513	470	446	882	1430	994
Waldhof	-	1804	1103	674	819	491	546	-	558	1030	1927	1203
	-	1634	1115	608	769	293	327	-	423	829	2007	1073
Schmücke	-	1524	1096	632	891	545	633	634	749	805	1187	1126
	-	1501	1051	601	786	485	537	608	743	876	1146	1075
Schauinsland	-	1014	1059	576	690	315	420	519	588	484	626	807
	-	938	948	593	485	264	408	415	637	503	506	803
Hohenpeissenberg	901	1135	1014	479	298	194	217	171	246	444	631	748
	959	1155	872	497	276	173	206	148	225	463	556	745
Košetice	1004	1324	889	467	305	278	208	192	-	883	2516	1032
	851	1350	737	454	270	237	187	164	-	692	1470	1025
Rigi	822	900	1001	499	337	204	219	203	252	431	624	716
	801	863	855	485	335	172	206	176	211	405	458	676
La Tardière	911	835	850	333	245	570	645	610	611	720	1152	1059
	923	868	771	351	261	649	534	571	578	690	1110	1038
Peyrusse Vieille	1720	1040	992	540	304	446	559	-	-	-	-	1129
	1658	991	869	577	202	354	576	-	-	-	-	1086
	PROPENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	64	-	-	24	23	43	30	51	33	50	33	-
	64	-	-	27	20	34	29	38	31	53	32	-
Zingst	-	220	295	242	362	300	412	460	91	349	466	296
	-	187	285	216	301	295	388	428	93	291	450	282
Neuglobsow	-	201	452	249	343	316	346	333	176	292	483	425
	-	197	334	222	232	300	308	324	126	304	404	308
Waldhof	-	196	367	242	341	311	408	383	140	354	527	365
	-	179	290	234	316	293	389	383	117	297	561	303
Schmücke	-	191	348	243	459	321	384	444	113	363	349	338
	-	176	253	232	314	329	412	376	110	293	380	345
Schauinsland	-	-	442	292	507	456	478	-	156	300	316	322
	-	-	373	271	405	464	503	-	140	273	298	309
Hohenpeissenberg	96	140	70	22	16	19	21	17	22	49	94	57
	79	86	44	17	14	15	17	15	18	37	64	45
Košetice	176	138	72	50	39	29	37	58	-	163	316	201
	175	116	62	36	33	26	34	44	-	116	247	146
Rigi	87	94	72	33	27	27	25	28	28	52	96	43
	65	62	51	31	26	24	25	25	26	43	46	35
La Tardière	124	117	78	30	75	57	92	56	87	123	134	110
	119	99	63	30	51	54	80	55	91	99	133	63
Peyrusse Vieille	152	98	86	60	65	57	83	-	-	-	-	73
	159	87	86	69	66	64	69	-	-	-	-	68

	ETHYNE (ACETYLENE)											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	632	-	-	244	167	63	44	44	81	149	257	-
	683	-	-	241	162	60	44	33	77	131	218	-
Zingst	-	468	426	191	264	405	327	621	569	624	706	436
	-	434	402	215	249	454	347	618	616	639	651	375
Neuglobsow	-	561	475	191	448	323	230	368	648	440	822	354
	-	502	428	185	429	341	175	329	622	464	894	354
Waldhof	-	566	413	223	409	414	283	444	566	415	912	422
	-	513	414	235	400	391	288	505	568	502	923	422
Schmücke	-	568	361	246	361	305	348	398	355	463	650	394
	-	540	323	241	356	380	303	418	253	487	670	360
Schauinsland	-	-	459	208	414	397	234	404	456	497	412	429
	-	-	395	209	336	440	225	308	472	500	319	439
Hohenpeissenberg	841	1343	911	429	222	144	141	123	188	386	642	421
	734	1143	735	421	174	131	136	114	155	384	492	382
Košetice	706	818	373	154	109	69	45	29	-	241	580	851
	540	859	345	157	111	64	43	26	-	279	525	725
Rigi	517	620	630	250	160	93	81	88	106	187	405	251
	401	492	497	245	142	82	77	78	82	165	248	231
La Tardi��re	608	700	679	324	203	52	40	52	112	236	642	447
	504	584	698	320	178	42	29	52	84	180	511	371
Peyrusse Vieille	588	512	683	289	172	35	48	-	-	-	-	346
	613	525	634	283	179	35	30	-	-	-	-	333
	N-BUTANE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	518	-	-	131	47	60	26	46	83	107	169	-
	495	-	-	102	41	38	19	27	78	100	153	-
Zingst	-	531	391	207	362	167	244	95	215	358	768	484
	-	472	406	200	444	22	181	26	141	331	704	455
Neuglobsow	-	584	345	181	128	211	352	22	119	431	728	452
	-	562	361	175	138	113	330	23	102	399	714	436
Waldhof	-	575	347	236	229	170	377	53	194	656	878	558
	-	522	364	225	171	99	398	20	97	644	900	503
Schm��cke	-	534	281	197	332	280	343	141	386	378	441	533
	-	525	289	210	156	205	299	171	355	366	449	541
Schauinsland	-	-	352	231	222	233	418	202	362	221	273	334
	-	-	280	235	161	38	429	162	321	192	189	337
Hohenpeissenberg	338	413	327	134	89	72	90	68	102	174	256	259
	344	440	252	131	86	59	83	63	98	156	212	242
Ko��setice	353	420	263	163	80	78	62	65	-	311	422	341
	335	429	220	128	73	66	62	53	-	332	411	335
Rigi	313	348	360	178	129	95	106	104	127	179	276	232
	291	306	306	164	128	79	99	94	105	153	159	220
La Tardi��re	367	283	275	133	88	82	176	80	72	122	299	267
	341	276	214	125	81	63	174	69	53	121	276	257
Peyrusse Vieille	714	336	325	220	174	187	136	-	-	-	-	284
	675	336	305	231	190	133	93	-	-	-	-	283

	2-METHYLPROPANE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	276	-	-	56	26	17	13	16	40	56	91	-
	276	-	-	57	23	13	11	11	36	55	76	-
Zingst	-	277	216	79	77	65	87	122	105	250	456	256
	-	241	212	73	71	66	89	98	85	243	433	236
Neuglobsow	-	300	247	79	92	131	84	124	90	202	459	255
	-	285	193	80	82	62	85	97	87	169	444	248
Waldhof	-	310	180	117	143	94	135	107	114	291	525	292
	-	292	167	101	106	57	67	93	85	293	499	261
Schmücke	-	286	152	101	113	76	108	123	124	174	280	289
	-	285	141	87	88	59	124	125	121	171	231	281
Schauinsland	-	-	206	106	154	85	116	151	166	120	161	177
	-	-	150	98	98	45	89	111	172	106	104	172
Hohenpeissenberg	210	267	220	93	64	50	62	48	74	118	191	146
	197	268	170	91	60	41	57	41	66	105	130	136
Košetice	234	267	176	110	59	63	45	48	-	356	452	240
	206	272	143	103	51	51	48	43	-	251	331	214
Rigi	189	202	219	99	69	48	53	49	61	96	152	109
	163	170	174	93	68	41	49	43	48	81	81	102
La Tardière	208	166	157	72	39	42	-	28	32	57	156	137
	186	162	123	73	39	27	-	17	26	60	135	140
Peyrusse Vieille	525	224	233	210	171	135	47	-	-	-	-	222
	477	215	220	231	178	45	42	-	-	-	-	216
	BUTENES											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Zingst	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Neuglobsow	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Waldhof	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Schmücke	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Schauinsland	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Hohenpeissenberg	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Košetice	44	75	75	82	70	148	150	122	-	181	126	113
	36	75	72	66	60	147	156	119	-	115	132	113
Rigi	55	65	67	59	62	72	66	70	62	57	72	39
	43	49	62	58	61	69	67	69	61	53	45	37
La Tardière	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Peyrusse Vieille	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-

	1-BUTENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	15 14	- -	- -	8 8	7 6	11 12	7 7	10 10	8 7	10 11	8 8	- -
Zingst	- -	33 30	29 24	39 17	30 27	32 32	39 38	37 42	33 33	32 30	48 42	28 24
Neuglobsow	- -	37 34	37 36	26 22	27 31	28 31	43 40	41 40	40 31	27 28	54 48	32 28
Waldhof	- -	36 35	32 27	20 22	37 30	31 29	41 45	38 40	30 29	40 29	57 59	35 26
Schmücke	- -	39 29	25 22	20 21	27 30	32 33	43 38	44 37	35 32	28 23	33 31	37 36
Schauinsland	- -	- 26	34 17	15 40	35 48	45 44	47 44	- -	43 45	23 24	21 18	24 24
Hohenpeissenberg	18 15	23 19	16 12	7 7	6 6	6 5	6 5	5 5	6 6	11 9	18 13	12 10
Košetice	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Rigi	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
La Tardi��re	34 27	37 36	25 23	18 19	22 18	18 17	23 18	15 15	17 16	27 24	26 26	26 23
Peyrusse Vieille	40 34	32 30	26 26	21 21	17 17	15 15	18 18	- -	- -	- -	- -	13 14
	TRANS_2-BUTENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	3 3	- -	- -	4 3	4 3	4 3	4 3	3 3	3 3	3 3	4 3	- -
Zingst	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Neuglobsow	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Waldhof	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Schmücke	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Schauinsland	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Hohenpeissenberg	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Košetice	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Rigi	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
La Tardi��re	4 3	56 62	16 21	9 6	6 3	3 3	4 3	3 3	3 3	8 3	9 11	7 3
Peyrusse Vieille	3 3	43 35	35 37	18 17	3 3	3 3	5 3	- -	- -	- -	- -	3 3

	CIS_2-BUTENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	6 3	- -	- -	4 3	3 3	4 3	3 3	3 3	3 3	3 3	3 3	- -
Zingst	- -	25 20	39 39	63 42	64 53	57 54	99 99	108 100	36 36	30 27	35 32	27 26
Neuglobsow	- -	21 21	75 45	40 36	59 46	69 62	81 71	75 67	57 54	27 26	31 30	31 24
Waldhof	- -	24 23	60 45	41 41	58 55	66 58	98 89	93 91	48 39	30 27	38 38	29 26
Schmücke	- -	19 20	51 39	39 38	82 66	66 60	91 95	116 99	44 47	37 29	34 33	29 27
Schauinsland	- -	- 60	72 48	50 75	88 114	106 123	116 -	- 64	38 35	39 41	33 31	
Hohenpeissenberg	6 6	8 8	7 6	4 4	3 3	3 3	3 3	3 3	3 3	4 3	6 4	5 5
Košetice	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
Rigi	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	- -	
La Tardière	7 3	6 3	3 3	3 3	5 3	3 3	4 3	3 3	3 3	6 3	4 3	4 3
Peyrusse Vieille	3 3	3 3	4 3	3 3	3 3	3 3	5 3	- -	- -	- -	- -	3 3
	N-PENTANE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	99 102	- -	- -	21 22	13 10	11 8	8 8	24 17	25 26	32 29	44 30	- -
Zingst	- -	143 131	113 123	48 41	48 47	53 34	67 59	96 52	85 60	206 190	363 359	194 177
Neuglobsow	- -	179 168	120 93	43 47	61 65	63 42	77 80	102 76	86 77	153 163	389 329	197 176
Waldhof	- -	168 162	115 129	71 49	109 65	82 61	101 61	102 60	108 83	245 266	479 421	254 212
Schmücke	- -	165 142	97 84	56 52	76 58	75 75	113 81	110 100	99 87	131 133	212 163	214 187
Schauinsland	- -	102 59	115 83	131 91	125 74	76 35	100 81	131 70	152 152	103 66	100 47	119 117
Hohenpeissenberg	113 101	166 149	120 85	62 53	52 42	43 31	59 48	43 33	65 61	104 79	112 83	77 70
Košetice	127 121	151 157	96 87	67 63	46 33	50 43	55 42	169 174	- -	- -	- -	- -
Rigi	124 94	174 117	174 130	116 84	104 75	69 50	87 59	77 55	110 67	133 98	195 76	84 73
La Tardière	260 244	212 219	239 238	295 263	281 255	177 129	55 26	245 176	126 113	158 147	191 161	215 203
Peyrusse Vieille	156 113	76 74	69 50	30 28	27 27	16 15	218 196	- -	- -	- -	- -	88 68

	2-METHYLBUTANE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	171	-	-	34	15	28	11	29	33	36	51	-
	168	-	-	26	12	12	9	15	28	33	31	-
Zingst	-	156	136	73	92	76	99	122	123	257	330	246
	-	144	118	61	90	57	98	97	87	190	359	237
Neuglobsow	-	183	117	42	127	109	95	137	167	171	367	250
	-	175	94	36	122	67	61	129	105	185	392	229
Waldhof	-	175	121	86	168	106	111	109	145	252	478	287
	-	161	119	64	126	59	73	94	104	291	475	267
Schmücke	-	186	95	78	133	97	132	170	113	167	236	269
	-	171	93	84	124	58	115	178	106	160	222	232
Schauinsland	-	121	133	124	206	58	116	259	245	158	136	149
	-	86	116	102	130	49	105	163	119	114	87	143
Hohenpeissenberg	151	205	154	88	76	70	87	82	107	144	170	111
	147	198	127	80	73	66	77	78	104	125	135	99
Košetice	163	186	111	83	61	71	82	255	-	-	-	-
	151	191	92	76	41	59	60	255	-	-	-	-
Rigi	188	241	232	164	156	134	142	157	190	187	258	143
	153	189	211	149	142	116	126	138	154	160	132	127
La Tardi��re	163	120	113	84	62	58	95	60	51	75	151	113
	148	107	94	73	53	44	39	42	40	90	130	103
Peyrusse Vieille	179	91	86	48	35	20	101	-	-	-	-	79
	168	84	76	44	30	22	73	-	-	-	-	65
	N-HEXANE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	35	-	-	5	16	5	4	8	8	8	15	-
	38	-	-	3	6	3	3	7	8	8	10	-
Zingst	-	43	38	18	27	26	36	39	41	87	107	62
	-	38	39	17	29	23	36	33	42	86	98	56
Neuglobsow	-	46	27	16	25	26	35	39	50	63	118	73
	-	46	23	17	23	28	32	31	51	60	103	66
Waldhof	-	45	30	27	44	36	40	39	53	86	122	77
	-	44	33	21	30	25	29	36	45	80	122	69
Schm��cke	-	46	28	19	32	32	39	41	48	55	59	71
	-	41	25	20	30	32	33	35	42	64	48	67
Schauinsland	-	31	31	27	41	35	41	38	59	48	38	43
	-	25	25	25	34	35	39	33	61	49	29	42
Hohenpeissenberg	29	37	27	13	9	9	10	10	14	16	21	19
	28	37	19	11	7	7	8	8	13	14	16	17
Ko��etice	43	48	26	15	10	12	9	11	-	60	65	37
	41	47	24	14	8	10	9	9	-	59	59	37
Rigi	27	28	28	14	13	12	13	14	20	20	31	22
	23	22	21	12	11	10	12	13	17	18	17	20
La Tardi��re	39	35	28	15	13	11	25	6	3	6	29	30
	38	33	23	14	13	11	3	3	3	3	30	25
Peyrusse Vieille	44	26	24	13	8	7	10	-	-	-	-	23
	38	29	20	13	3	3	3	-	-	-	-	22

	ISOPRENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	5 4	- -	- -	6 4	4 4	26 14	33 30	4 4	4 4	4 4	4 4	- -
Zingst	- -	6 6	5 4	4 3	36 25	216 195	169 100	298 197	91 75	19 18	16 16	12 9
Neuglobsow	- -	7 7	6 7	9 7	52 38	137 122	69 49	154 136	62 47	20 18	26 22	21 17
Waldhof	- -	7 7	6 6	7 7	22 19	43 42	44 39	39 30	40 37	21 17	21 21	13 14
Schmücke	- -	8 5	5 5	9 10	37 29	32 29	65 45	52 49	20 20	15 12	14 14	15 13
Schauinsland	- -	8 7	10 7	17 17	99 73	60 45	57 44	198 126	86 64	38 27	13 13	9 8
Hohenpeissenberg	5 4	6 5	7 5	24 8	45 17	52 19	39 13	60 17	35 14	11 8	8 6	4 3
Košetice	15 9	7 7	3 3	24 10	70 68	78 57	59 53	116 86	- -	12 12	14 11	17 12
Rigi	9 9	12 12	10 8	26 13	41 24	64 33	55 28	94 38	43 17	21 14	20 17	22 11
La Tardière	6 3	25 23	14 13	55 50	303 196	321 201	711 665	341 260	278 224	120 65	13 14	4 3
Peyrusse Vieille	22 6	24 24	18 15	202 204	780 701	1224 688	362 300	- -	- -	- -	- -	5 3
	BENZENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	202 227	- -	- -	90 78	74 61	26 25	23 24	33 21	39 34	52 42	97 73	- -
Zingst	- -	414 319	342 259	129 107	139 135	118 105	116 99	150 128	137 128	321 263	635 616	222 185
Neuglobsow	- -	538 452	261 190	128 122	145 133	116 116	118 119	139 139	145 141	307 217	863 677	229 198
Waldhof	- -	578 485	278 216	161 156	179 161	123 104	123 122	116 108	121 88	350 307	928 1170	237 209
Schmücke	- -	517 446	241 182	155 148	156 138	115 115	122 95	126 119	133 125	242 195	382 312	260 281
Schauinsland	- -	289 260	304 222	156 142	183 136	167 140	127 119	154 137	179 168	185 241	184 122	153 142
Hohenpeissenberg	213 188	315 270	231 182	101 95	52 45	35 29	40 37	33 30	48 45	97 90	157 125	116 107
Košetice	376 272	388 426	196 169	124 118	72 69	54 50	49 48	43 39	- -	250 282	523 483	249 212
Rigi	190 157	215 177	231 184	94 90	59 55	39 33	38 36	37 32	43 32	84 71	145 93	94 87
La Tardière	195 163	181 160	158 122	79 79	52 44	36 36	37 24	26 21	29 19	65 71	165 137	129 113
Peyrusse Vieille	180 184	136 136	162 125	66 69	34 33	25 22	48 38	- -	- -	- -	100 97	

	TOLUENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
Zingst	-	335	274	193	370	353	383	366	319	406	511	255
	-	295	270	166	372	337	362	348	344	392	444	216
Neuglobsow	-	489	384	293	459	575	468	546	699	458	742	504
	-	471	369	278	431	561	479	455	627	448	731	466
Waldhof	-	330	265	227	394	403	437	369	448	431	641	320
	-	347	276	244	390	396	461	357	444	484	652	271
Schmücke	-	367	292	297	381	447	458	449	416	380	427	396
	-	366	268	285	398	423	396	415	336	396	456	347
Schauinsland	-	-	398	382	545	-	420	543	601	415	320	373
	-	-	372	354	545	-	419	511	597	361	276	263
Hohenpeissenberg	131	183	116	58	50	53	60	55	73	110	135	80
	117	159	99	50	45	47	49	45	75	100	106	68
Košetice	169	162	83	81	27	144	163	51	-	236	224	139
	160	157	71	88	27	63	150	59	-	152	214	106
Rigi	153	218	186	102	95	90	93	90	122	148	214	75
	106	134	152	78	84	76	74	79	95	116	94	60
La Tardi��re	177	190	107	64	66	76	45	80	70	106	177	144
	148	138	80	57	34	50	42	55	74	92	186	79
Peyrusse Vieille	175	102	83	48	32	35	71	-	-	-	-	76
	168	80	73	42	27	20	46	-	-	-	-	67
	ETHYLBENZENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
Zingst	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Neuglobsow	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Waldhof	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Schm��cke	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Schauinsland	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Hohenpeissenberg	21	32	21	12	10	11	10	10	12	18	24	14
	19	27	19	11	9	9	9	9	13	17	18	11
Ko��setice	20	25	14	11	7	18	12	20	-	45	38	18
	22	22	16	9	5	9	9	19	-	21	40	14
Rigi	23	34	26	15	12	16	13	13	17	23	34	12
	16	23	20	12	11	13	12	12	14	21	18	10
La Tardi��re	37	44	36	20	20	19	14	16	14	24	38	36
	26	32	29	21	18	14	14	3	12	22	35	22
Peyrusse Vieille	38	22	17	9	4	6	20	-	-	-	-	12
	35	21	13	6	3	3	14	-	-	-	-	11

	m+p-XYLENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Zingst	-	66	72	44	60	56	66	89	125	86	136	41
	-	60	65	34	65	52	55	74	86	81	118	36
Neuglobsow	-	88	91	45	53	70	87	73	137	62	135	54
	-	78	52	43	57	76	67	72	131	68	82	35
Waldhof	-	81	61	48	65	60	88	63	75	101	165	61
	-	84	55	44	54	42	67	59	75	76	156	28
Schmücke	-	108	47	51	59	75	69	66	82	71	89	57
	-	73	41	40	47	49	46	68	57	62	58	32
Schauinsland	-	83	84	43	92	107	87	100	107	60	41	19
	-	57	61	41	90	113	90	83	82	51	22	13
Hohenpeissenberg	51	77	46	23	18	21	22	20	23	44	69	36
	42	60	36	20	16	17	16	17	20	37	50	27
Košetice	49	43	24	20	12	15	19	88	-	59	90	45
	55	33	26	13	8	13	9	92	-	49	86	42
Rigi	60	97	59	30	25	32	29	26	35	56	105	33
	38	62	42	23	20	26	25	24	27	47	46	28
La Tardière	96	117	105	65	61	75	62	63	44	59	77	95
	57	83	91	56	57	51	43	40	41	56	67	48
Peyrusse Vieille	88	59	47	47	34	28	55	-	-	-	-	35
	78	43	48	43	31	19	37	-	-	-	-	26
	o-XYLENE											
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
Pallas	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Zingst	-	27	32	16	26	22	27	31	32	43	61	15
	-	23	31	12	24	17	20	33	36	42	60	13
Neuglobsow	-	41	32	17	39	29	41	38	55	45	67	28
	-	41	24	15	26	32	32	35	58	49	53	18
Waldhof	-	34	26	22	46	24	39	34	23	57	80	24
	-	38	26	26	23	20	32	31	21	61	79	15
Schmücke	-	46	21	22	37	29	30	26	27	44	50	24
	-	35	19	19	17	15	28	27	21	49	44	12
Schauinsland	-	38	38	20	50	45	44	42	48	40	36	9
	-	28	26	19	60	54	39	36	39	39	14	7
Hohenpeissenberg	-	-	-	-	-	-	-	-	-	-	-	-
	-	-	-	-	-	-	-	-	-	-	-	-
Košetice	14	16	8	5	3	4	23	13	-	45	24	13
	15	13	8	4	2	3	9	12	-	14	26	12
Rigi	25	36	25	12	10	12	10	10	14	24	43	14
	16	23	19	10	8	10	9	9	11	19	22	11
La Tardière	28	46	28	11	9	12	20	16	5	12	26	29
	19	33	34	7	3	3	19	3	3	13	21	18
Peyrusse Vieille	31	34	23	9	4	3	15	-	-	-	-	7
	27	35	22	6	3	3	9	-	-	-	-	3

**Monthly mean and median concentrations
(first and second line, respectively)
of carbonyls ($\mu\text{g m}^{-3}$)**

METHANAL (FORMALDEHYDE)													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	1.173	0.698	1.198	-	1.329	-	2.087	1.947	1.488	-	1.062	0.336	
	1.140	0.664	1.160	-	1.422	-	1.895	1.710	1.080	-	1.104	0.313	
Peyrusse Vieille	1.047	0.917	0.879	0.757	1.145	1.098	1.819	1.866	-	-	0.527	0.569	
	1.145	0.642	0.881	0.741	0.913	0.920	1.903	1.612	-	-	0.503	0.356	
ETHANAL (ACETALDEHYDE)													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	0.952	0.632	0.594	-	0.926	-	0.975	0.989	0.670	-	0.748	0.328	
	1.065	0.582	0.656	-	1.062	-	0.918	0.806	0.565	-	0.664	0.345	
Peyrusse Vieille	0.772	0.658	0.401	0.449	0.608	0.544	0.475	0.783	-	-	0.427	0.331	
	0.739	0.537	0.399	0.454	0.507	0.464	0.527	0.864	-	-	0.422	0.230	
PROPANONE (ACETONE)													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	1.436	1.478	1.963	-	2.082	-	3.079	2.984	2.515	-	1.954	0.698	
	1.584	1.585	2.035	-	1.686	-	3.296	2.786	2.183	-	1.893	0.658	
Peyrusse Vieille	1.262	1.465	1.615	2.063	2.241	1.876	2.147	2.107	-	-	1.337	0.852	
	1.243	1.059	1.485	2.113	1.989	1.764	2.272	2.361	-	-	1.318	0.858	
PROPANAL													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	0.195	0.037	0.383	-	0.408	-	0.114	0.149	0.112	-	0.569	0.027	
	0.152	0.030	0.272	-	0.282	-	0.110	0.139	0.102	-	0.664	0.027	
Peyrusse Vieille	0.259	0.048	-	-	-	-	-	-	-	-	-	0.200	
	0.375	0.046	-	-	-	-	-	-	-	-	-	0.085	
BUTANALS													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	0.078	0.010	0.014	-	0.085	-	0.093	0.127	0.108	-	0.066	0.023	
	0.065	0.005	0.005	-	0.089	-	0.087	0.130	0.109	-	0.072	0.020	
Peyrusse Vieille	0.051	0.010	0.012	0.049	0.099	0.119	0.038	0.099	-	-	0.013	0.019	
	0.052	0.005	0.005	0.049	0.100	0.122	0.045	0.123	-	-	0.006	0.017	
2-PROPENAL													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	-	0.005	0.006	-	0.006	-	-	0.009	0.016	-	0.007	0.007	
	-	0.005	0.005	-	0.006	-	-	0.008	0.008	-	0.006	0.007	
Peyrusse Vieille	-	0.005	0.005	0.006	0.006	0.005	0.006	0.006	-	-	0.006	0.011	
	-	0.005	0.005	0.007	0.006	0.005	0.006	0.006	-	-	0.006	0.006	
BUTANONE													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	0.541	0.518	0.731	-	0.595	-	0.336	0.254	0.257	-	0.514	0.187	
	0.486	0.470	0.692	-	0.553	-	0.375	0.245	0.267	-	0.550	0.165	
Peyrusse Vieille	0.351	0.425	0.551	0.690	0.887	0.900	1.358	1.406	-	-	0.261	0.150	
	0.331	0.360	0.524	0.749	0.890	0.903	1.313	1.174	-	-	0.228	0.120	
3-BUTEN-2-ONE													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	0.135	0.041	0.049	-	0.037	-	0.089	0.106	0.043	-	0.036	0.017	
	0.082	0.034	0.049	-	0.039	-	0.090	0.079	0.050	-	0.033	0.007	
Peyrusse Vieille	0.068	0.034	0.031	0.029	0.006	0.027	0.026	0.017	-	-	0.012	0.011	
	0.061	0.039	0.031	0.029	0.006	0.030	0.021	0.006	-	-	0.006	0.006	
2-METHYLPROPENAL													
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	
La Tardi��re	0.018	0.005	0.006	-	0.015	-	0.100	0.148	0.090	-	0.006	0.007	
	0.006	0.005	0.005	-	0.006	-	0.084	0.099	0.060	-	0.006	0.007	
Peyrusse Vieille	0.013	0.005	0.005	0.006	0.088	0.133	0.176	0.255	-	-	0.006	0.006	
	0.006	0.005	0.005	0.007	0.066	0.142	0.121	0.221	-	-	0.006	0.006	

BENZENCARBALDEHYDE												
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
La Tardière	0.104 0.092	0.119 0.092	0.159 0.152	-	0.107 0.068	-	0.108 0.083	0.033 0.033	0.035 0.033	-	0.089 0.084	0.014 0.007
Peyrusse Vieille	0.032 0.032	0.051 0.037	0.066 0.046	0.053 0.022	0.078 0.068	0.176 0.137	0.133 0.161	0.312 0.231	-	-	0.104 0.123	0.067 0.070
ETHANEDIAL (GLYOXAL)												
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
La Tardière	0.025 0.018	- -	- -	-	0.088 0.090	-	0.037 0.019	0.026 0.029	0.021 0.006	-	0.040 0.033	0.067 0.068
Peyrusse Vieille	0.050 0.025	0.152 0.143	0.059 0.041	0.084 0.091	0.062 0.047	0.037 0.035	0.117 0.094	0.145 0.158	-	-	0.006 0.006	0.036 0.026
N-HEXANAL												
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
La Tardière	0.019 0.008	0.047 0.037	0.057 0.053	-	0.097 0.085	-	0.064 0.065	0.068 0.053	0.029 0.029	-	0.030 0.006	0.025 0.024
Peyrusse Vieille	0.041 0.043	0.069 0.067	0.041 0.037	0.037 0.028	0.104 0.062	0.041 0.052	0.048 0.006	0.020 0.006	-	-	0.006 0.006	0.012 0.006
2-OXOPROPANAL												
	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC
La Tardière	0.046 0.044	0.122 0.129	0.115 0.098	-	0.362 0.098	-	0.084 0.042	0.191 0.056	0.081 0.031	-	0.032 0.026	0.017 0.007
Peyrusse Vieille	0.050 0.035	0.090 0.073	0.063 0.064	0.019 0.007	0.040 0.025	0.186 0.140	0.199 0.191	0.280 0.296	-	-	0.014 0.006	0.020 0.006

Appendix B

Time series of VOC measured in 2011

Explanations and synonyms to component names

ethyne:	acetylene
butane:	n-butane
isobutane:	i-butane
pentane:	n-pentane
isopentane:	i-pentane
hexane:	n-hexane
methanal:	formaldehyde
ethanal:	acetaldehyde
propanone:	acetone
2-propenal:	acrolein
2-butanone:	methyl ethyl ketone
3-buten-2-one:	methyl vinyl ketone
2-methyl propenal:	methacrolein
benzenecarbaldehyde:	benzaldehyde
ethanedial:	glyoxal
2-oxoproanal:	methyl glyoxal

