EMEP/CCC-Report 4/2018

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

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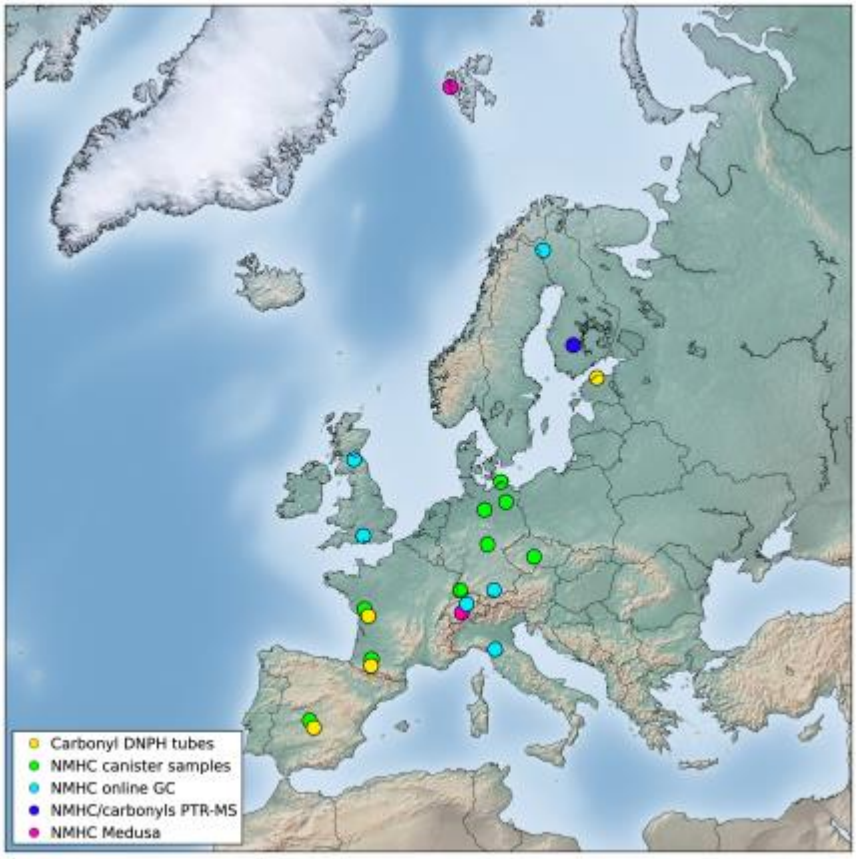

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VOC measurements 2016

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Summary

This report presents VOC measurements carried out during 2016 at EMEP monitoring sites. In total, 19 sites reported VOC data from EMEP VOC sites this year. Some of the data sets are considered preliminary and are not included in the report.

The monitoring of NMHC (non-methane hydrocarbons) has become more diverse with time in terms of instrumentation. Starting in the early 1990s with standardized methods based on manual sampling in steel canisters with subsequent analyses at the lab, the methods now consist of a variety of instruments and measurement principles, including automated continuous monitors and manual flask samples. For oxygenated VOCs (OVOCs), sampling in DNPH-tubes with subsequent lab-analyses is still the only method in use at EMEP sites.

Within the EU infrastructure project ACTRIS-2, data quality issues related to measurements of VOC have been an important topic. Many of the institutions providing VOC data to EMEP have participated in the ACTRIS-2 project, either as formal partners or on a voluntary basis. Participation in ACTRIS-2 has meant an extensive effort with data checking including detailed discussions between the ACTRIS community and individual participants. There is no doubt that this extensive effort has benefited the EMEP program and has led to improved data quality in general.

Comparison between median levels in 2016 compared to the medians of the previous 10-years period, revealed a similar north-to-south pattern for several species.

Changes in instrumentation, procedures, station network etc. during the last two decades make it difficult to provide a rigorous and pan-European assessment of long-term trends of the observed VOCs. In this report we have estimated the long-term trends in NMHC over the 2000-2016 period at six selected sites by two independent statistical methods. These estimates indicate marked differences in the trends for the individual species. Small or non-significant trends were found for ethane over this period followed by propane which also showed fairly small reductions. On the other hand, components linked to road traffic (ethene, ethyne and toluene) showed the strongest drop in mean concentrations, up to 60-80% at some stations. The trend in n-butane was between these two groups of species with an estimated drop in the annual mean concentration of 20-40% over the 2000-2016 period.

VOC measurements 2016

1. Introduction

1.1 Historical background

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 centred at noon GMT at each station. 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.

VOC measurements within EMEP started with the collection of grab samples of light hydrocarbons in mid 1992 and measurements of carbonyls in 1993. Initially, five stations were included in the monitoring programme: Rucava (LV0010), Košetice (CZ0003), Waldhof (DE0002), Tänikon (CH0032) and Donon (FR0008). Since then, the number and selection of VOC measurement sites have changed several times.

EMEP VOC measurements are reported annually, and presented in reports for consideration by EMEP-TFMM and the EMEP Steering Body. Previous results from the EMEP VOC programme have been presented in annual reports (e.g. Solberg, 2013 and references therein). 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).

VOC-data from the EMEP-network have been published and documented in numerous publications, e.g. Waked et al. (2016), Hellen et al. (2015), Hoerger et al. (2015), Malley et al. (2015), Solberg (2013), Tørseth et al. (2012), Worton et al. (2012), Sauvage et al. (2009), Plass-Dülmer et al. (2009), Plass-Dülmer et al. (2006), Hakola et al. (2006), Borbon et al. (2004), Solberg et al. (2001), Solberg et al. (1996).

1.2 Underlying protocols for VOC

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: sulfursulfursulfur, 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 sulfursulfursulfur 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 limits 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. An important difference from the previous protocol is that the emission ceilings now are given as percentage reductions from 2005 to 2020 and thereafter. Furthermore, $PM_{2.5}$ 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).

1.3 Cooperation with other bodies – GAW and ACTRIS

At some stage, initiatives were taken to increase the cooperation and exchange of VOC data between GAW (Global Atmospheric Watch) and EMEP. Harmonization of data quality objectives (DQOs) and using a common audit questionnaire were recommended, and it was 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 GAW species and their DQOs are given in Table 1 together with the original list of so-called required and desirable compounds within EMEP as defined at the Lindau workshop in 1989 (EMEP/CCC, 1990). Table 1 also lists the ACTRIS species and their DQOs, as explained below.

Most of the GAW compounds are already part of the EMEP VOC programme with some exceptions: Alcohols, terpenes, DMS (dimethyl sulfide) and acetonitrile are not part of the original EMEP VOC programme. 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 instrumentation than presently used for the hydrocarbons and carbonyls.

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

Within the EU FP7 infrastructure project ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure), data quality issues related to measurements of VOCs were important topics. ACTRIS, the European Research Infrastructure for the observation of Aerosol, Clouds, and Trace gases was launched as a EU FP7 project in 2011 and includes a large number of partners with experience in VOC monitoring, including most of EMEP laboratories. The aim was to evaluate the performance, repeatability and uncertainty of the present NMHC monitoring, as well as to develop guidelines and data quality objectives for the monitoring. Highly ambitious DQOs were defined for a number of individual species as shown in Table 1.

Furthermore, GAW are in the process of adopting the ACTRIS DQOs and are introducing the terminologies “GAW basic performance” and “GAW target performance”, the latter corresponding to the ACTRIS DQOs as given in Table 1.

Table 1: Original list of required and desirable VOCs within EMEP. The GAW priority species with required accuracy/precision and the ACTRIS priority species with required uncertainty/repeatability are also listed.

	EMEP required	EMEP desirable	GAW priority[1]		ACTRIS priority[2]	
Alkanes			accuracy	precision	uncert.	repeat.
Ethane	X		10%	5%	5%	2%
Propane	X		10%	5%	5%	2%
n-butane	X		10%	5%	5%	2%
i-butane	X		10%	5%	5%	2%
n-pentane	X		10%	5%	5%	2%
i-pentane	X		10%	5%	5%	2%
n-hexane		X			5%	2%
i-hexanes		X			5%	2%
n-heptane		X			5%	2%
i-heptanes		X			5%	2%
n-octane		X			5%	2%
i-octanes					5%	2%
Cyclohexane					5%	2%
Alkenes					5%	2%
Ethene	X				5%	2%
Propene	X				5%	2%
butenes		X			5%	2%
pentenes		X			5%	2%
1,3-butadiene					5%	2%
Isoprene	X		20%	15%	5%	2%
Alkynes					5%	2%
Acetylene	X		15%	5%	5%	2%
Propyne					5%	2%
Styrene		X			5%	2%
Aromatics					5%	2%
Benzene	X		15%	10%	5%	2%
Toluene	X		15%	10%	5%	2%
o-xylene	X				5%	2%
m,p-xylene	X				5%	2%
Ethylbenzene	X				5%	2%
trimethylbenzenes	X				5%	2%
propylbenzenes		X			5%	2%
Ethyltoluenes		X			5%	2%
Carbonyls					5%	2%
Formaldehyde	X		20%	15%		
Acetaldehyde	X					
Propionaldehyde		X				
Acetone	X		20%	15%		
Methylethylketone		X				
Methylvinylketone		X				
Other						
Monoterpenes			20%	15%		
Acetonitrile			20%	15%		
Methanol			20%	15%		
DMS			20%	15%		

[1] Accuracy = 20 ppt, Precision = 15 ppt if level < 0.1 ppb
[2] Uncertainty = 5 ppt, Repeatability = 2 ppt if level < 0.1 ppb

2. Status of the measurement programme in 2016

2.1 The station network

The locations of the EMEP monitoring sites for VOC in 2016 are shown in Figure 1 and an overview of the measurement programme and the responsible laboratories is given in Table 2. In total, 19 measurement sites are included in the list. Some data, as explained later, are not included in this report since they are still regarded as preliminary, either due to data format technicalities, or due to unresolved questions relating to data quality and filtering (flagging local influences).

The measured VOCs consist of two groups of species; non-methane hydrocarbons hereafter named NMHC and oxygenated species hereafter named OVOC. Monitoring of NMHC is carried out at all sites except Lahemaa, Estonia, whereas carbonyls are measured at a few sites only; two sites in France, one in Spain and at the Estonian site. As seen from Figure 1 and Table 2 the two French sites and the Spanish site measure both NMHC and OVOC.

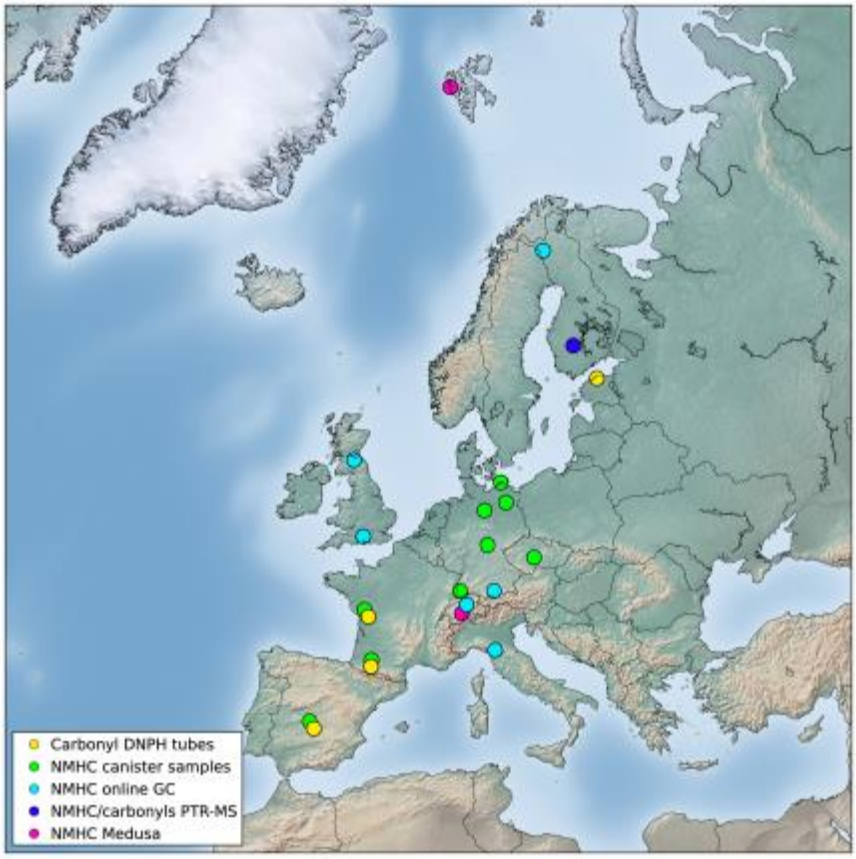

Figure 1: Monitoring sites for VOC in 2016.

The NMHC monitoring at EMEP sites has become more diverse with time in terms of instrumentation. Starting in the early 1990s with a standardized method based on manual sampling in steel canisters and subsequent lab analyses, the methods now

comprise a variety of instruments and measurement principles, including PTR-MS (Hyytiälä), Medusa monitors (Zeppelin Mountain and Jungfraujoch) and specialized online GC monitors for hydrocarbons. For OVOCs the original method based on sampling in DNPH adsorption tubes with subsequent lab analyses is still the only method used at the EMEP sites. It should be said that the PTR-MS could also be used for monitoring of certain OVOCs, but no such data were reported in 2016.

Although a substantial number of sites has contributed to the EMEP VOC programme since the early 1990s, very few sites have long and continuous time series. This pose a problem for making reliable long-term trend assessments of VOCs at European background sites. Additionally, shifts in instrumentation imply possible breaks in the time series. At some sites these shifts are a matter of upgrading the GC monitor, with minor effects on the measured values, while at other sites they represent significant breaks in the data time series.

As given in Table 2, some of the data series were considered questionable and – in one case – not included in this report. NMHC data from San Pablo (ES0001) have for several years shown substantial differences compared to the expected levels and compared to the other sites. The differences become particularly evident when inspecting ratios of certain NMHCs; e.g. an annual mean level of n-hexane being almost two orders of magnitude larger than at the other sites whereas the annual mean of ethyne is only 10% of that seen at the other sites. There has been a dialogue between NILU and the data provider in Spain on this issue and the status of the data is still regarded unsettled and hence the data are not included in this report.

In the UK, NMHC data have previously been monitored at two EMEP sites – Harwell (GB0036) and Auchencorth Moss (GB0048). In January 2016, the hydrocarbon monitoring was moved from Harwell to Chilbolton Observatory (GB1055) approximately 50 km to the south.

The data from Harwell and now Chilbolton observatory, located in southern England, clearly reflect the influence of populated areas in the whole of southern England, including e.g. road traffic emissions, and include a number of short-term spikes in the data.

The NMHC data for Auchencorth Moss in Scotland, a rural location around 20 km south of Edinburgh also show very spiky time series with peak values of propane, n-butane and other species resembling what is seen at street level in major cities. Furthermore, the ratios between pairs of components as mentioned above for San Pablo, also differ substantially at Auchencorth Moss compared to other sites. Based on this, together with the fact that there has been little response to the QA/QC issues raised within ACTRIS-2 to the Auchencorth Moss data, all data from this station are regarded preliminary and unsettled. It seems, though, that the median values are less affected by these issues, and thus the data have been included to some extent in the present report.

Table 2: VOC monitoring at EMEP sites in 2016. The columns give the station names, site code, and the sampling frequencies for hydrocarbons (HC) and carbonyl compounds (Carb). The institute responsible for the chemical analyses is also given. Whether the station is part of the ACTRIS-2 project is also indicated.

Station	Code	HC[1)]	Institute[2)]	Carb[1)]	Method	ACTRIS	Comment
Zeppelin Mtn.	NO0042	Cont.	NILU	-	MEDUSA	y	
Pallas	FI0096	Cont.	FMI	-	GC/MS	y	
Hyytiälä	FI0050	Cont	UHel	-	PTR-MS.	y	
Lahemaa	EE0009	-	EERC	Reg.	DNPH tubes	n	Very few valid data
Auchencorth Moss	GB0048	Cont.	Ricardo	-	GC/MS	y	Extreme spikes in data. Need further evaluation
Chilbolton Obs.	GB1055	Cont.	Ricardo	-	GC/MS	y	Strong local influences
Waldhof	DE0002	Reg.	UBA	-	Flask samples	y[3)]	Twice//week
Schauinsland	DE0003	Reg.	UBA	-	"	y[3)]	"
Neuglobsow	DE0007	Reg.	UBA	-	"	y[3)]	"
Schmücke	DE0008	Reg.	UBA	-	"	y[3)]	"
Zingst	DE0009	Reg.	UBA	-	"	y[3)]	"
Hohenpeissenberg	DE0043	Daily	DWD	-	GC/FID	y	2/day (noon, midnight)
Košetice	CZ0003	Reg.	CHMI	-	Flask samples	y	Twice/week
Jungfraujoch	CH0001	Cont.	EMPA	-	MEDUSA	y	
Rigi	CH0005	Cont.	EMPA	-	GC/MS	y	
Peyrusse Vieille	FR0013	Reg.	EMD	Reg.	Flask/DNPH samples	y	NMHC 2/week. OVOC 1/week
La Tardière	FR0015	Reg.	EMD	Reg.	"	y	"
Mt. Cimone	IT0009	Cont.	UU	-	GC/MS	y	
San Pablo	ES0001	Reg.	MMA	Reg.	Flask/DNPH samples	n	Twice/week. Prelim. NMHC data not included in the report.

1) Reg. = regularly, Scat. = scattered, n.m. = not measured., n.a. = not yet analysed, cont. = Continuous
2) CHMI = Czech Hydrometeorological Institute
DWD = Deutscher Wetterdienst
EERC = Estonian Environmental Research Centre
EMD = Ecole des Mines de Douai (France)
EMPA = Swiss Federal Lab. for Materials Testing and Research
FMI = Finnish Meteorological Institute
UHel = Univ. Helsinki
UBA = Umweltbundesamt (Germany)
UU = University of Urbino
MMA = Minestrio de Medio Ambiente
3) Participated voluntarily in ACTRIS-2 without being a formal partner

2.2 Analytical procedures, quality control and intercomparisons

Quality control of the VOC measurements includes QA/QC procedures at all stages from sampling to chemical analyses and integration. The QA procedures for the original EMEP methods (manual sampling of NMHC in stainless steel canisters and OVOC in DNPH adsorption tubes with subsequent analyses in the lab) are described in the EMEP manual (EMEP/CCC, 2014) and are more or less identical to the original manual description given in EMEP/CCC 1995.

Measurement guidelines for the original EMEP method based on manual sampling of NMHCs has furthermore been provided in detail by GAW (GAW, 2012). A review of the various methods for VOC monitoring within GAW has also been prepared based on a GAW expert workshop in 2006 (WMO/GAW, 2006).

As mentioned, the ACTRIS consortium and the subsequent ACTRIS-2 consortium played a central role in the quality control of data from the regular monitoring by the laboratories of the particpating institutes. A comprehensive Standard Operation Procedures (SOP) manual for VOCs has alsobeen developed as part of the project.

VOC data from ACTRIS-stations are presented by representatives from each institution and discussed in detail at dedicated workshops annually (normally in May the following year). Associated institutions not being formal ACTRIS-2 partners are invited to take part in this, and UBA (Umweltbundesamt) in Germany has used this opportunity.

Based on statistical tools developed within ACTRIS and ACTRIS-2, potential outliers and errors in the data were discussed, and recommendations for database flagging were agreed on through discussion at the workshop. EMPA, the ACTRIS-2 task leader for VOC, had a key role in this process. Detailed inspection of the data has furthermore been done by NILU in parallel with the tools developed at EMPA. Dialogue between EMPA, NILU and the data providers has been essential in this work and a web based issue tracker has been developed and used in the process.

The whole QA/QC process has become fairly complex and demanding but is now (by 2018) “on track” and goes much more smoothly than in the first years. The procedures developed within ACTRIS and ACTRIS-2 follow somewhat in the footsteps of the AGAGE project for greenhouse gases and ozone depleting substances (e.g. Prinn et al., 2018). The elaborate QA work on VOC provided by ACTRIS-2 and EMPA in particular, has meant a significant improvement in the general data quality. Hopefully, these procedures and routines will incorporated into the parts of the EMEP VOC program and stations that are not formally part of ACTRIS-2.

In conjunction with EBAS, the templates for data submission were further developed to the requirements of GAW, EMEP and WIGOS. The data flow for VOC data collected at EMEP/ACTRIS stations are shown in Figure 2.

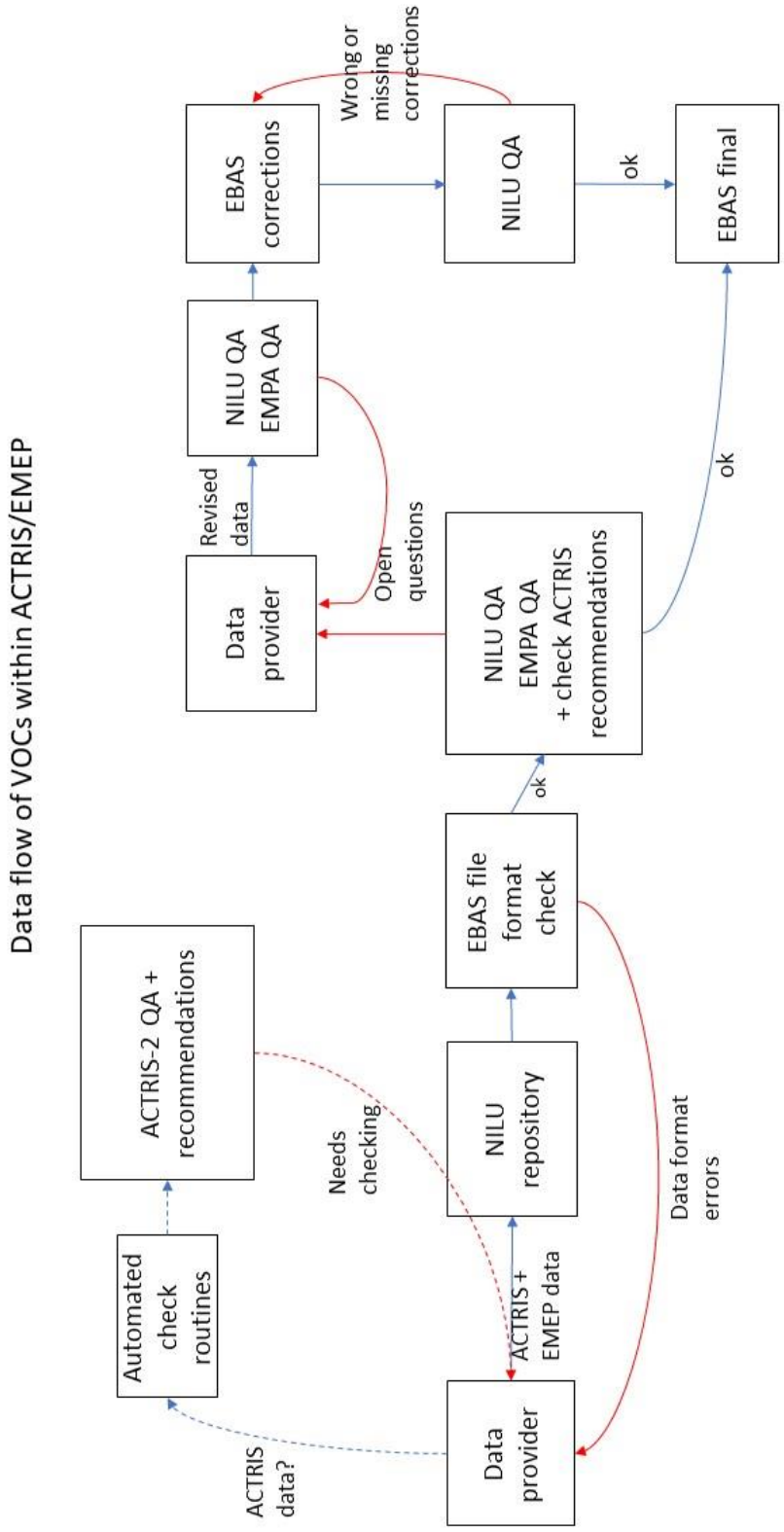

Figure 2: Flow diagram for the VOC data submitted within ACTRIS/EMEP.

2.2.1 Intercomparisons

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

Since then, various intercomparisons for VOCs have been carried out, e.g. through the NOMHICE (Nonmethane Hydrocarbon Intercomparison Experiment) (Apel, 2003, and references therein) and AMOHA (Accurate Measurements of Hydrocarbons in the Atmosphere) (Slemr et al., 2002; Plass-Duelmer et al., 2006) projects, with participation from a large number of laboratories in Europe and elsewhere. A major part of the AMOHA 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 ±25% of the median for the lighter alkanes. For some aromatics and unsaturated hydrocarbons as well as the C_6-C_7 alkanes a large spread in the values was seen, indicating measurement difficulties with these compounds. The spread in the results was, however, much less for those laboratories using an NPL standard for calibration (Aas et al., 2001). Thus, it was 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.

Details of the ACTRIS NMHC intercomparison can be found in Hoerger et al. (2015). The intercomparison covered a list of 34 NMHCs, including C_2-C_8 alkanes, C_2-C_5 alkenes, five aromatics and two alkynes. One canister with a mixture of 30 NMHCs at 1 ppb level in N_2 and one canister with whole air sampled in an suburban area (Dübendorf, Switzerland) were distributed to all participating laboratories for analysis. For calibration, the laboratories were asked to use their own certified multicomponent standards, traceable to the GAW scale. Three laboratories served as reference labs, analyzing the starting cylinders before and after the exercise: The WCC-VOC (World Calibration Centre for VOC, Karlsruhe Institute of Technology, Garmisch-Partenkirchen), DWD (Deutscher Wetterdienst at Hohenpeissenberg) and EMPA (Swiss Federal Laboratories for Materials Science and Technology), Switzerland.

The intercomparison showed the best results for the gas mixture in N_2 and for the lighter alkanes. In comparison, the results were clearly poorer for the whole air sample. C_4-C_5 alkenes and C_7-C_8 species (alkanes and aromatics) were the most problematic species. For all species/laboratories, almost 62% of the results from the N_2 canisters fell within the 5% DQO and 90% within the former 10% DQO of GAW. For the real air samples, larger and more frequent deviations were found. Only 50% of the results were within the ACTRIS 5% DQO and 79% within the 10% group

As seen in previous intercomparison studies (e.g. AMOHA, Plass-Duelmer et al. (2006)), the type of calibration standard is important for the performance of the

laboratory. In the ACTRIS study, it turned out that systems based on direct calibrations with standards in the ppb-range performed better than those based on a two-step calibration using more concentrated standards. Furthermore, ethyne was a problem in several systems, and direct calibration of ethyne turned out to be essential for the result. Additionally, almost all the participating laboratories/instruments showed indications of losses of the C_7-C_8 aromatics, most probably due to adsorption effects.

In general, the best results were provided by GC-FID instruments. GC-MS systems also delivered good results; however they require more frequent calibrations since they are less stable. The only commercially available system, the Perkin Elmer Online Ozone Precursor Analyzer, provided reasonable results although not among the best. A main conclusion from the ACTRIS study is that the very ambitious ACTRIS DQOs for NMHCs could be met. It will, however, require experienced personnel, well-characterized instrumentation and detailed procedures for quality control at all stages.

The ACTRIS project also included an intercomparison for NMHCs in which 18 laboratories with 23 different GC instruments participated and the results were recently published (Hoerger et al., 2015). In addition, a side-by-side intercomparison for OVOCs (aldehydes and ketones) was carried out within ACTRIS at Hohenpeissenberg, with synthetic test mixtures and ambient air. The results of this exercise are, however, not yet published.

In addition to the intercomparison for NMHCs discussed above, a side-by-side intercomparison for oxygenated VOC (OVOC) was carried out within ACTRIS at Hohenpeissenberg, with synthetic test mixtures and ambient air. The results of this exercise are, however, not yet published.

3. VOC concentrations in 2016

3.1 General levels

Time series of the diurnal means of all compounds at all stations during 2016 are given in the Appendix. Figure 3 shows the spread of data values for each station and NMHC species in 2016 in box and whisker plots, and the annual median values based on the previous 10 years of data (2006-2015) are included for comparison for stations that had such data. The sites are arranged from north to south going from left to right in the panels. Thus, the panels in Figure 3 indicate both the north-to-south differences, the deviation of the 2016 concentration levels relative to the previous 10 years climatology as well as the spread in 2016 data at each site separately.

For some species (e.g. ethane, propane, n-butane and 2-methylpropane) there is a striking similarity between the variation in median values in 2016 compared to the 10 year climatology. For other species, particularly toluene, benzene, n-pentane and n-hexane there are substantial differences between the 2016 levels and the 10 year climatology. Note that previous monitoring problems at UBA's sites in Germany explain parts of this – most evident for toluene showing very high median levels for the 2006-2015 period that presumably are not real.

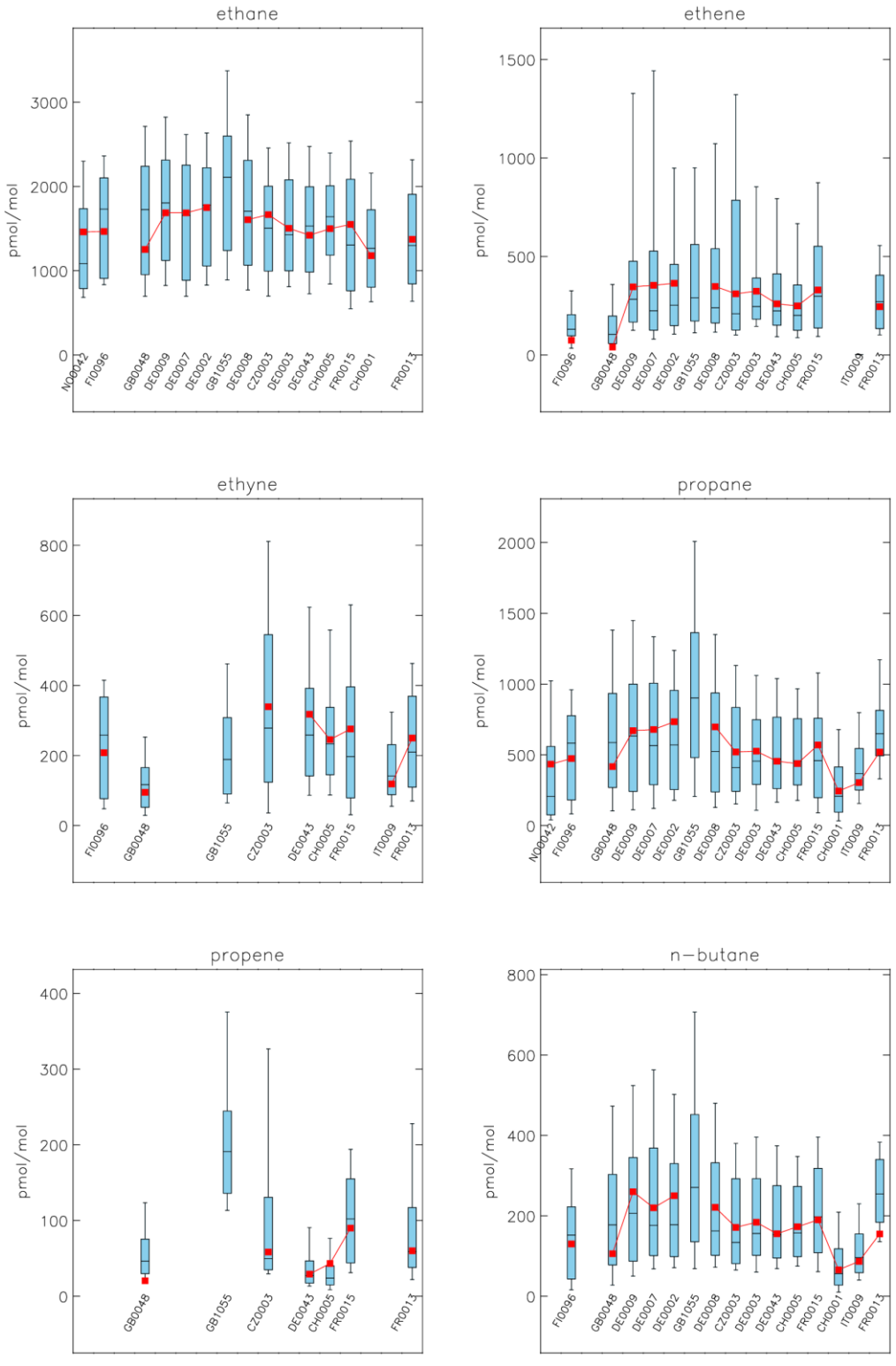

Figure 3: Box- and whisker-diagrams for light hydrocarbons based on all measurements in 2016. The boxes enclose the 25- and 75-percentile with the median marked inside. The whiskers extend out to the 9th and 91th percentile. The red squares connected with a red line mark the medians based on all data for the previous 10 years (2006-2015).

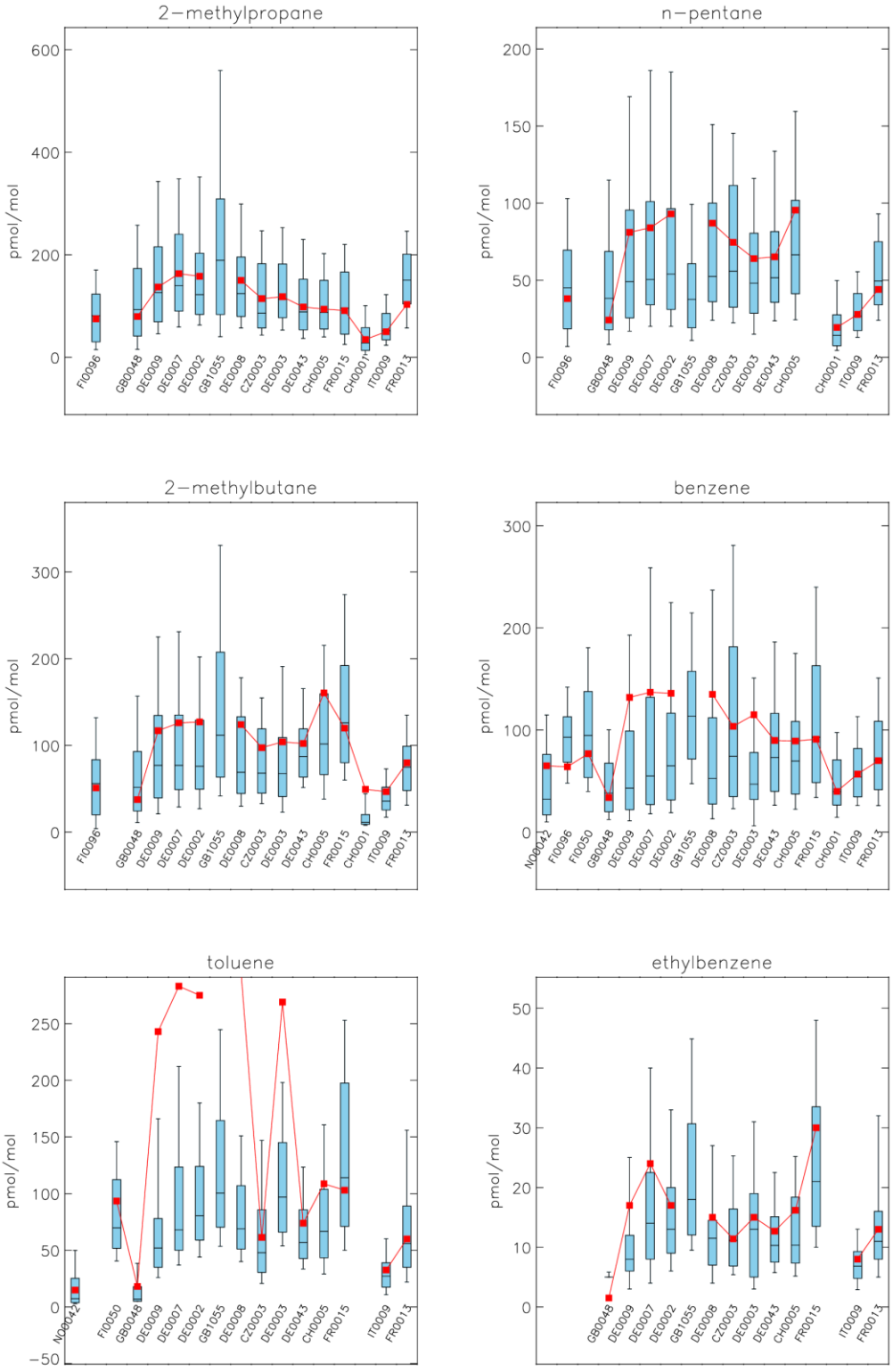

Figure 3. (cont.)

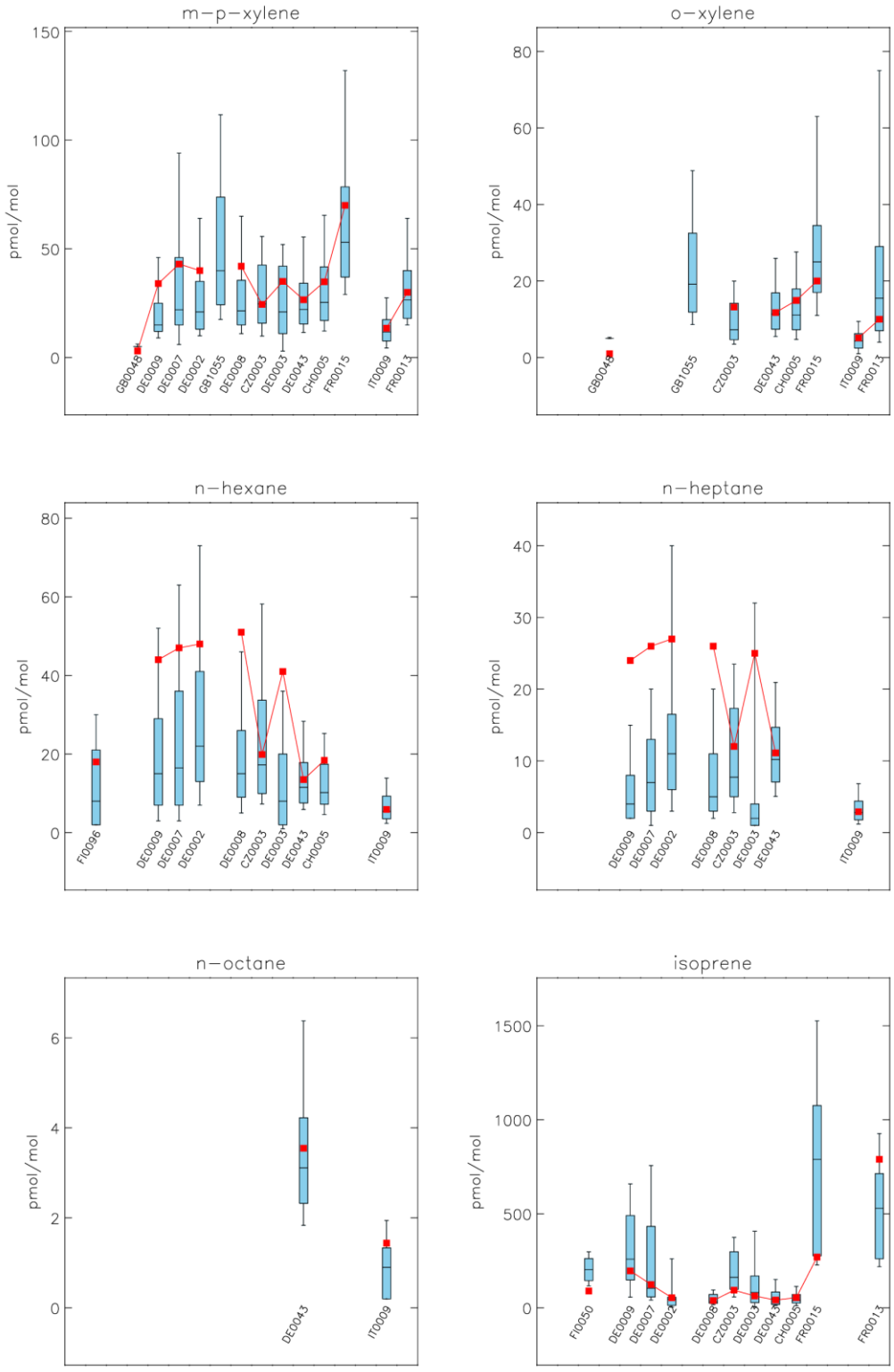

Figure 3. (cont.). Note that for isoprene only data for June-August (whole day) were used.

Similar box and whisker plots for a number of carbonyls based on the whole year of monitoring are shown in Figure 4. Only four stations contributed with such data in 2016 and one of these (Lahemaa) had so few real (non-missing) data, that the box and whiskers could not be made.

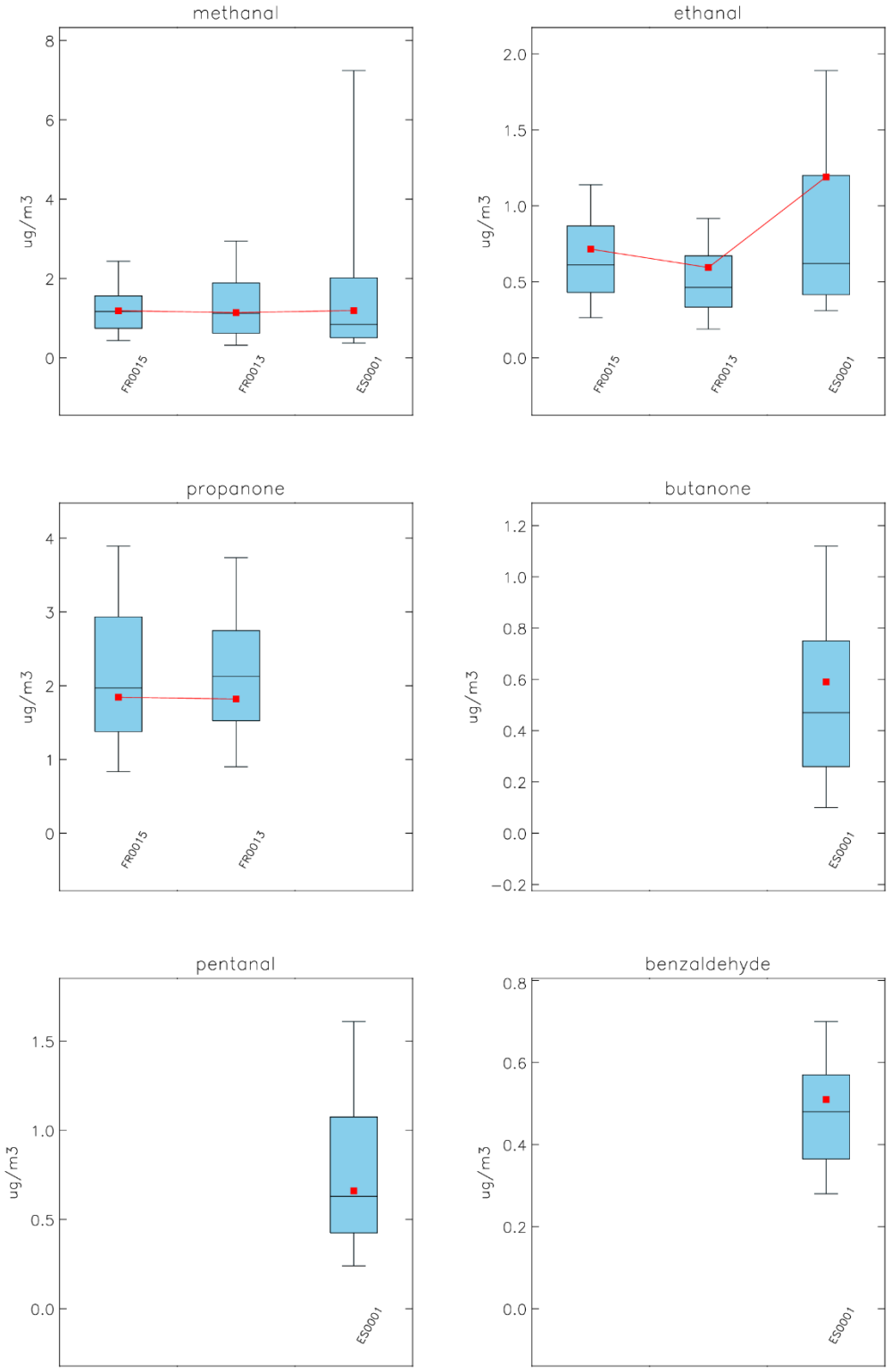

Figure 4: Box- and whisker-diagrams for carbonyls based on all measurements in 2016. The boxes enclose the 25- and 75-percentile with the median marked inside. The whiskers extend out to the 9th and 91th percentile. The red squares connected with a red line mark the medians based on all data for the previous 10 years (2006-2015).

3.2 Regional distribution of VOC

Figure 5 shows maps with the stations' annual median concentrations of light hydrocarbons in 2016. Note that since the steel canisters are all sampled at daytime (normally at noon), a bias could be inherent in these plots when compared with the 24 h daily average values from online GCs. A bias for other species is also likely to a varying extent. The mountain stations (Hohenpeissenberg and Mt Cimone) are influenced by the diurnal venting of the planetary boundary layer, and will receive upslope polluted air masses at daytime and cleaner free tropospheric air at night.

Similar maps for selected carbonyl species based on data from the whole year are given in Figure 6. The number of monitoring sites for carbonyls are much less than for NMHC and the sampling frequency is in general somewhat poorer. Particularly poor data coverage was seen at the site in Estonia (Lahemaa) and thus the annual medians calculated in Figure 6 were based on just a very few values.

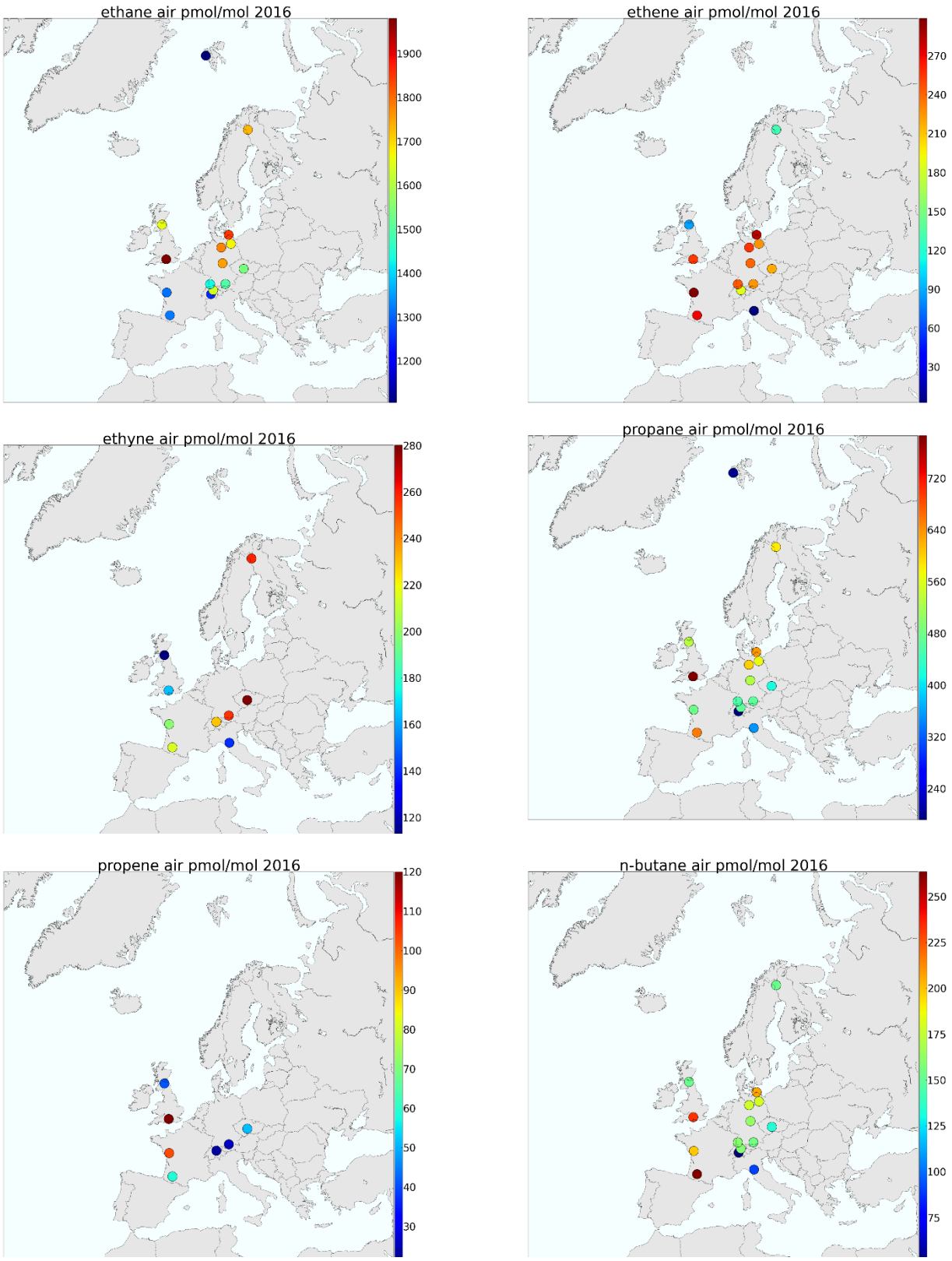

Figure 5: Annual median concentration of NMHCs in 2016.

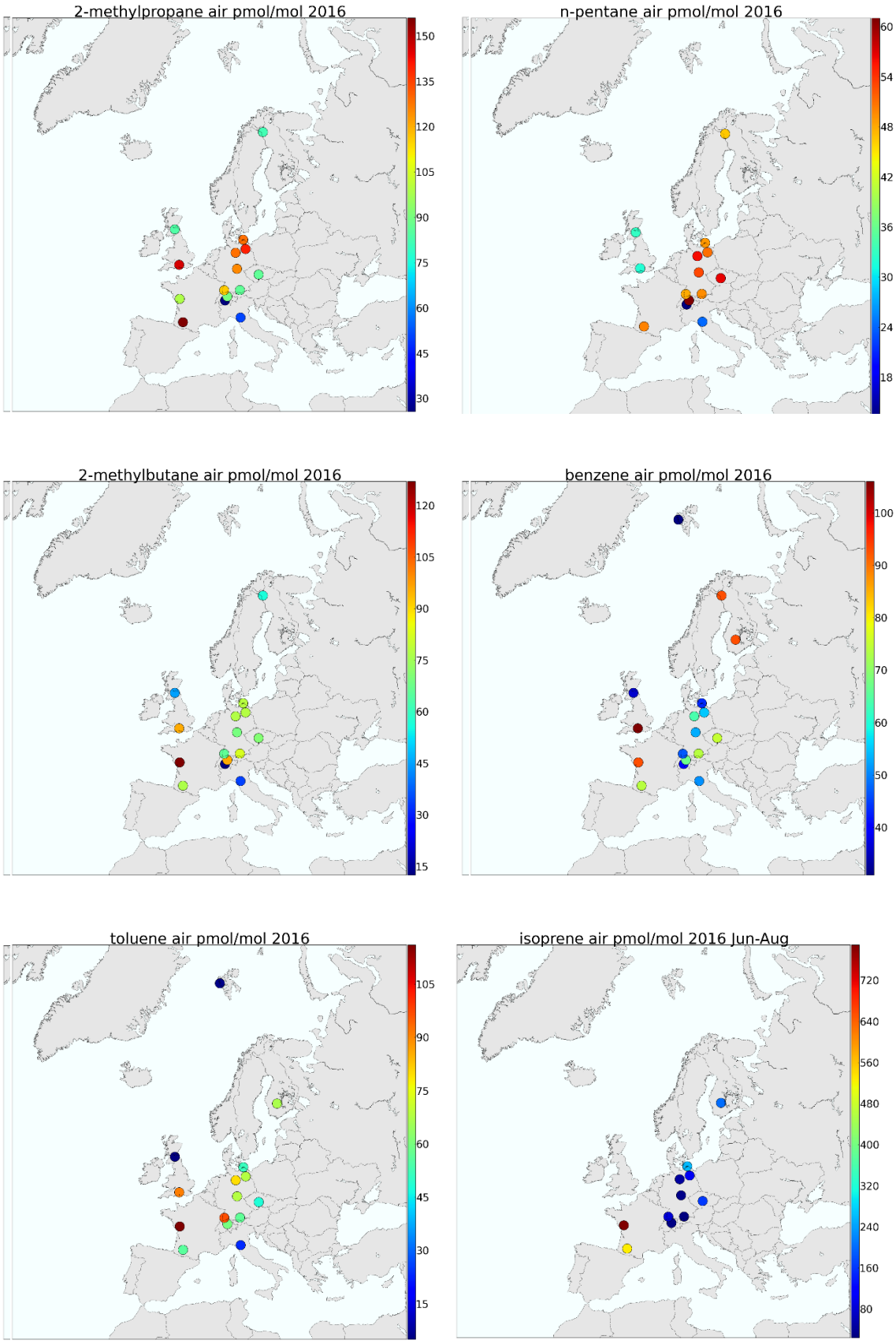

Figure 5 (contd.). Note that for isoprene the summer median (Jun-Aug) is shown.

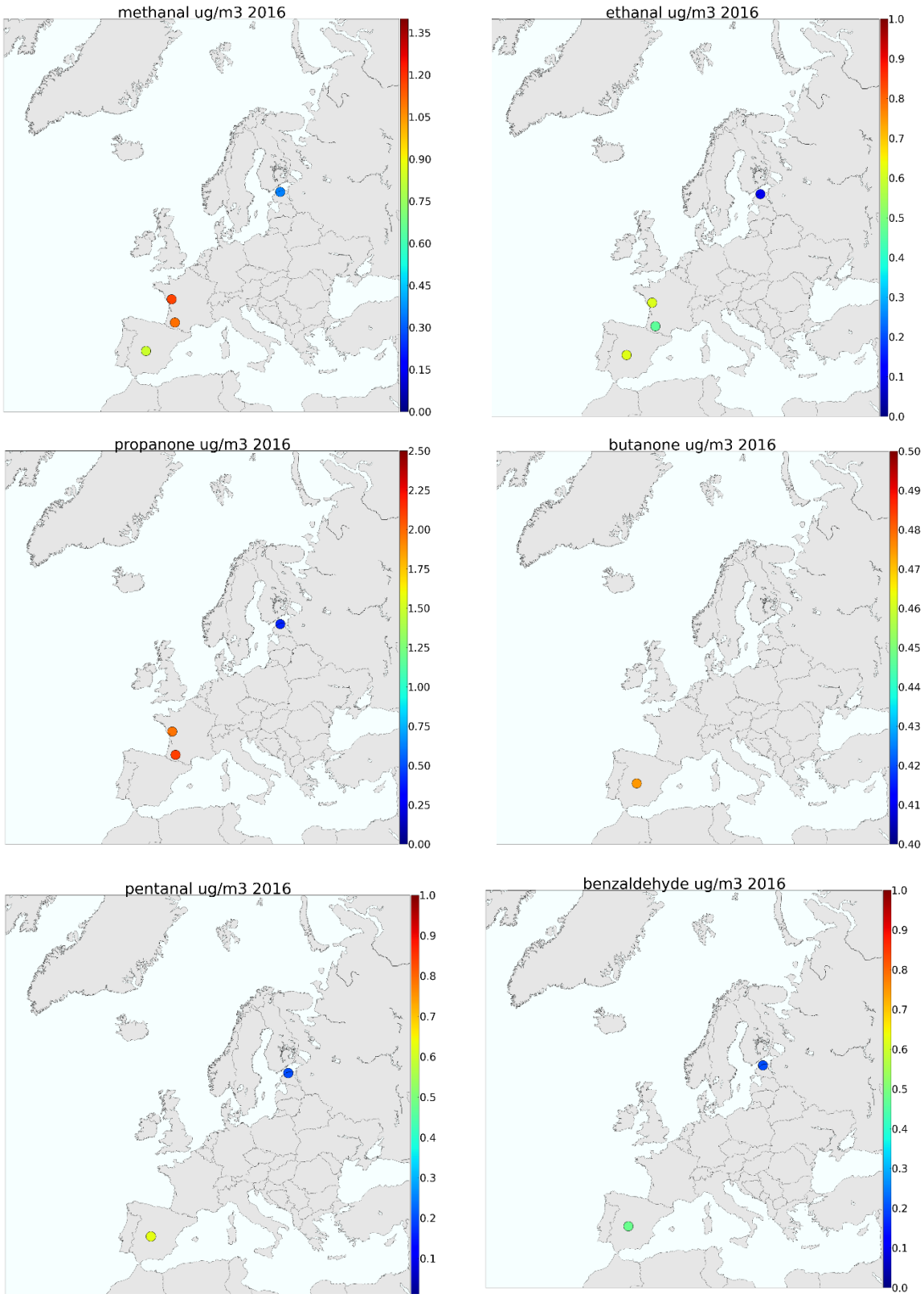

Figure 6: Annual median concentration of carbonyls in 2016.

4. Long-term trends in VOC

According to the official emission data, there have been substantial reductions in anthropogenic emissions of VOCs in the last two decades, of the order of 40-50% since 1990, and with an overall reduction of 27% from 2000 to 2015 for the EMEP region as a whole (Fagerli et al., 2017). There are substantial regional and national differences in these emission trends within the EMEP domain, particularly when comparing the western and eastern part of the region (Fagerli et al., 2017; Fagerli et al., 2018).

Declines in the measured concentrations of hydrocarbons have been reported from suburban/urban sites at several locations. Based on a network of high-frequency continuous monitoring of C_2-C_8 hydrocarbons in the UK, mostly at urban/suburban locations, Derwent et al. (2014) found substantial declines in concentrations with present levels close to an order of magnitude below the levels in the early 1990s. They estimated exponential declines in concentrations of the order of -11% y^{-1} to -22% y^{-1} for the period 1994-2012. They also found a marked difference between ethane and propane which showed relatively stable levels, while other alkanes showed pronounced declines.

Long-term monitoring data from an urban network in Switzerland (Hüglin, pers. comm.) also show strong declines in the concentrations of NMHCs and OVOCs from the start of the 1990s to the present.

Various trend studies have been carried out for VOC data from EMEP rural sites as well. Sauvage et al. (2009) found clear decreases at the French EMEP sites of most NMHCs. Ethane was an exception to this and showed more stable levels.

Analyses of the twenty years NMHC monitoring at the EMEP/GAW site Pallas in Northern Finland revealed, however, a significant downward trend only for ethyne (Hellen et al., 2015). They concluded that other source regions than the EU were dominating the NMHC levels at the site. Based on source area estimates they found that the Eastern parts of the continent were the main source regions for high concentrations at Pallas.

A simple 1:1 relationship between observed VOC concentrations at rural background sites and the overall European emission numbers is 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.

Furthermore, various methods used for trend analyses in terms of mathematical method, selection of time periods and stations etc could give different results. In the following we have looked at daily measurements of selected VOCs at six EMEP sites with long term data over the period 2000-2016. Based on these data, best-fit seasonal trend curves were calculated by a non-linear least squares fit method by use of a standard statistical package (Markwardt, 2009) to the following equation:

$$c(t) = \left[a_0 + a_1(\sin(2\pi(t - a_2))\right] exp[a_3(t - t_0)] \qquad (Eq.\ 1)$$

where

c(t) = value at time *t* measured in years

t_0 = 2000

and the coefficients a_0, a_1, a_2, a_3 were determined by the non-linear least squares fit using the iterative algorithm by Markwardt, 2009. It turned out that *Eq. 1* converged for all our cases with less than 10 iterations, even when using very strict tolerance criteria for the iterations. In addition to best fit values for the coefficients a_0, a_1, a_2, a_3, the algorithm also provided confidence intervals for each of the coefficients.

Eq 1 represents a simple seasonal cycle with a mean level a_0, amplitude a_1 and seasonal phase displacement a_2 that change exponentially over time 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 ensure positive solutions. the least squares fit was applied to log-transformed data, i.e. in *Eq 1* we defined c(t) = log(cc(t)), where cc(t) is the actual daily observed mixing ratio in pmol mol^{-1}. This also implies a weighting of the data, increasing the weight of the low-level concentrations relative to the high-level ones. Without such a weighting, the least squares fit would be strongly determined by the highest concentration values that are mostly observed in winter.

For comparison, we included a best fit of the daily data (not log-transformed) to a linear polynomial function as presented by Simmonds et al (2006) for AGAGE trace gases:

$$f(t) = a + b \cdot N \cdot P_1\left(\frac{t}{N} - 1\right) + \frac{1}{3} \cdot d \cdot N^2 \cdot P_2\left(\frac{t}{N} - 1\right)$$
$$+c_1 \cdot \cos(2\pi t) + s_1 \sin(2\pi t) + c_2 \cos(4\pi t) + s_2 \sin(4\pi t) \qquad (Eq.\ 2)$$

where P_1 and P_2 are the Legendre polynomials of order 1 and 2, *t* is time measured in years (*t* = 0 for year 2000) and 2N is the total number of years. The coefficient *a* defines the average mole fraction, while coefficients *b* and *d* define the linear trend and the acceleration in trend, respectively. The coefficients *c* and *s* define the annual cycle in concentration. We note that the polynomial function in *Eq 2* could lead to negative values for species that are observed at low concentration levels, and that is indeed seen in the results below.

Figure 7 - Figure 13 shows the daily concentrations (blue marks) together with the curves fitted to *Eq 1* in red and to *Eq 2* in black for six selected sites: Pallas, Kosetice, Hohenpeissenberg, Rigi, La Tardiere and Peyrusse Vielle, all of which have sufficiently long time series of NMHCs. For the time series where a statistically significant trend was found by *Eq 1*, the percentage change (as given by *Eq 1*) from 2000 to 2016 using 2000 as a reference year is given in the title. We assumed significant trends if $|a_3| > 2\sigma$, where σ is the confidence interval as mentioned above.

Table 3 lists the percentage reductions of these species from 2000 to 2016 as estimated by *Eq 1* and *Eq 2*, respectively. We calculated the total reductions over the 2000-2016 period based on the annual mean values in 2000 and 2016 as calculated directly from *Eq 1* and from *Eq 2*, respectively. For *Eq 1* the reductions were not calculated if $|a_3| < 2\sigma$, and similarly for *Eq 2* ($|b| < 2\sigma$).

The results in Figure 7 - Figure 13 and in Table 3 indicate marked differences in the long-term trends between the individual species. The smallest percentage changes, or most frequent non-significant trends, are seen for ethane and propane. The largest reductions are found for ethyne, benzene and toluene as well as for ethene (depending on site). Furthermore, fairly similar reductions are found with both methods overall although there are differences for certain sites/species. In general, the polynomial fit (*Eq 2*) gave somewhat smaller confidence intervals than the non-linear fit (*Eq 1*) and thus more significant values.

These results indicate that the European background levels of light hydrocarbons have experienced a substantial reduction since 2000 amounting up to 50-80% in the annual means for ethene, ethyne and toluene. The species with the strongest reductions are all linked to emissions from combustion and this likely reflects strong reductions in emissions from road traffic in Europe.

Ethane on the other hand shows either no significant trend or only small changes since 2000, and fairly small changes are found for propane as well. This indicates that emissions linked to the exploration and use of natural gas have not dropped significantly over this period. It should be said that these species also have a long chemical lifetime in the atmosphere, making them significantly dependent on emissions from other continents in the northern hemisphere. For n-butane we estimate a reduction of the order of 20-40% from 2000 to 2016, which is a number in between the number found for the natural gas tracers and the tracers of road traffic emissions.

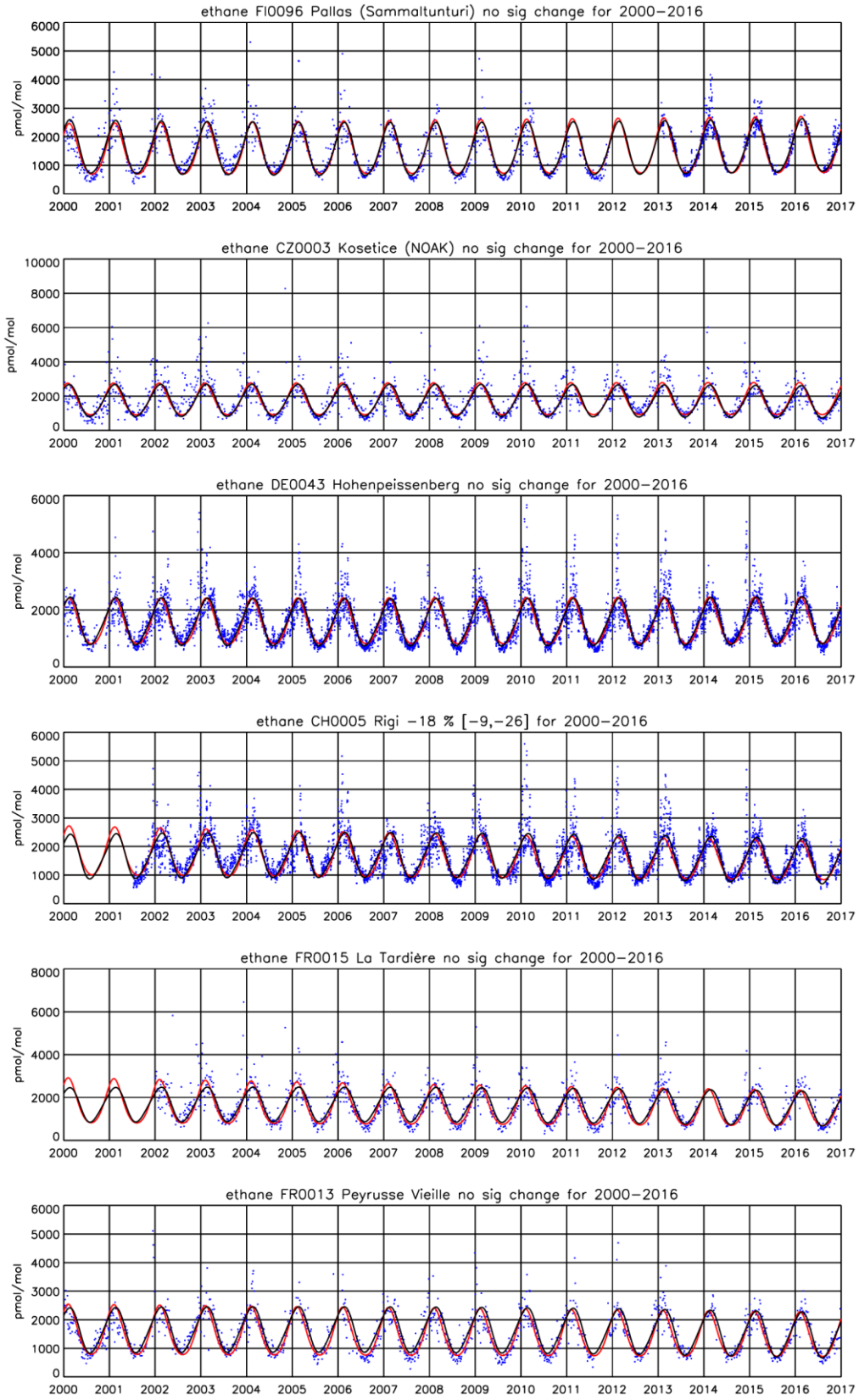

Figure 7: Daily mean ethane concentrations (blue) at six EMEP sites during 2000-2016 together with the estimated seasonal trend curve in red (see text for explanation). Significant trends are given with confidence intervals in the header as percentage change from 2000 to 2016.The black curve shows the curve fit using a polynomial function (see text).

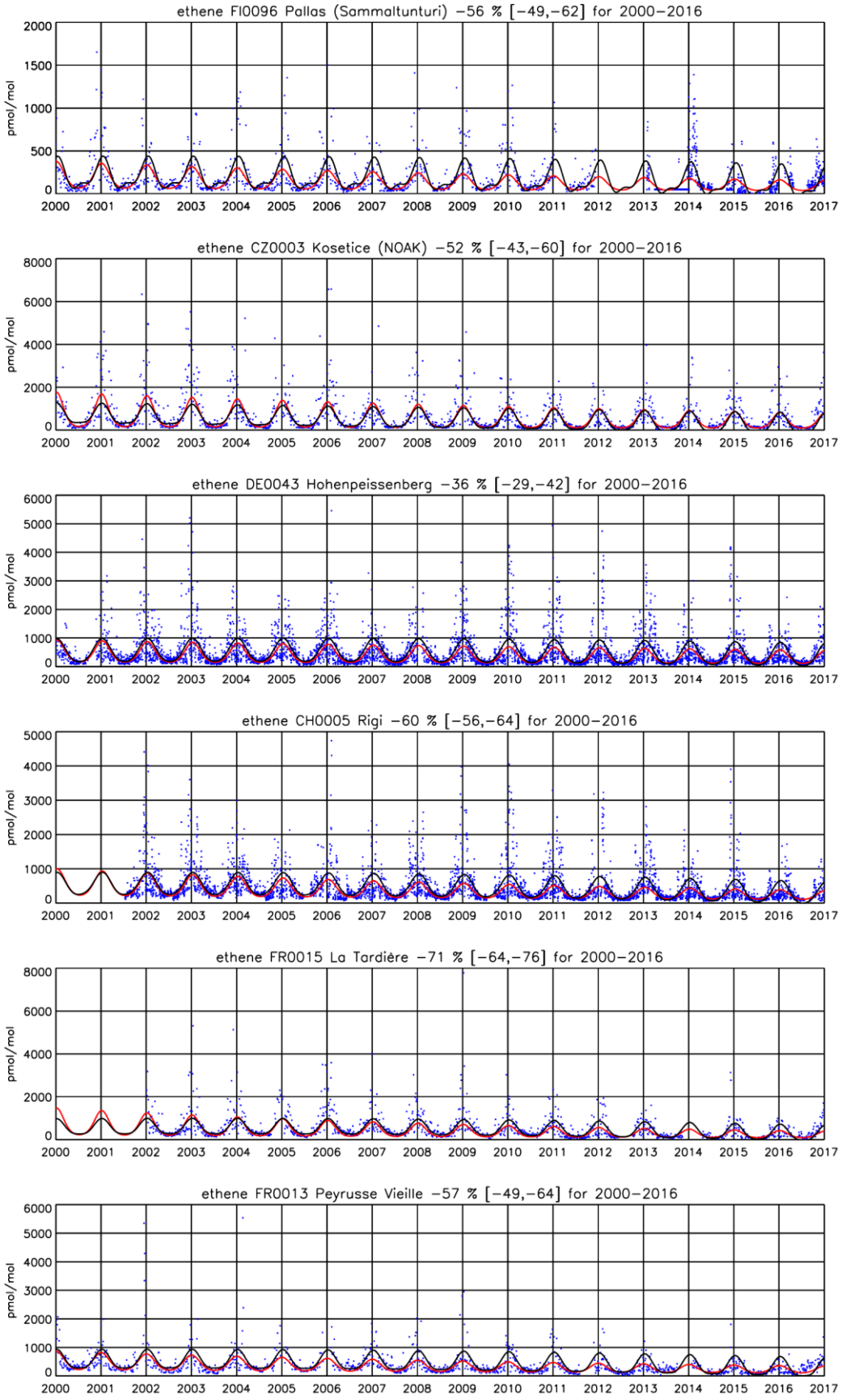

Figure 8. Same as Figure 7 for ethene.

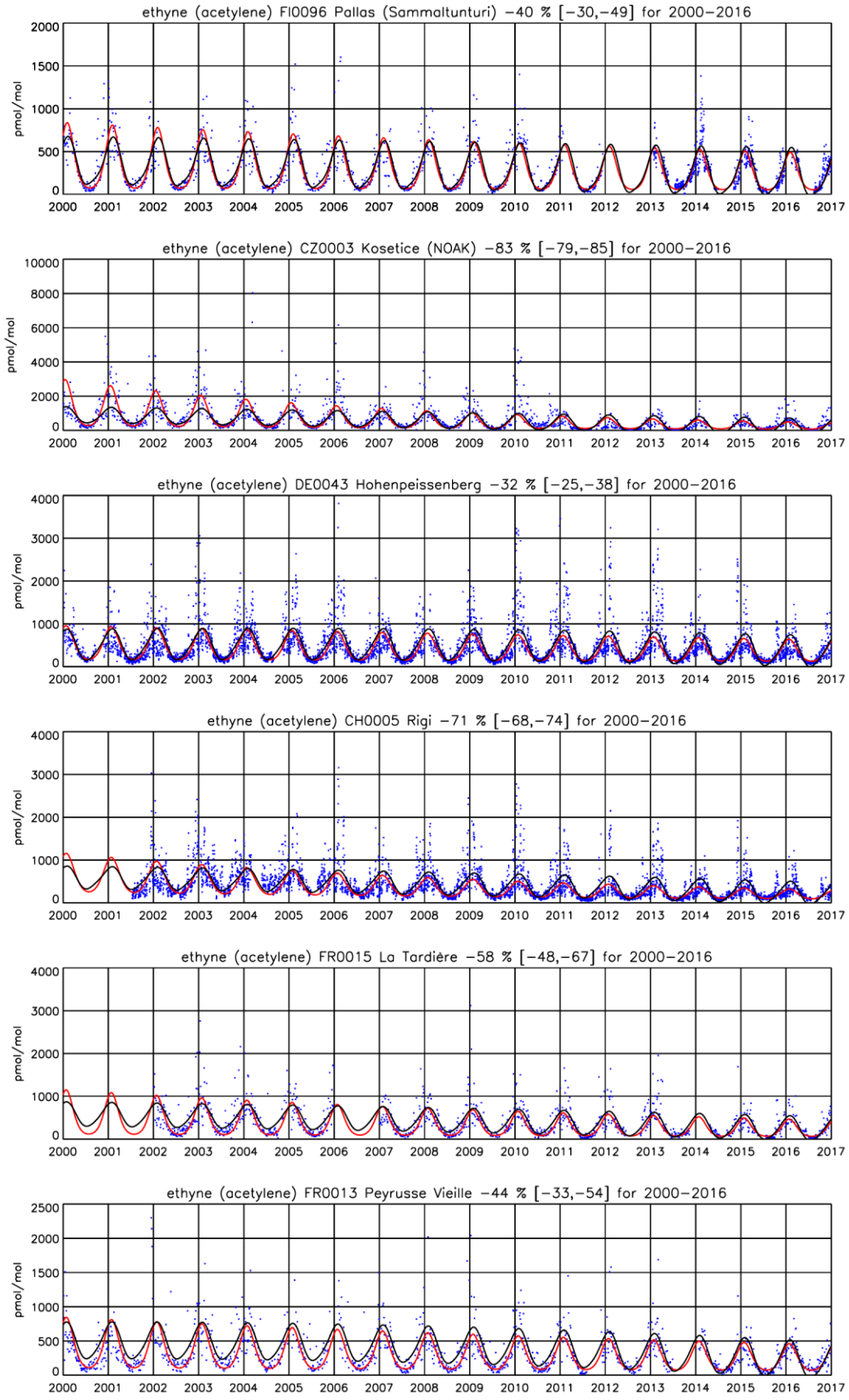

Figure 9. Same as Figure 7 for ethyne.

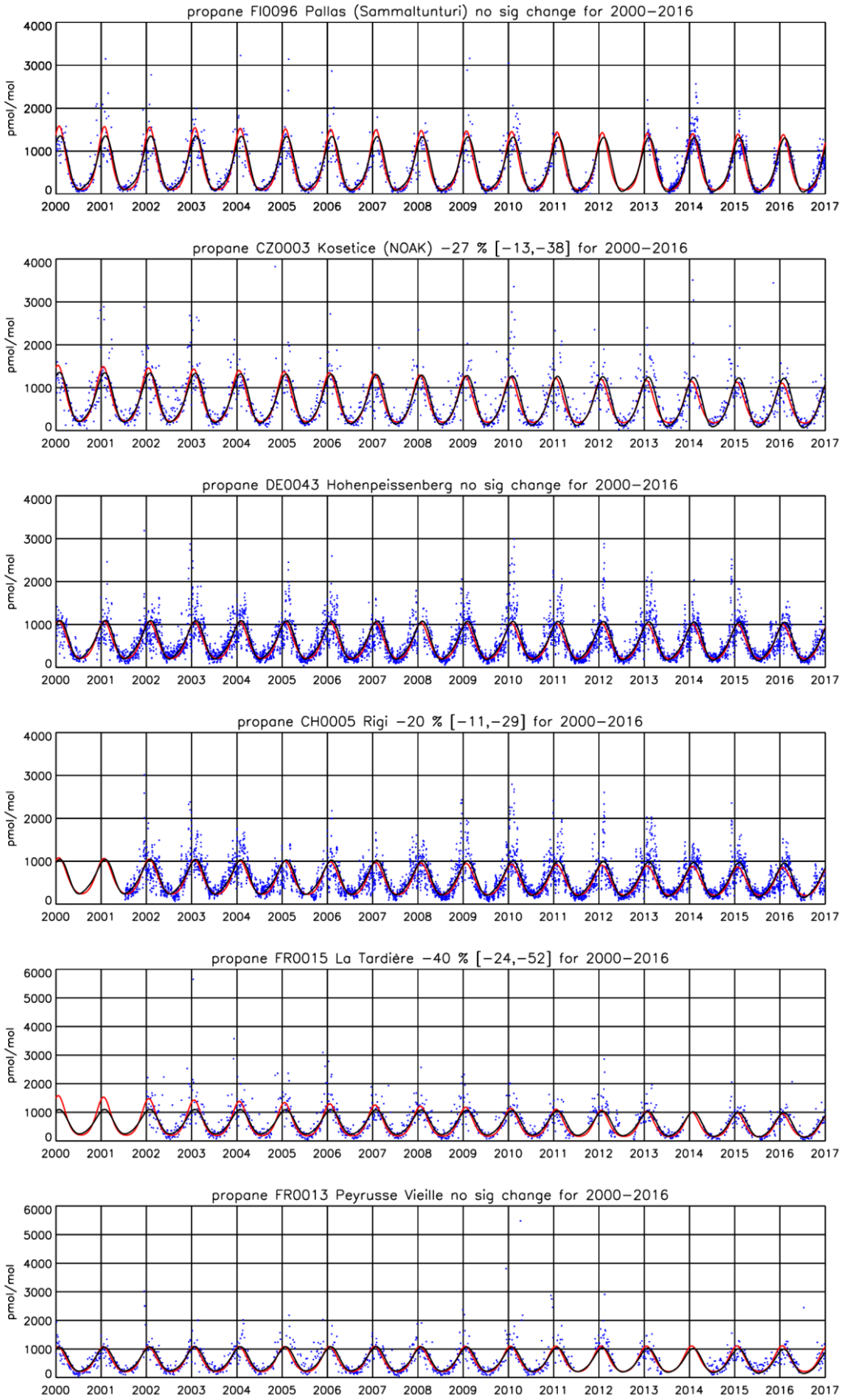

Figure 10. Same as Figure 7 for propane.

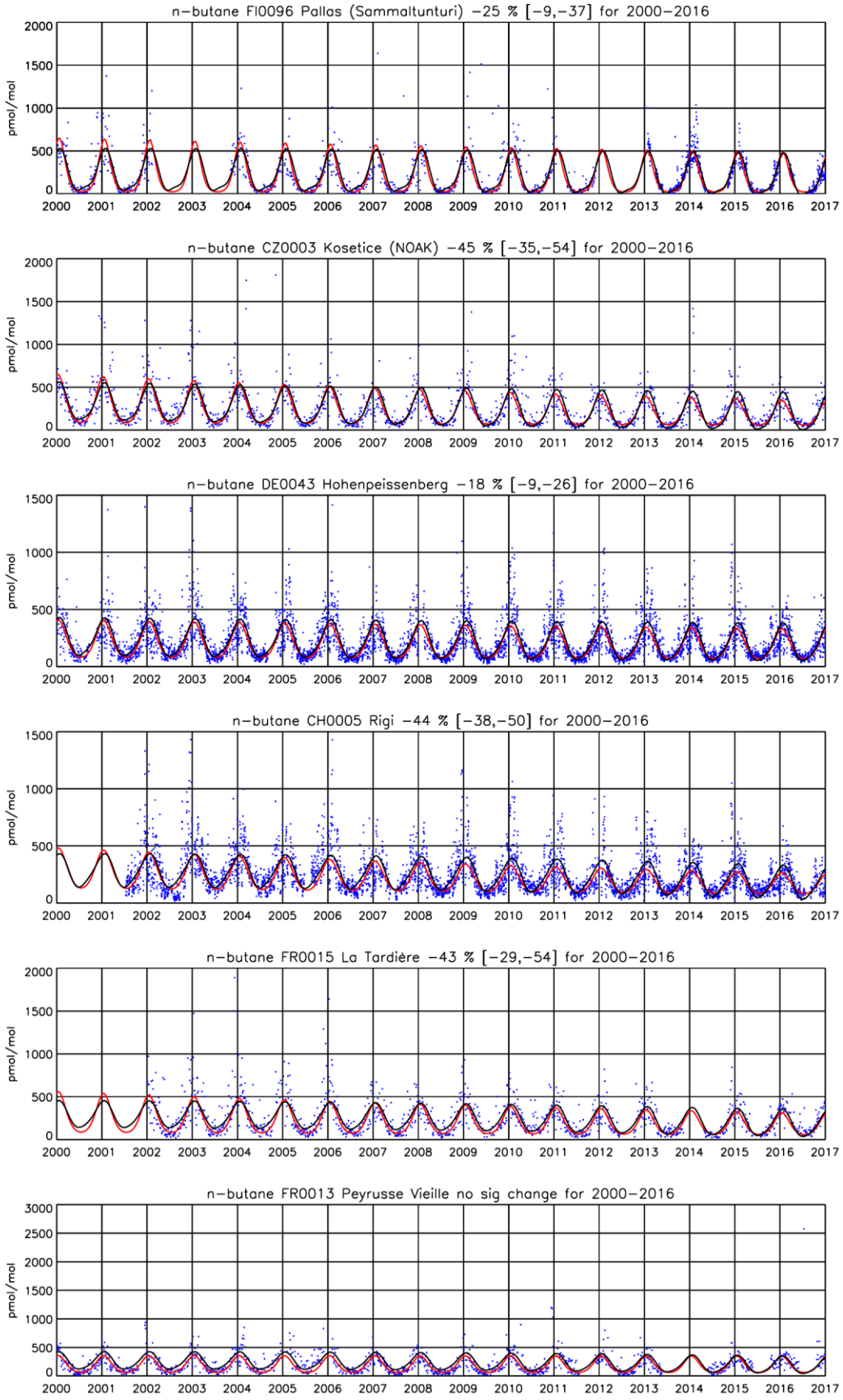

Figure 11. Same as Figure 7 for n-butane.

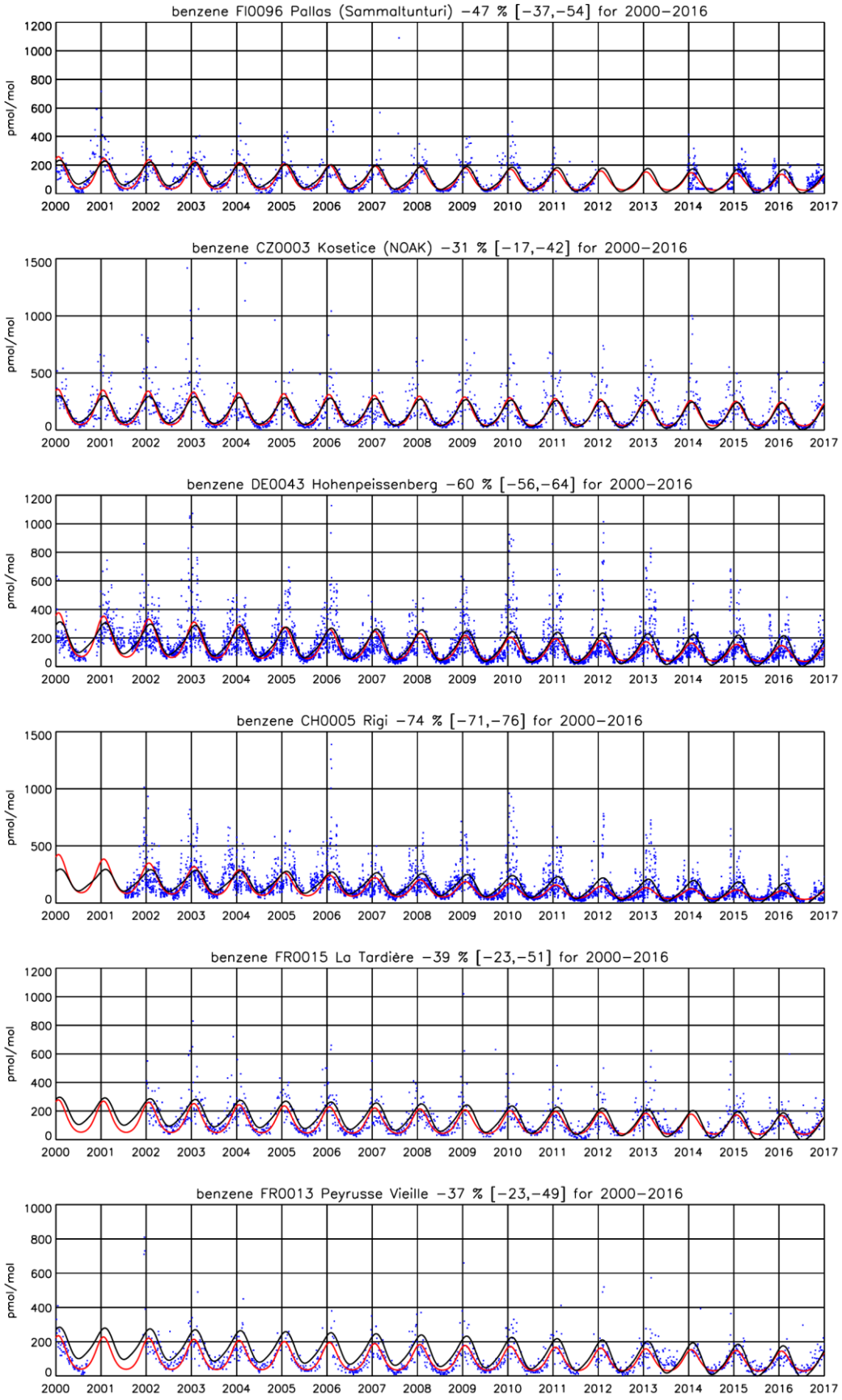

Figure 12. Same as Figure 7 for benzene.

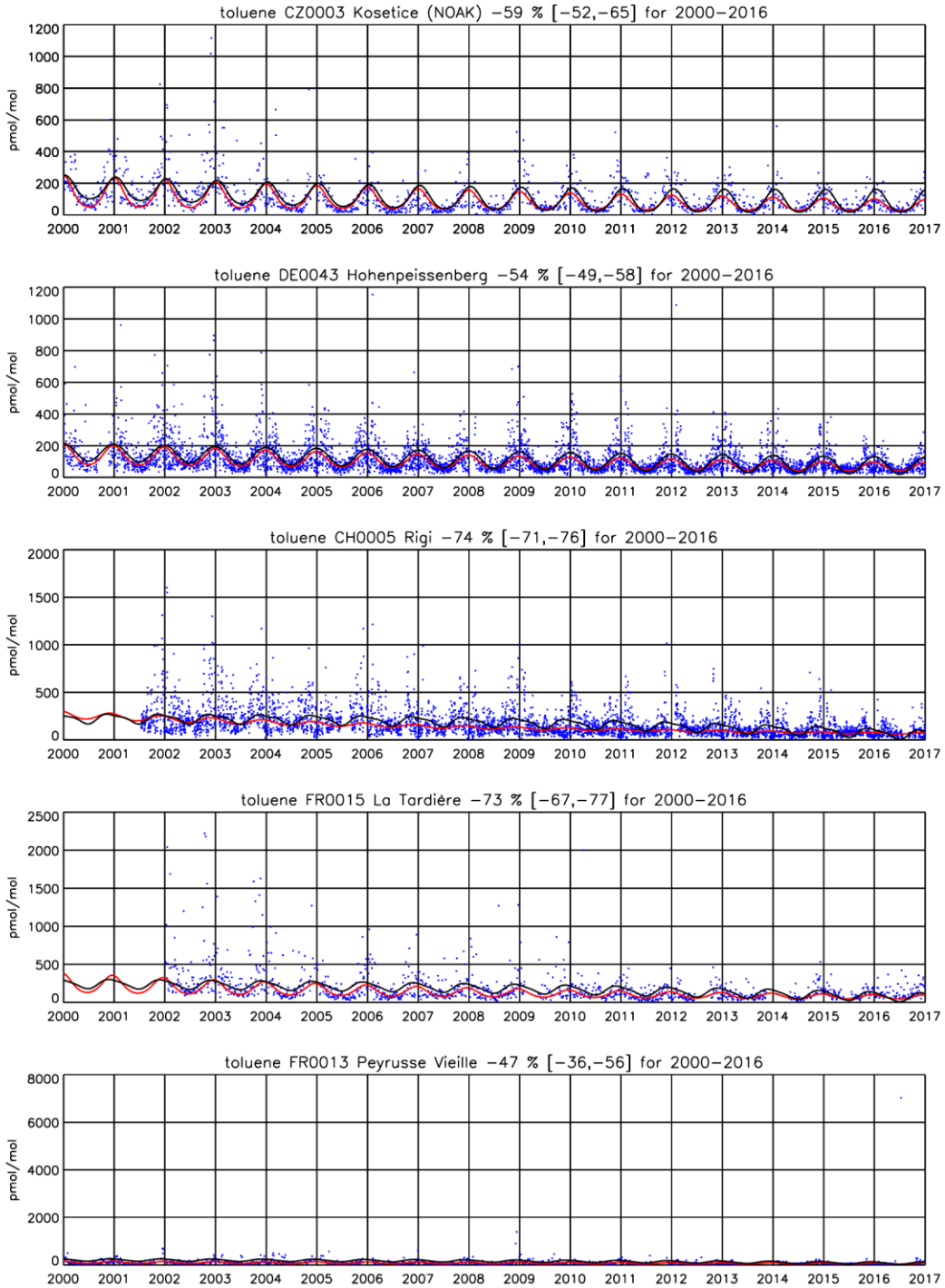

Figure 13. Same as Figure 7 for toluene.

Table 3. The percentage reduction in measured hydrocarbons over the period 2000-2016 relative to 2000 as the base year estimated from Eq 1 and Eq 2, respectively. 'ns' implies that no significant trend was found. The stations are: Pallas (PAL), Kosetice (KOS), Hohenpeissenberg (HPB), Rigi (RIG), La Tardiere (LTA) and Peyrusse Vieille (PEY).

	PAL		KOS		HPB		RIG		LTA		PEY	
	Eq 1	*Eq 2*	*Eq 1*	*Eq 2*	*Eq 1*	*Eq 2*	*Eq 1*	*Eq 2*	*Eq 1*	*Eq 2*	*Eq 1*	*Eq 2*
Ethane	*ns*	*ns*	*ns*	*ns*	*ns*	*ns*	18	10	*ns*	10	*ns*	10
Ethene	56	46	52	66	36	28	60	48	71	52	57	52
Ethyne	40	37	83	78	32	30	71	63	58	59	44	58
Propane	*ns*	*ns*	27	19	*ns*	*ns*	20	15	40	19	*ns*	13
n-butane	25	24	45	39	18	20	44	38	43	38	*ns*	30
Benzene	47	45	31	38	60	49	74	66	39	58	37	60
Toluene	-	-	59	50	54	53	74	72	73	71	47	65

5. Acknowledgement

Data originators for individual datasets can be found as part of the metadata by visiting http://ebas.nilu.no. Special thanks to the extensive effort and contribution provided by all participants through the ACTRIS-2 project including long-lasting detailed discussions on individual data values.

6. References

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URL: http://www.nilu.no/projects/ccc/manual/index.html [2015-09-10].

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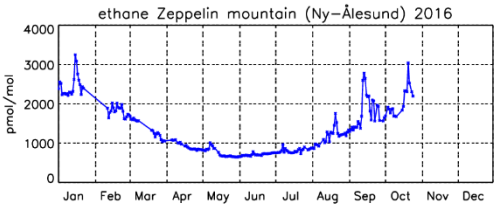
Appendix

Time series of daily means of VOC measured in 2016 listed from north to south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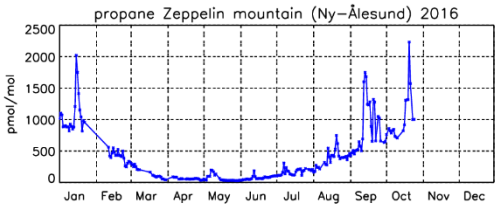
pmol/mol
4000
3000
2000
1000
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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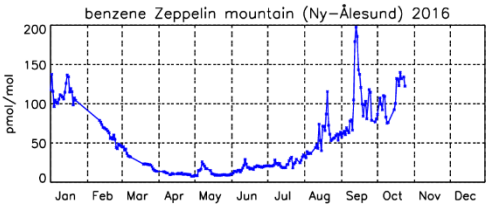
pmol/mol
2500
2000
1500
1000
500
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


benzene Zeppelin mountain (Ny−Ålesund) 2016
pmol/mol
200
150
100
50
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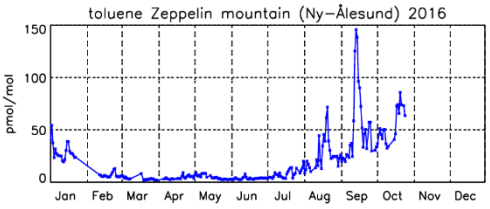
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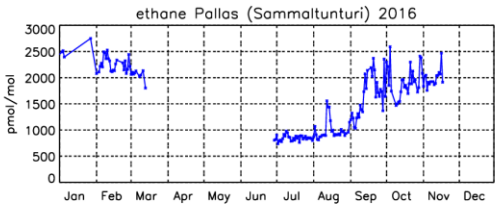
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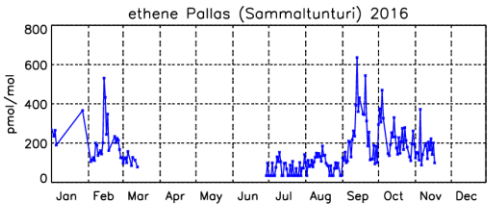
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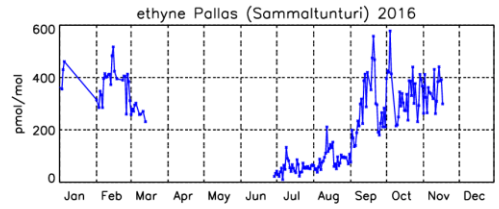
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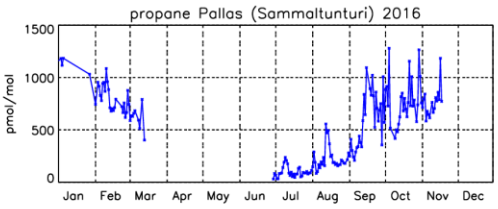
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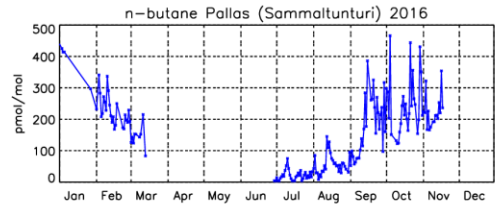
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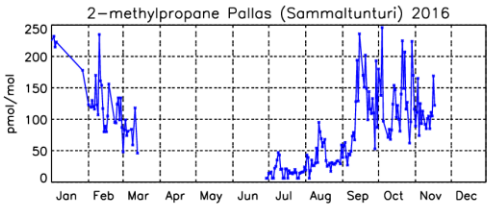
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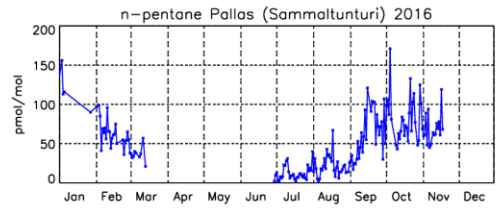
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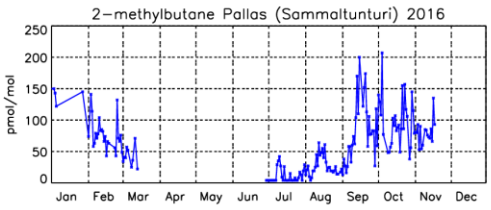
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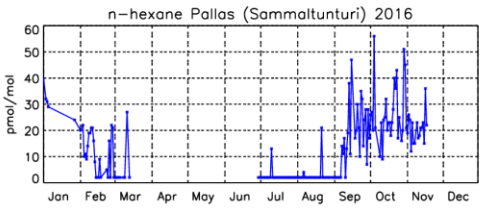
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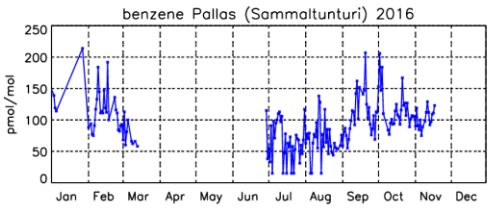
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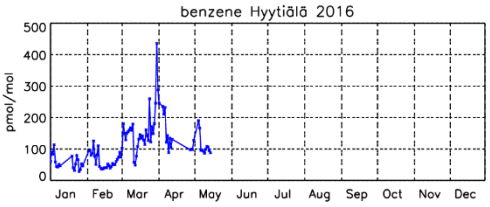
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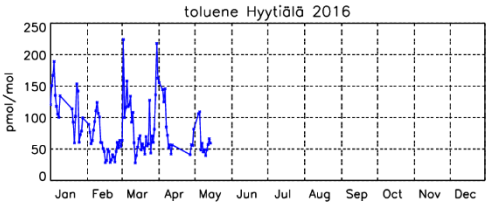
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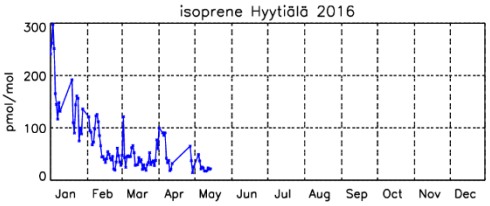
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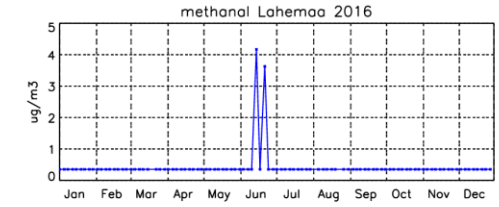
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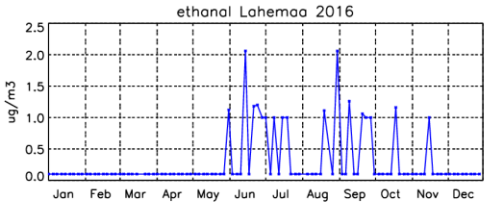
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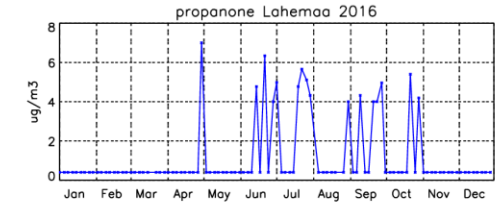
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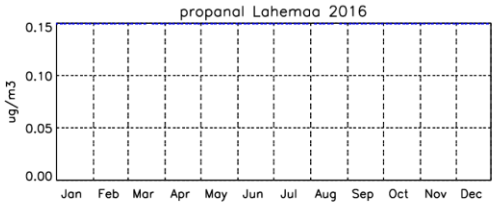
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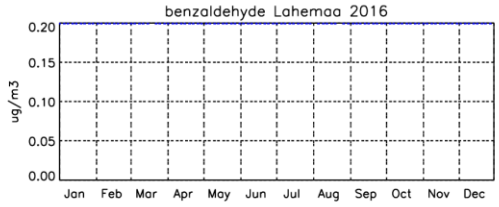
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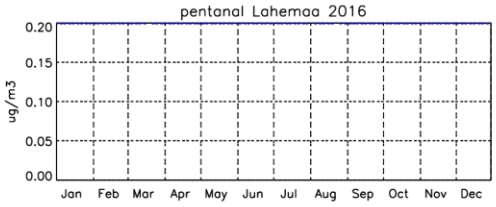
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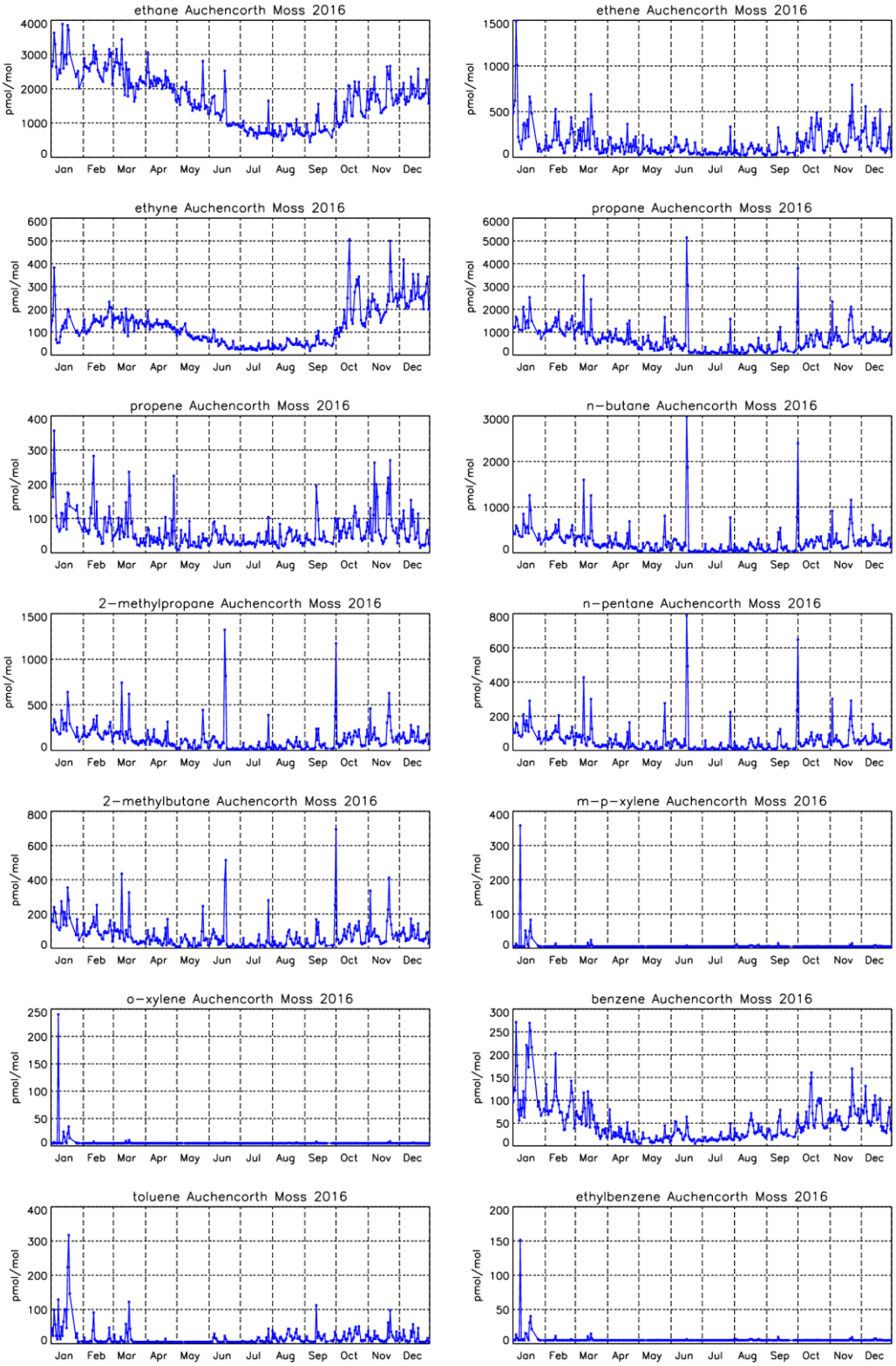
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2−methylpropane Auchencorth Moss 2016
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m−p−xylene Auchencorth Moss 2016
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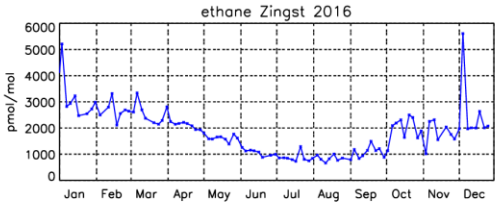
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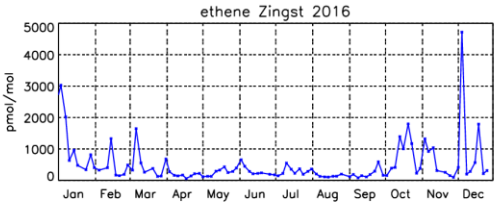
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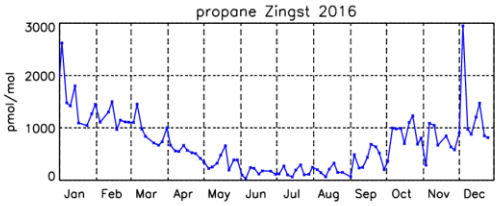
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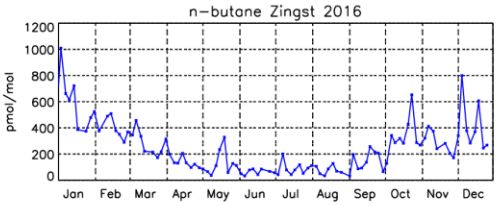
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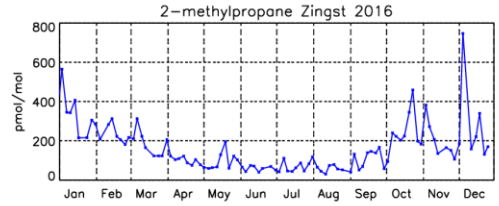
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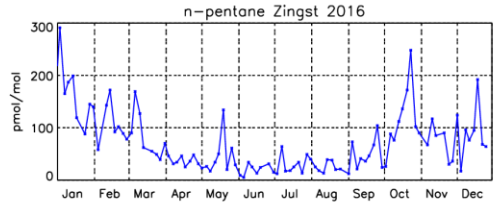
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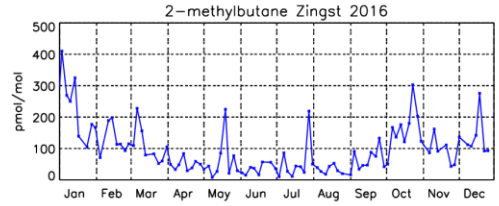
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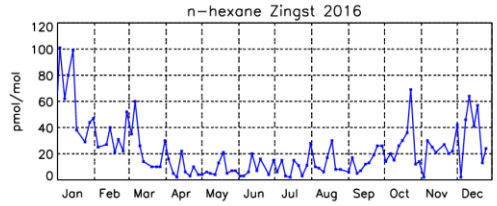
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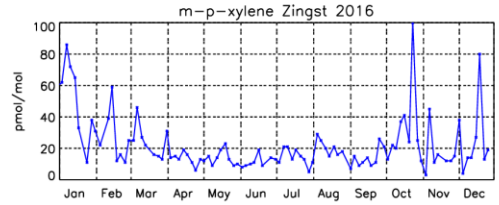
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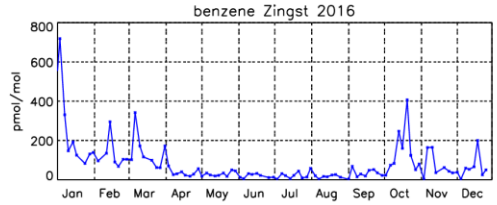
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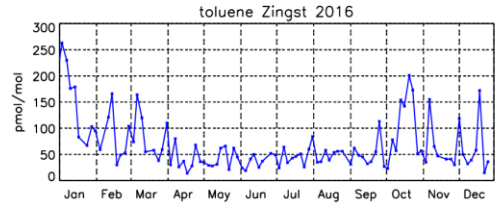
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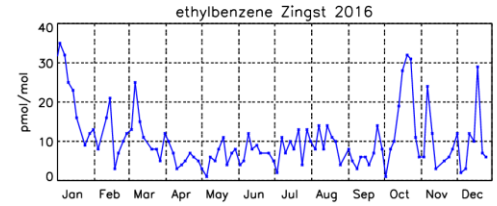
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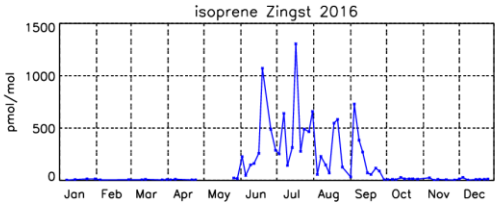
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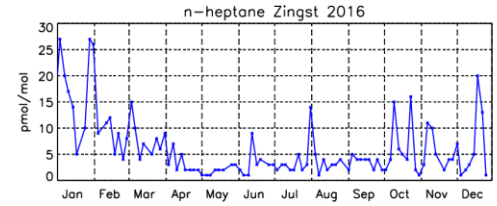
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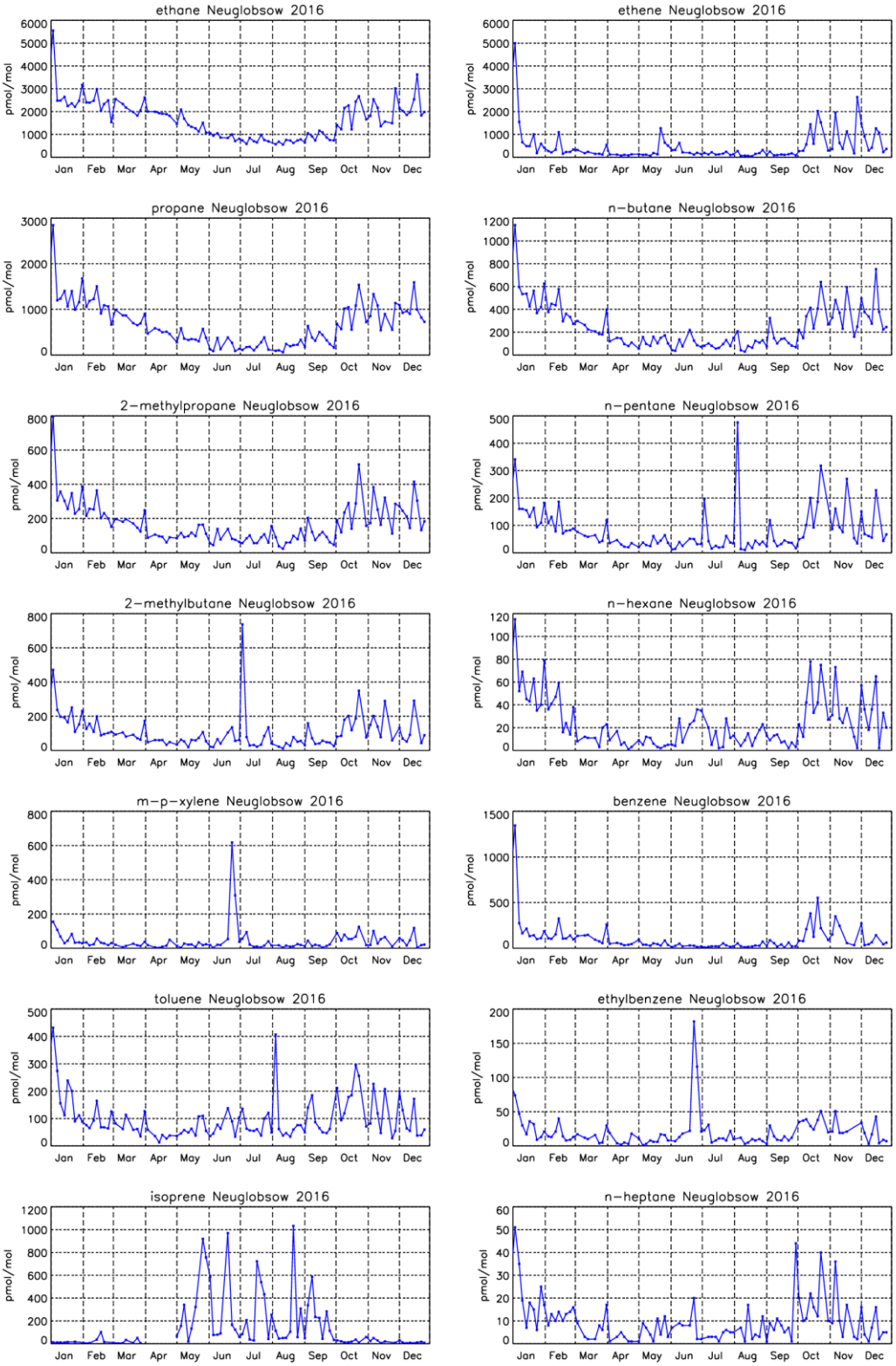
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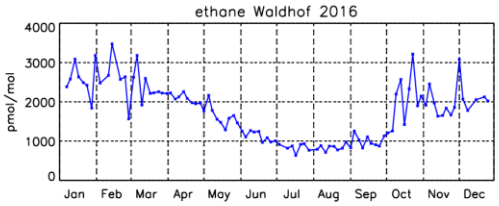
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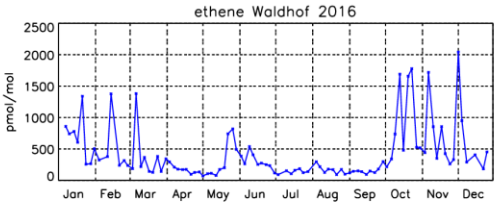
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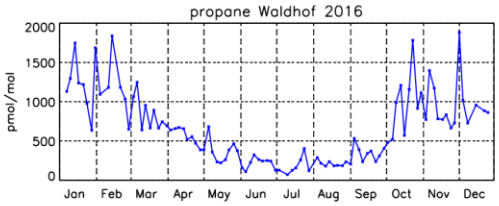
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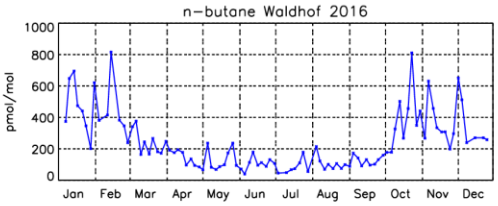
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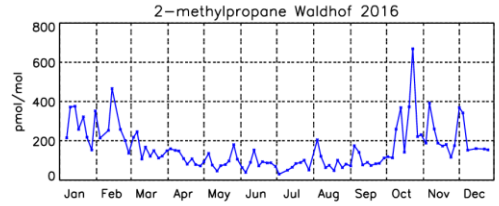
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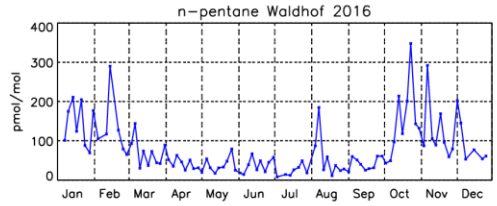
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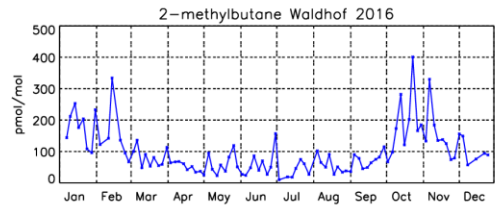
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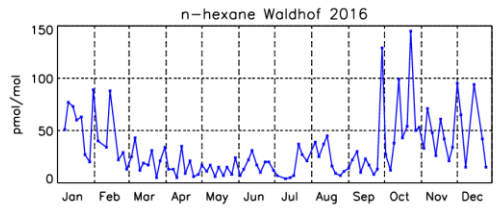
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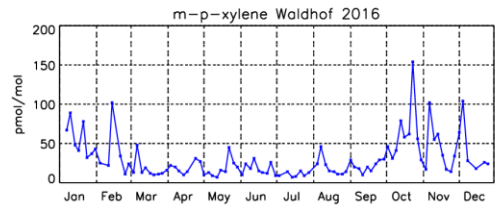
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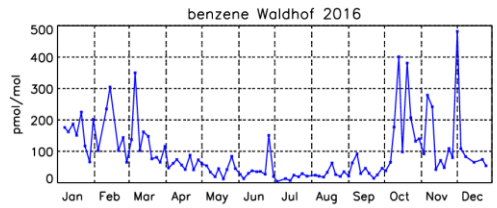
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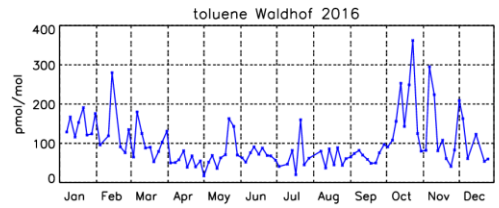
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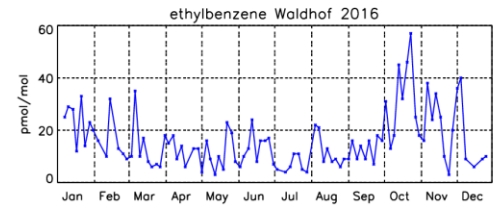
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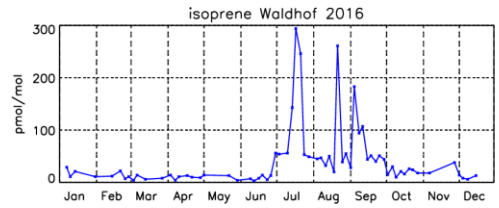
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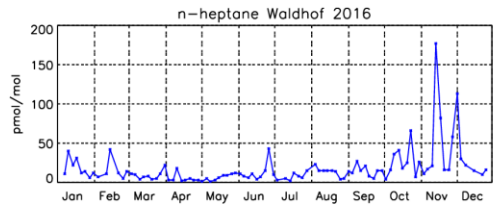
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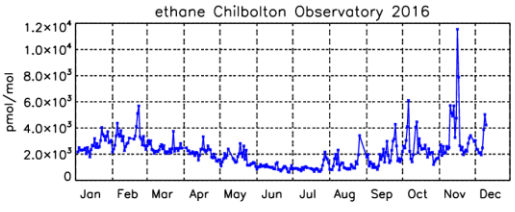
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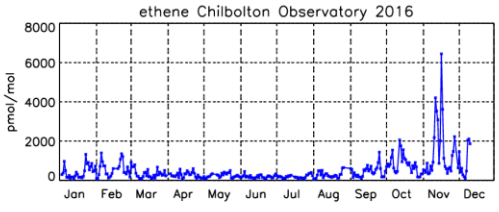
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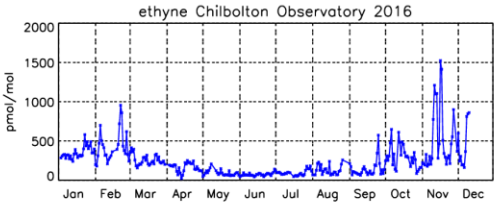
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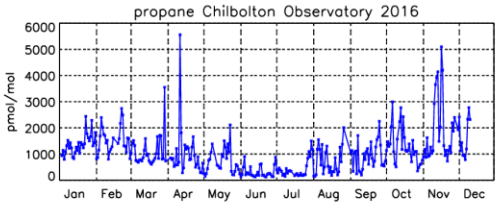
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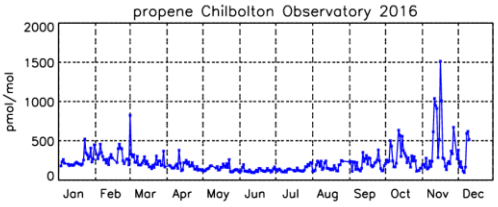
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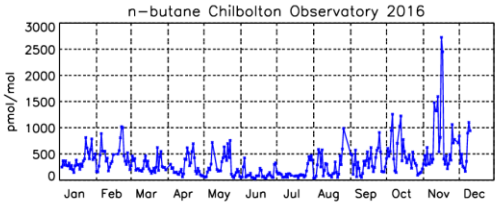
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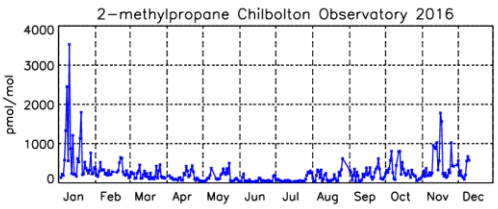
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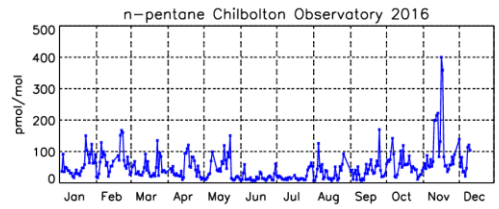
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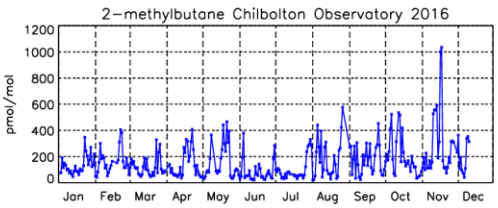
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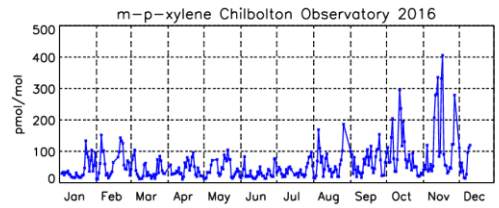
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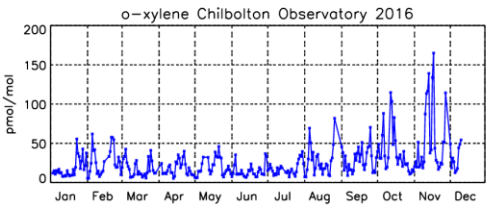
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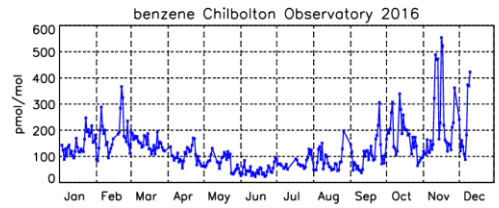
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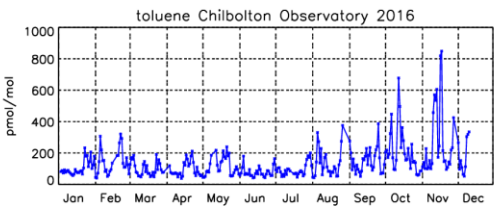
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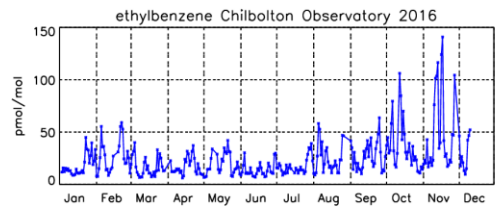
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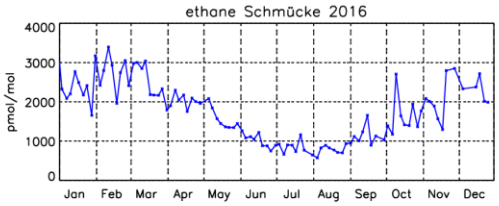
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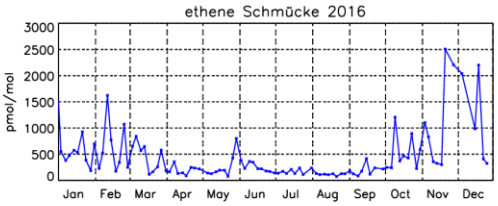
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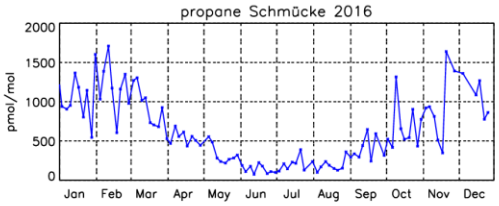
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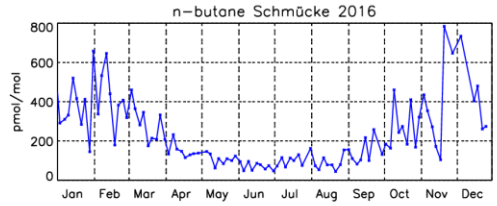
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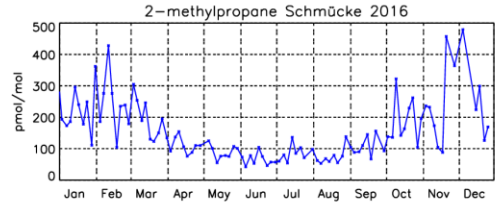
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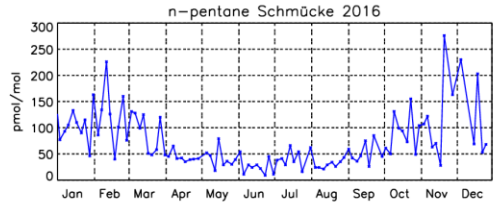
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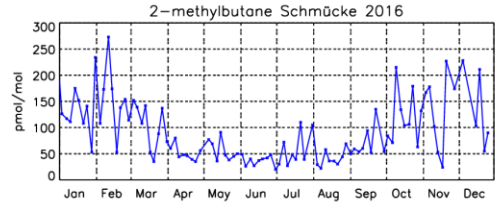
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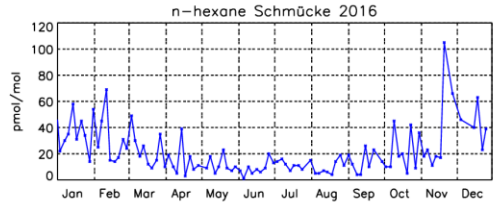
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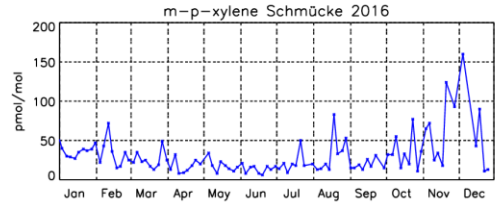
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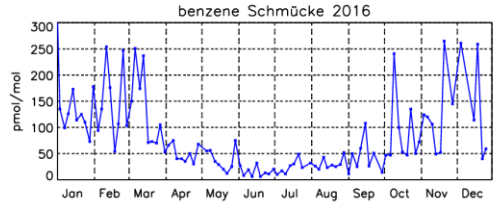
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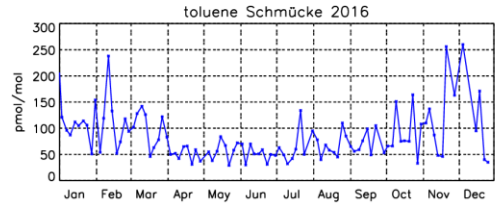
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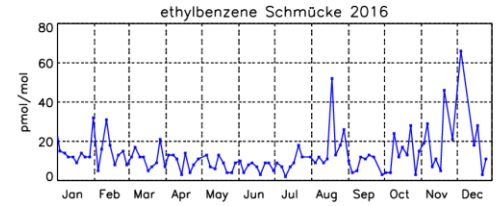
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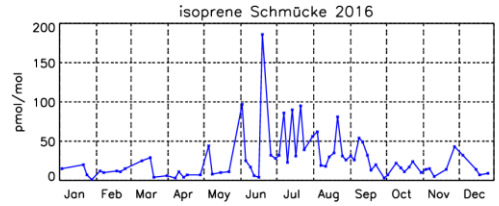
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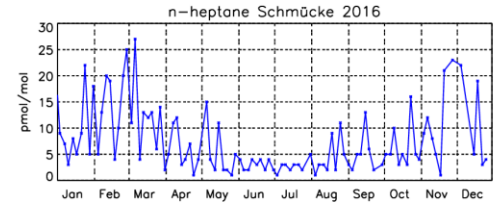
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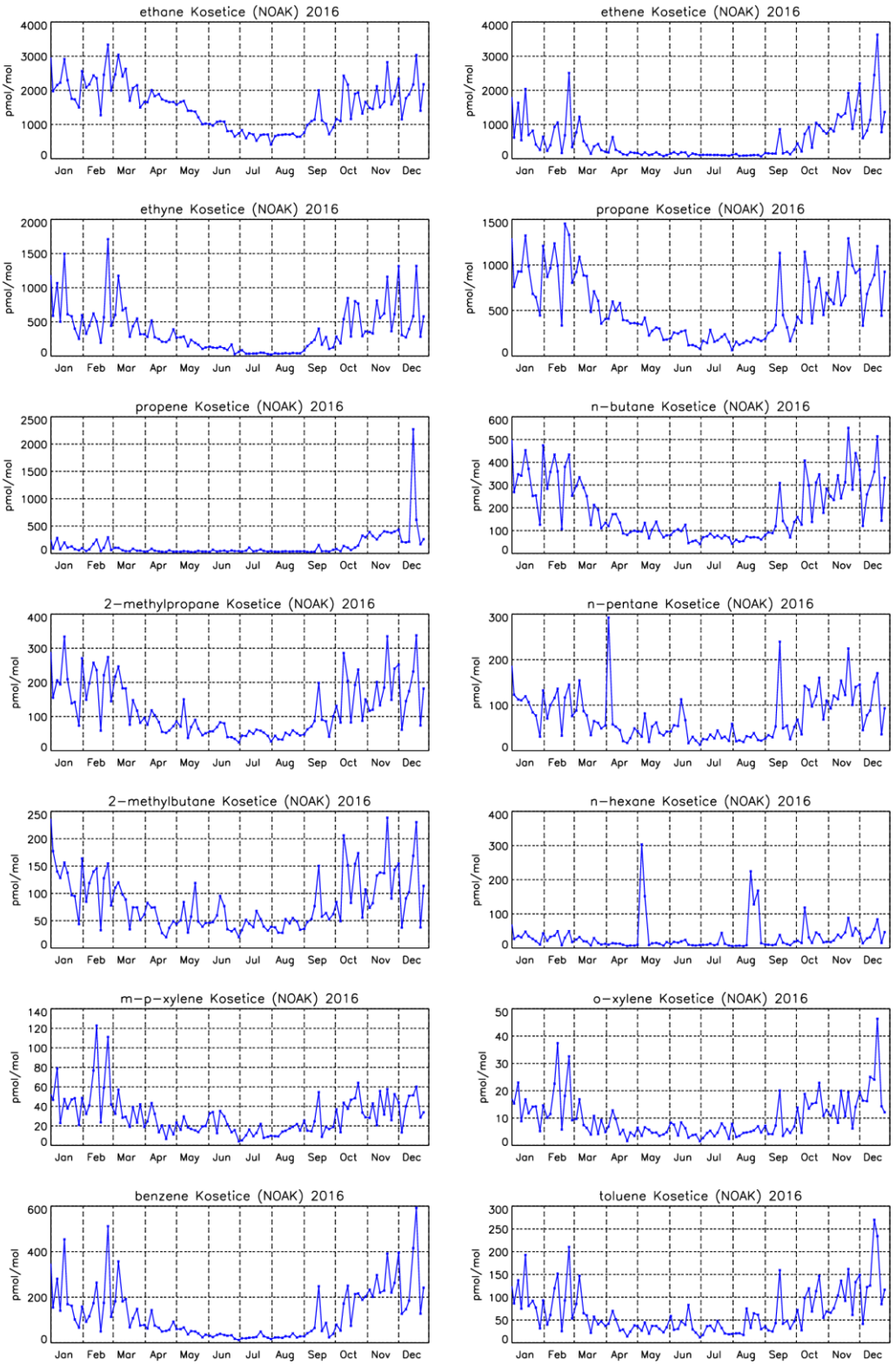
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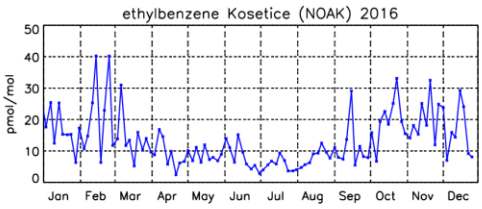
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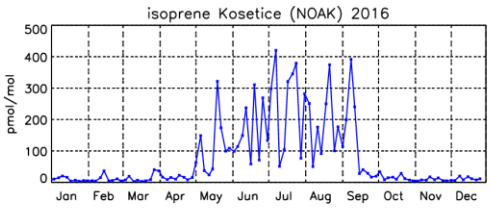
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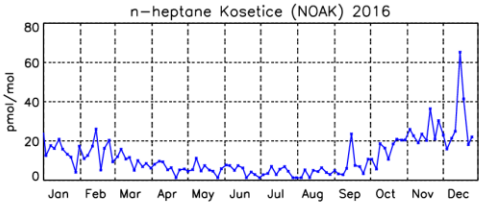
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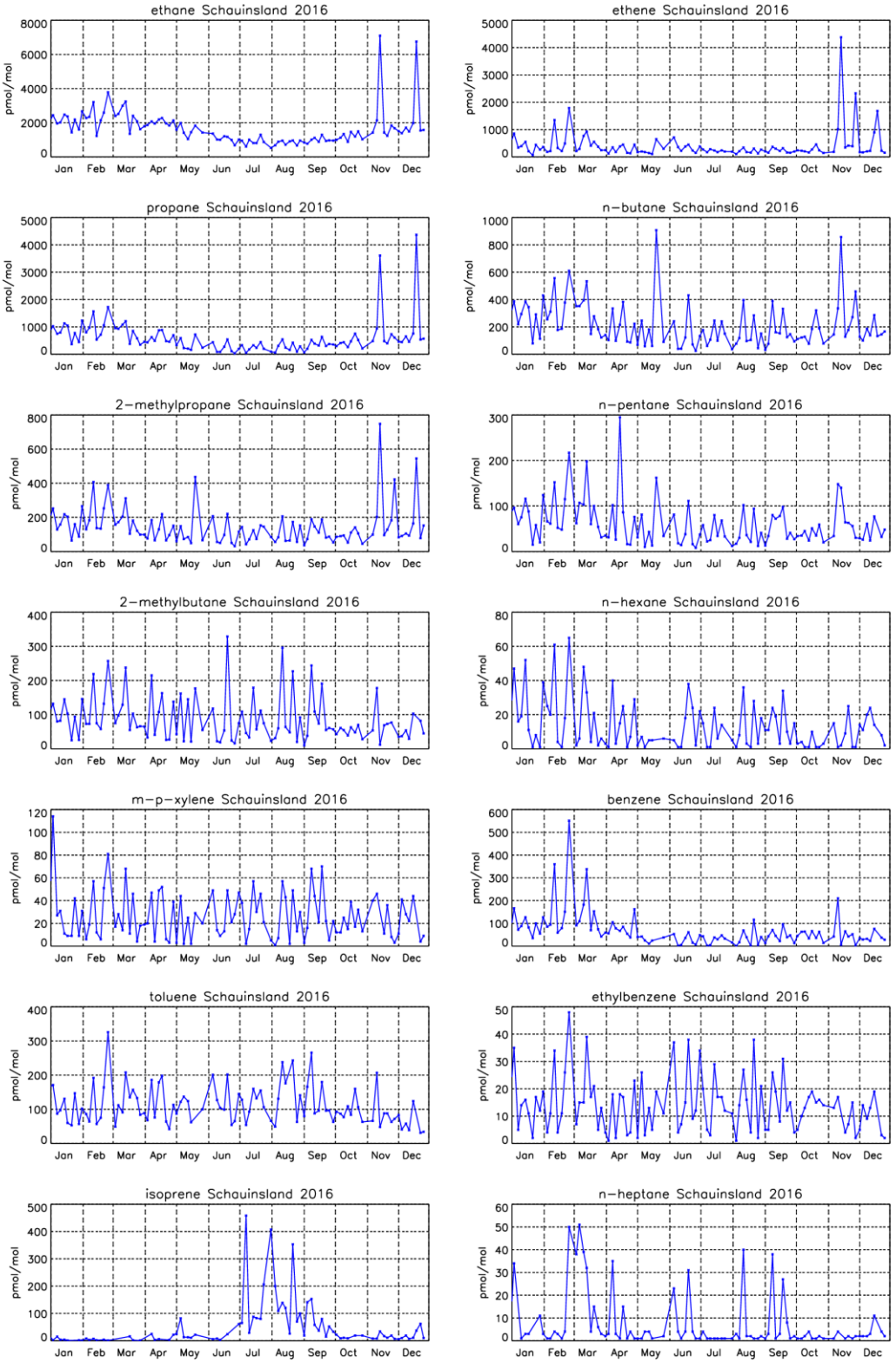
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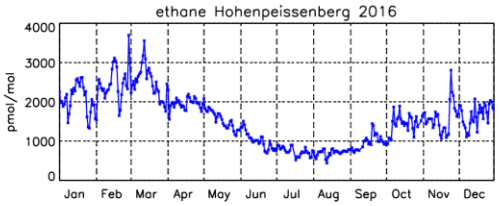
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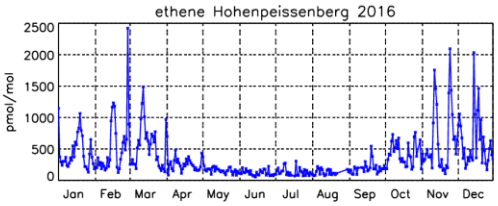
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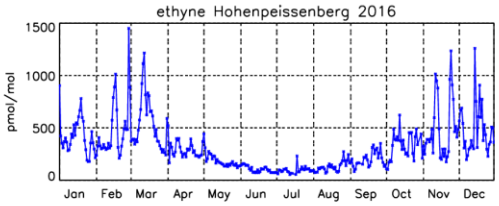
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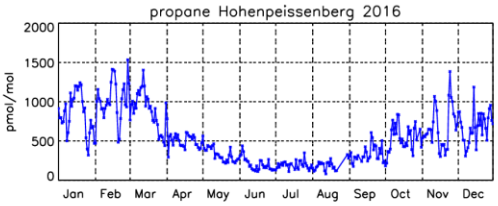
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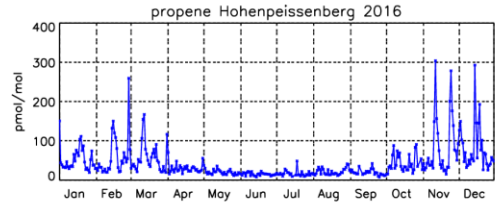
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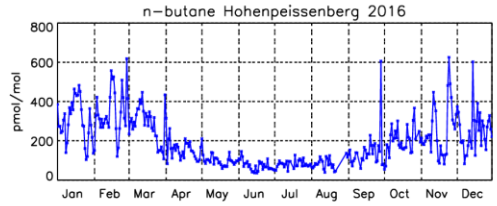
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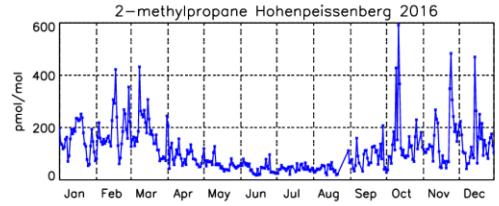
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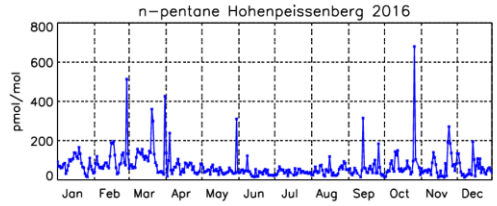
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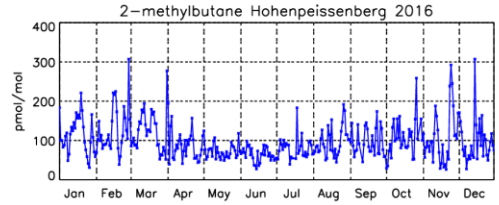
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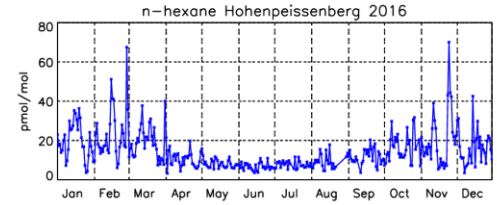
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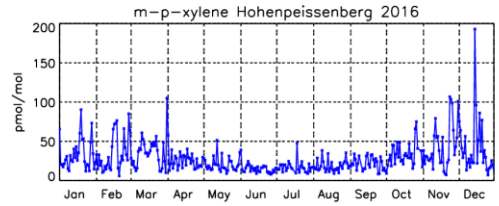
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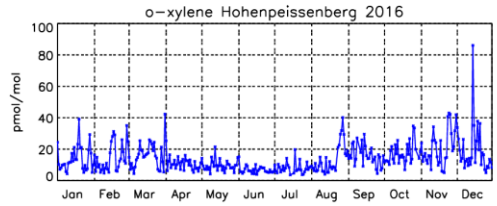
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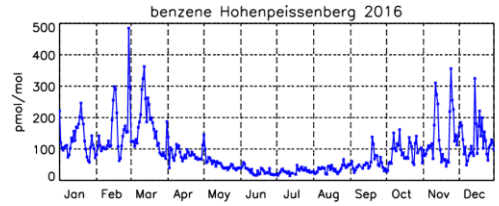
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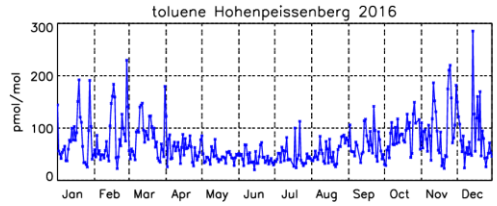
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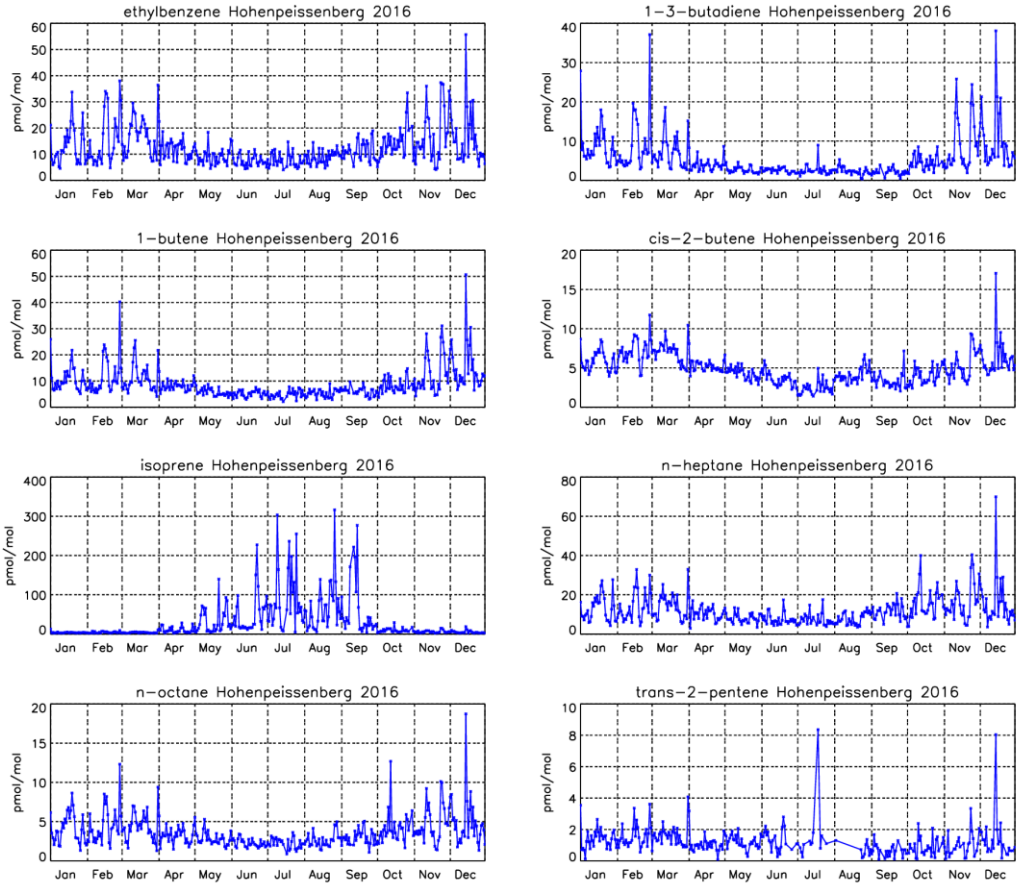
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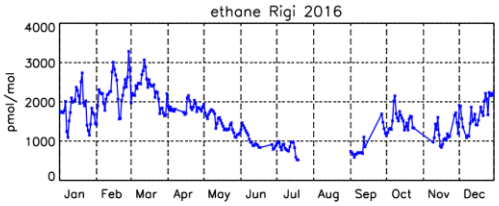
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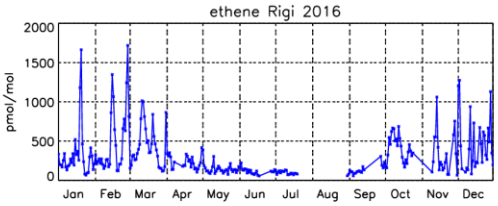
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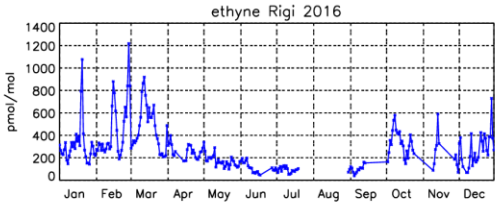
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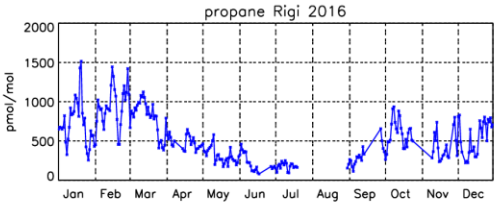
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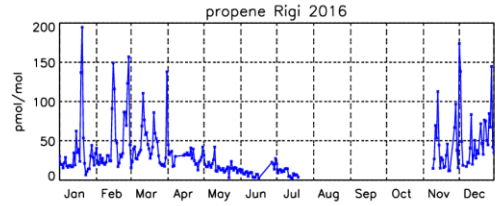
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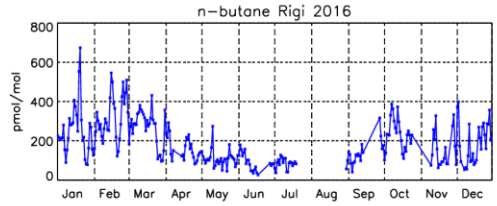
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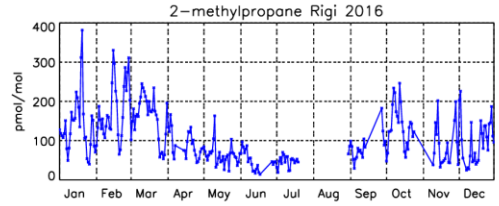
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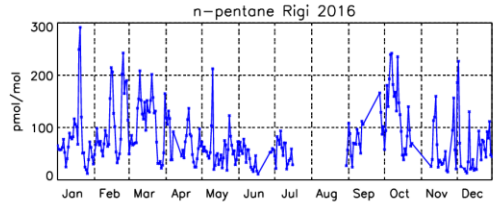
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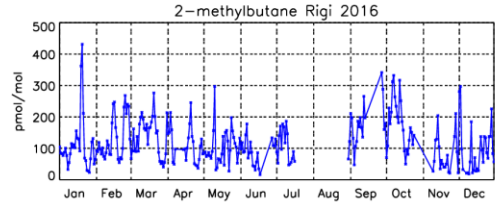
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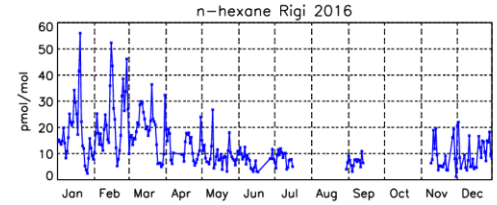
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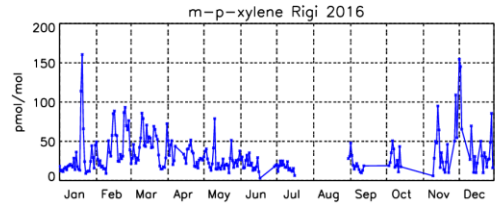
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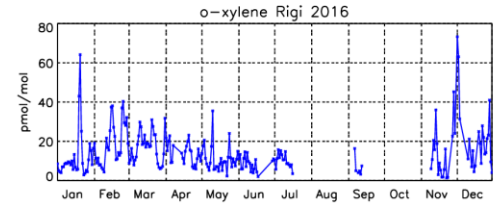
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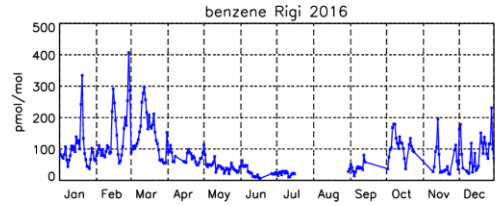
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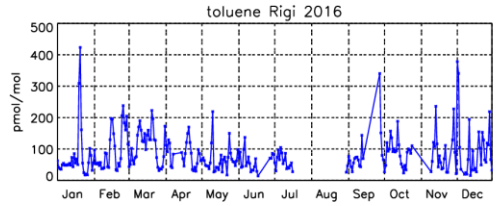
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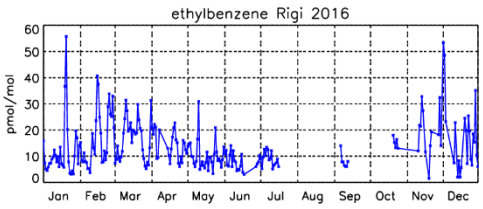
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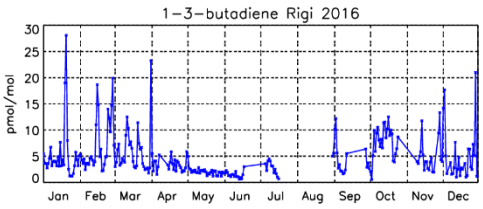
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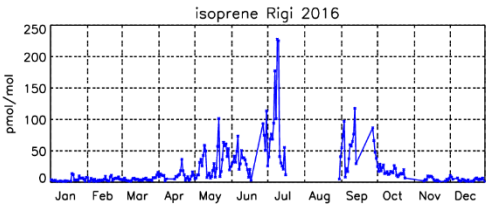
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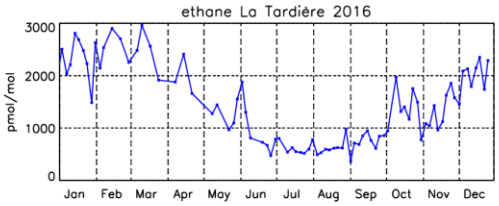
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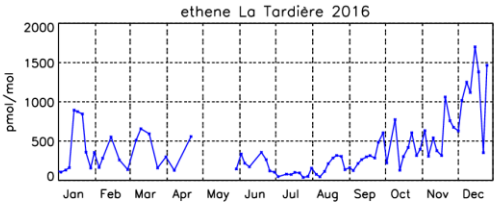
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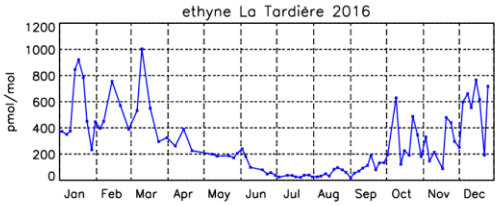
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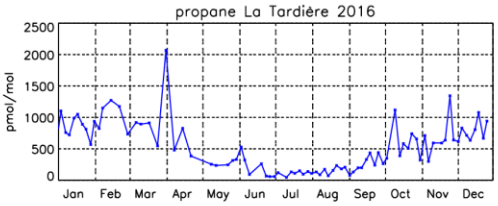
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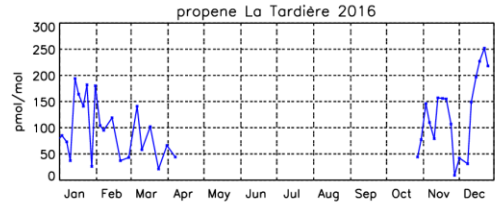
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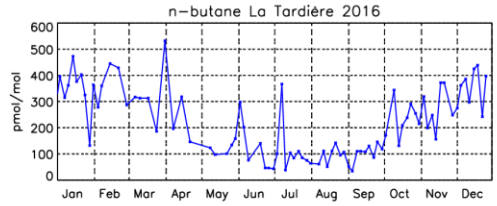
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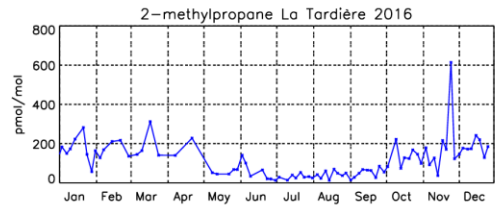
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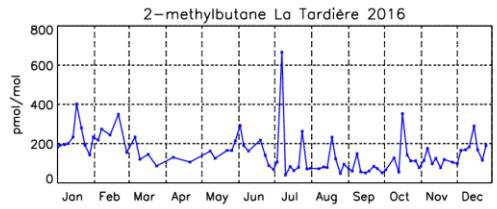
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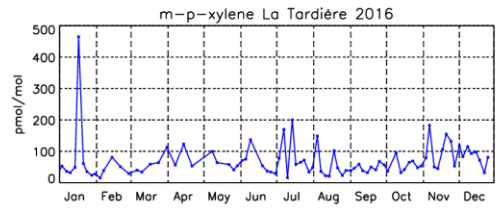
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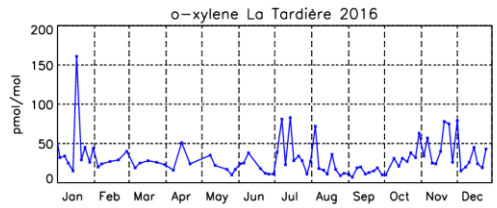
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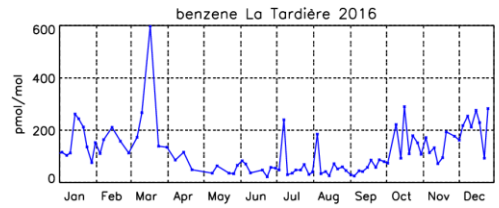
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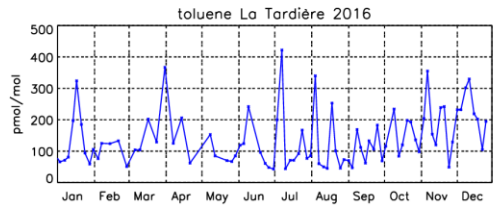
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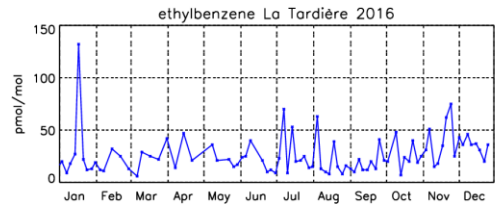
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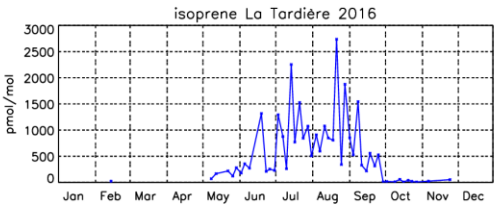
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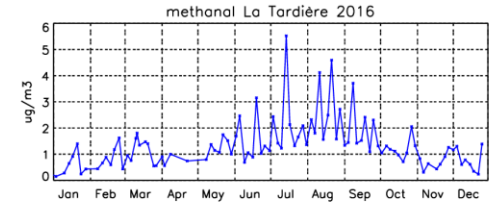
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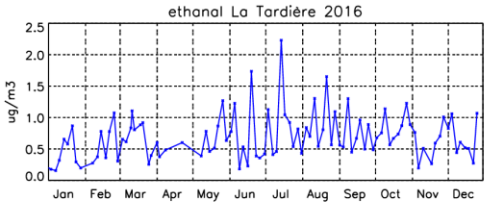
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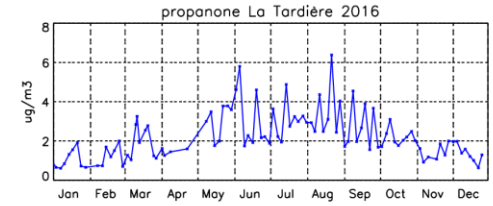
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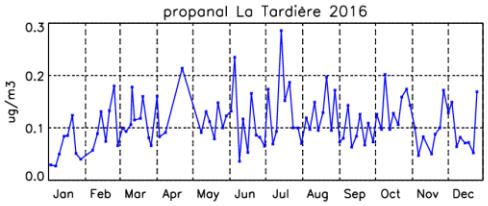
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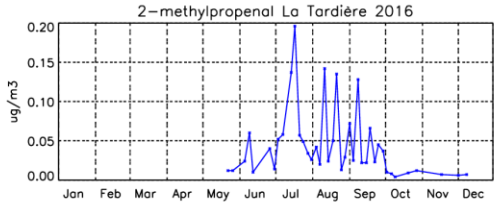
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ug/m3
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0.10
0.15
0.20
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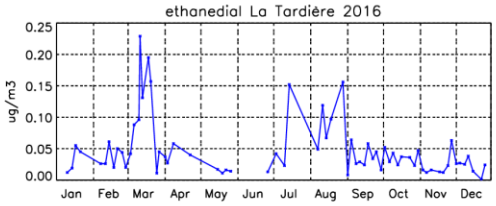
ethanedial La Tardière 2016
ug/m3
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0.10
0.15
0.20
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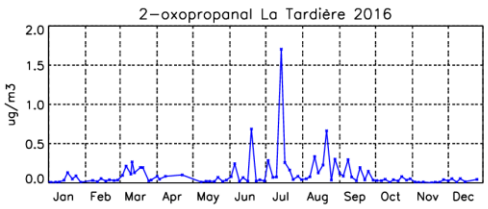
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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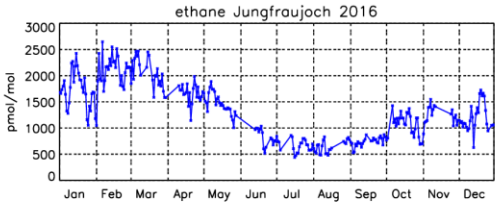
ethane Jungfraujoch 2016
pmol/mol
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2500
2000
1500
1000
500
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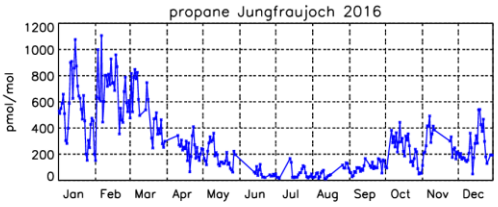
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propane Jungfraujoch 2016
pmol/mol
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800
600
400
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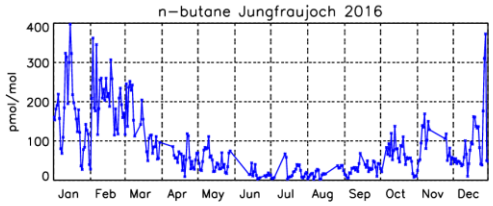
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n−butane Jungfraujoch 2016
pmol/mol
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300
200
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0
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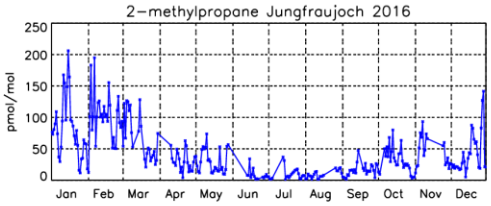
2−methylpropane Jungfraujoch 2016
pmol/mol
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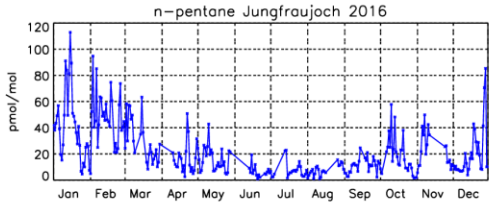
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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pmol/mol
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40
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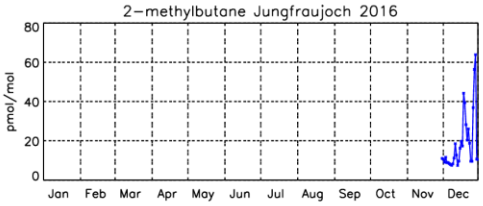
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2−methylbutane Jungfraujoch 2016
pmol/mol
80
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40
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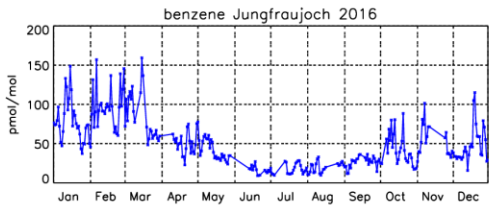
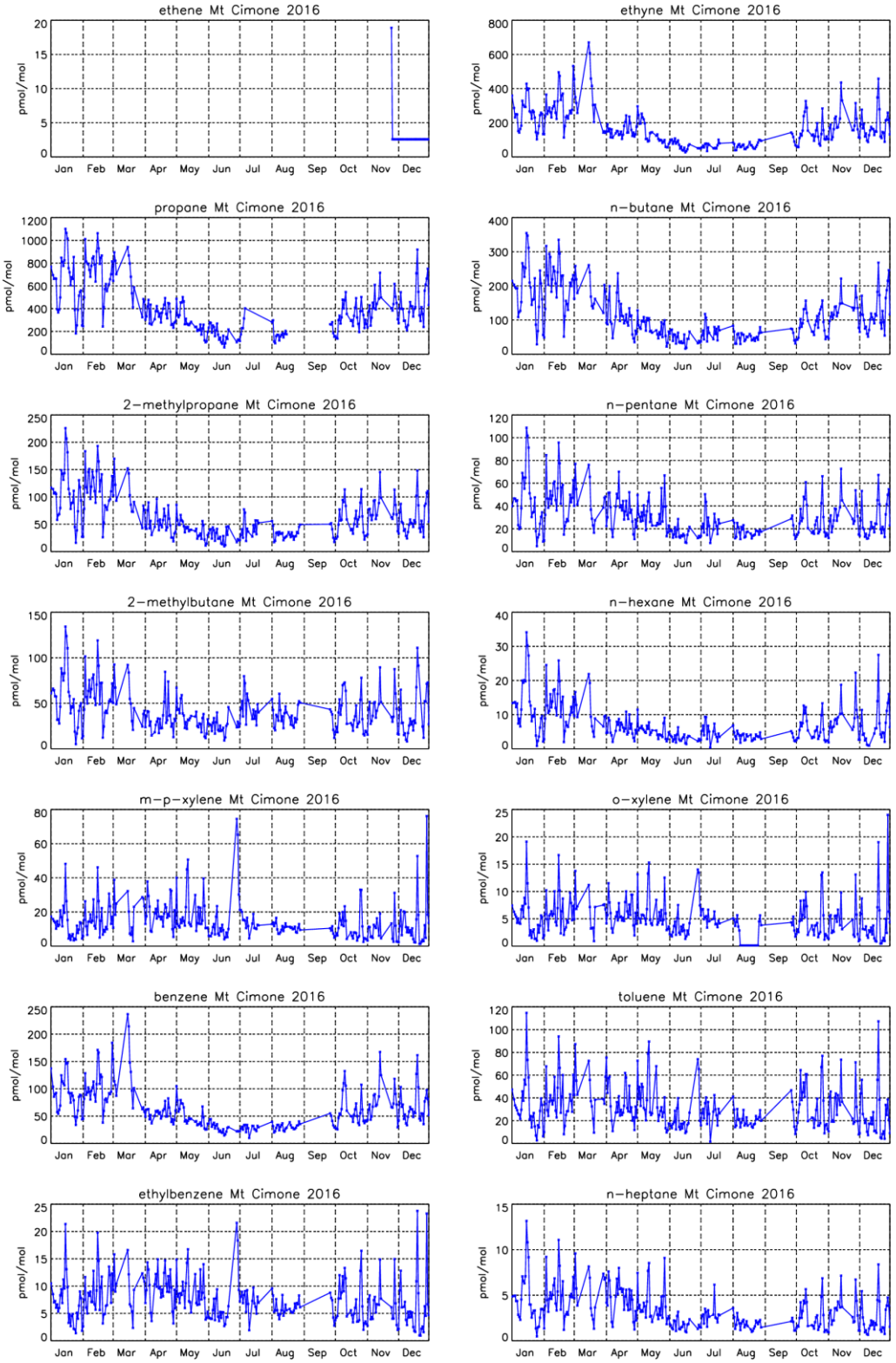
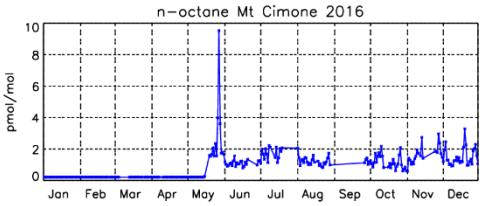
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


benzene Jungfraujoch 2016
pmol/mol
200
150
100
50
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethene Mt Cimone 2016
ethyne Mt Cimone 2016
propane Mt Cimone 2016
n−butane Mt Cimone 2016
2−methylpropane Mt Cimone 2016
n−pentane Mt Cimone 2016
2−methylbutane Mt Cimone 2016
n−hexane Mt Cimone 2016
m−p−xylene Mt Cimone 2016
o−xylene Mt Cimone 2016
benzene Mt Cimone 2016
toluene Mt Cimone 2016
ethylbenzene Mt Cimone 2016
n−heptane Mt Cimone 2016
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-octane Mt Cimone 2016
pmol/mol
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6
4
2
0
Jan
Feb
Mar
Apr
May
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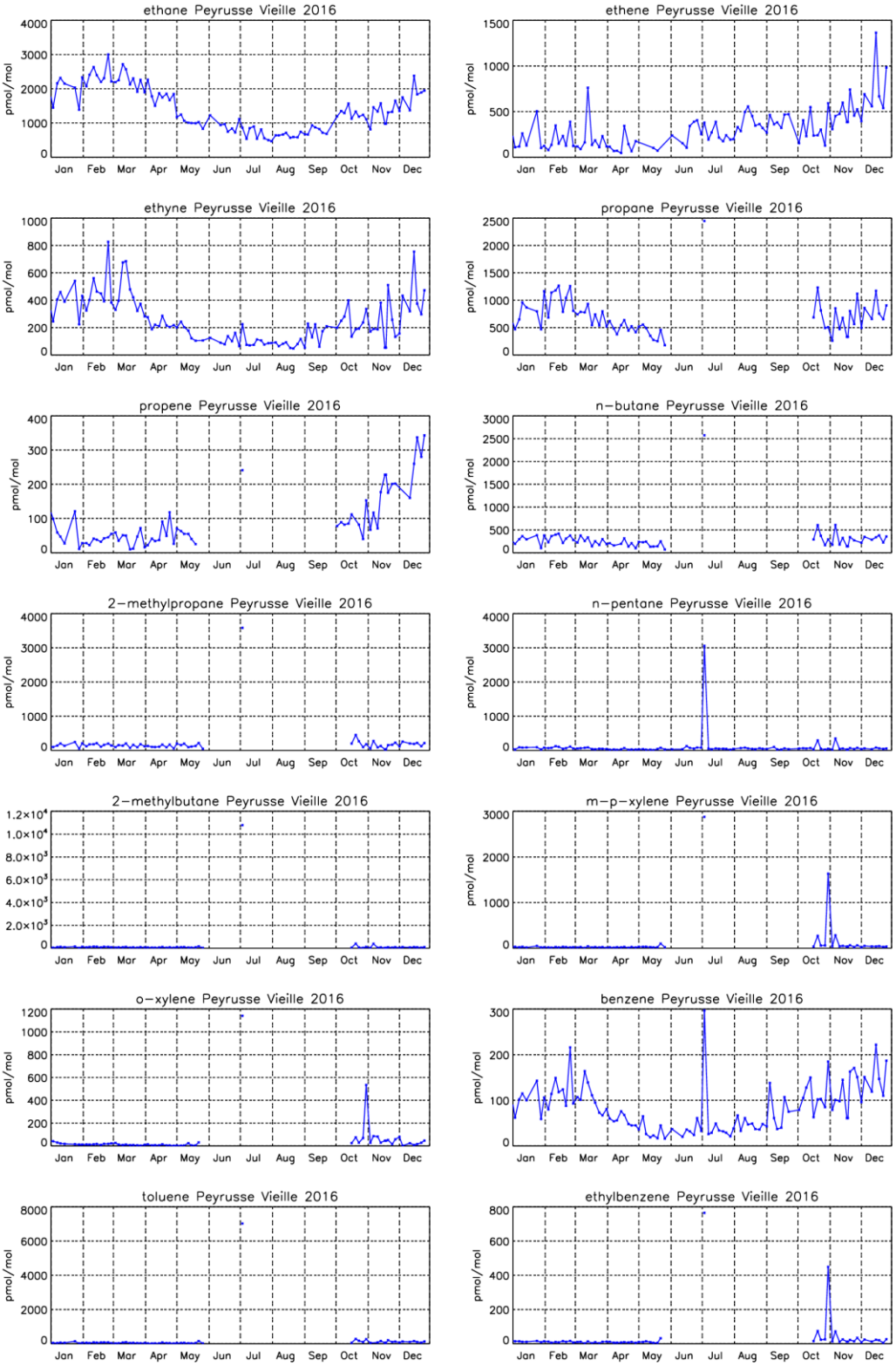
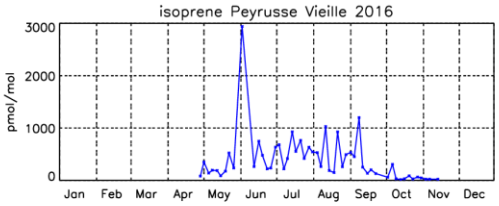
Sep
Oct
Nov
Dec


ethane Peyrusse Vieille 2016
ethene Peyrusse Vieille 2016
ethyne Peyrusse Vieille 2016
propane Peyrusse Vieille 2016
propene Peyrusse Vieille 2016
n−butane Peyrusse Vieille 2016
2−methylpropane Peyrusse Vieille 2016
n−pentane Peyrusse Vieille 2016
2−methylbutane Peyrusse Vieille 2016
m−p−xylene Peyrusse Vieille 2016
o−xylene Peyrusse Vieille 2016
benzene Peyrusse Vieille 2016
toluene Peyrusse Vieille 2016
ethylbenzene Peyrusse Vieille 2016
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


isoprene Peyrusse Vieille 2016
pmol/mol
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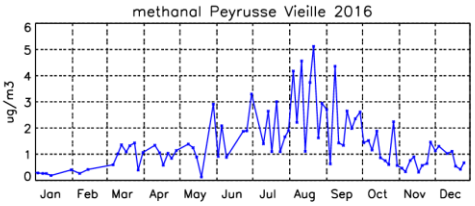
2000
1000
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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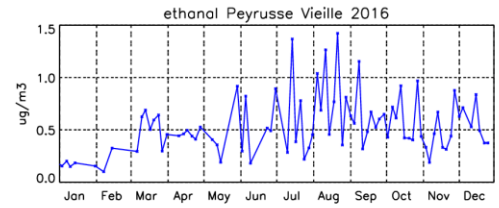
4
3
2
1
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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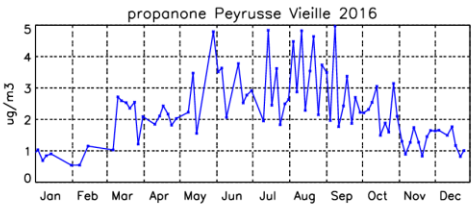
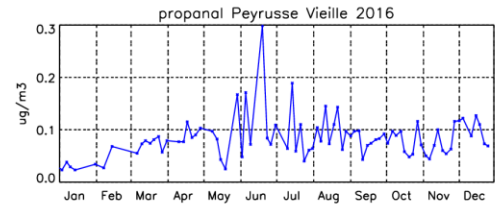
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propanone Peyrusse Vieille 2016
ug/m3
5
4
3
2
1
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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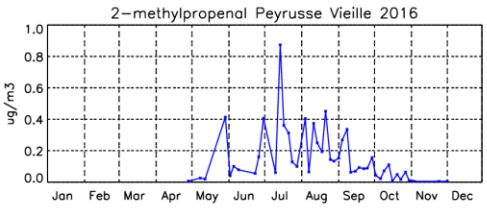
0.1
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2−methylpropenal Peyrusse Vieille 2016
ug/m3
1.0
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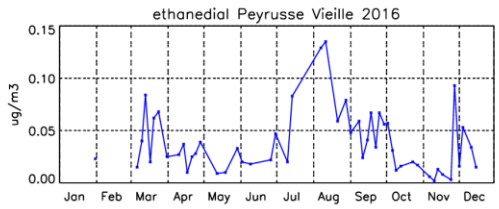
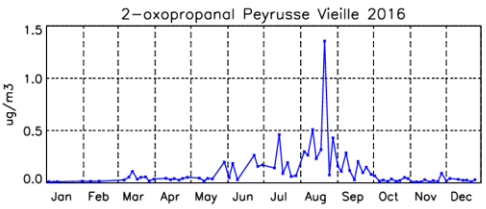
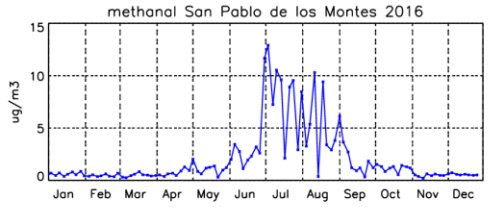
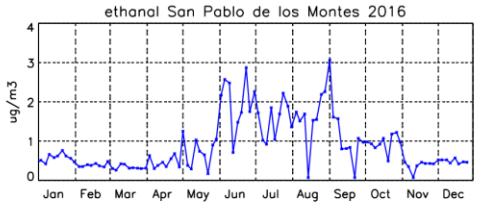
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanedial Peyrusse Vieille 2016
ug/m3
0.15
0.10
0.05
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2−oxopropanal Peyrusse Vieille 2016
ug/m3
1.5
1.0
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


methanal San Pablo de los Montes 2016
ug/m3
0
5
10
15
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanal San Pablo de los Montes 2016
ug/m3
0
1
2
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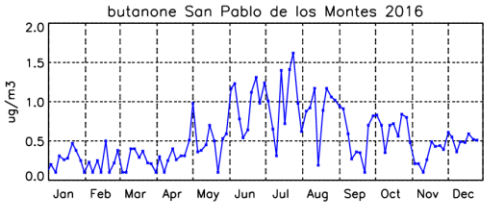
4
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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1.0
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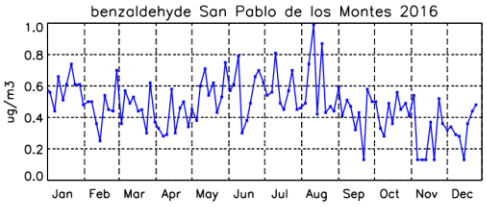
2.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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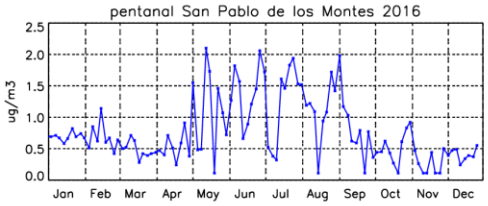
0.4
0.6
0.8
1.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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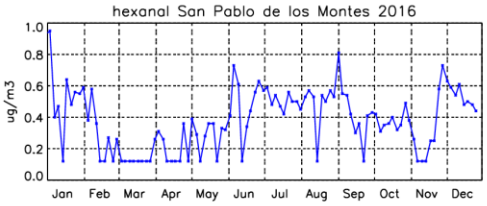
2.0
2.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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ug/m3
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0.8
1.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec