EMEP/CCC-Report 4/2017

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

VOC measurements 2014 - 2015

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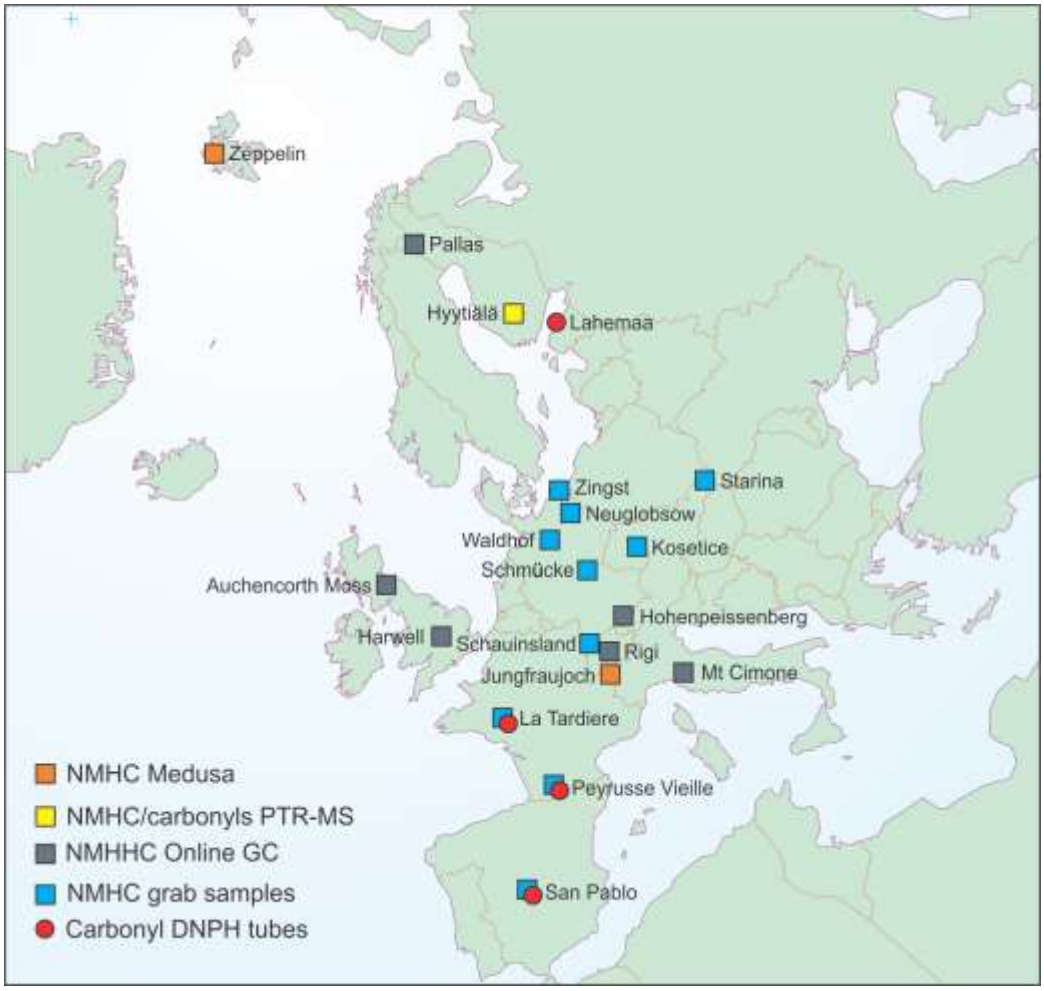

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EMEP Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

VOC measurements 2014 - 2015

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Summary

This report presents VOC measurements carried out during 2014-2015 at EMEP monitoring sites. Totally 20 sites reported VOC data from EMEP VOC sites these two years. Some of the data sets are considered preliminary and are thus 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 (OVOC), sampling in 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 VOCs has 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. The participation in ACTRIS-2 has meant an extensive effort with data checking including detailed discussions between the ACTRIS community and the individual participants. It is no doubt that this extensive effort has gained the EMEP program and has led to improved data quality in general.

The reported EMEP VOC data for the winter half year 2014-2015 show generally highest concentration levels in central Europe and lower levels in the north and at high mountain sites.

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 at three selected sites located at very different regions (Pallas in N-Finland, Hohenpeissenberg in S-Germany and Peyrusse Vielle in SW-France). An iterative procedure for performing a non-linear least squares fit to a seasonal-trend function was tried out. Based on this approach we found marked differences in the trends for the individual species. None of the three sites showed any significant trend in ethane and propane over the period 2000-2015. For other species inspected, we calculated significant reductions over the period. Particularly strong reductions were found for ethyne and benzene corresponding to a 30-60 % reduction over the 2000-2015 period.

VOC measurements 2014 - 2015

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.

The measurements of VOC within EMEP started with the collection of grab samples of light hydrocarbons in the middle of 1992 and measurements of carbonyls in 1993. In the beginning five stations were included in the monitoring programme, Rucava (LV10), Košetice (CZ03), Waldhof (DE02), Tänikon (CH32) and Donon (FR08). Since then the number and selection of VOC measurement sites have changed several times.

The 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). Highlights and findings from the EMEP VOC programme have also been presented in a number of scientific papers (Lindskog et al., 1995; Solberg et al., 1996; Hov et al., 1997; Solberg et al., 2001; Borbon et al., 2004; Hakola et al., 2006; Tørseth et al., 2012; Koohkan et al., 2013; Waked et al., 2016).

In 2016 no EMEP data report was published for VOCs due to problems meeting the reporting deadline, partly due to the very rigorous data quality control within ACTRIS-2 as well as adaption to the new data submission formats in EBAS. Thus, the present report documents the VOC measurements from two years, 2014-2015.

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: sulphur, NO_x, VOCs and ammonia. These ceilings were negotiated on the basis of scientific assessments of pollution effects and abatement options. Parties whose emissions have a more severe environmental or health impact and whose emissions are relatively cheap to reduce will have to make the biggest cuts. According to the Protocol, Europe's sulphur emissions should be cut by at least 63%, its NO_x emissions by 41%, its VOC emissions by 40% and its ammonia emissions by 17% compared to 1990. The Protocol also sets tight limit values for specific emission sources (e.g. combustion plant, electricity production, dry cleaning, cars and lorries) and requires best available techniques to be used to keep emissions down. VOC emissions from such products as paints or aerosols will also have to be cut.

In 2012 a revised Gothenburg protocol was adopted. A main difference from the previous protocol is that the emission ceilings now are given as percentage reductions from 2005 to 2020 and thereafter. Furthermore, $PM_{2.5}$ 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. Harmonisation 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. The alcohols (methanol and ethanol) are likely to become more important in the future due to increased use of biofuels in vehicles. Furthermore, terpenes are important as precursors for secondary organic aerosols. These compounds would be of interest to include in the EMEP monitoring as well, but require other sampling methods and instrumentations than presently used for the hydrocarbons and carbonyls.

In the 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 harmonise 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 it includes a large number of partners with experience in VOC monitoring, including most of the 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.

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 2014-2015

No EMEP VOC data report for 2014 was published and thus results from the two years 2014-2015 are documented in the following.

2.1 The station network

The location of the EMEP monitoring sites for VOC in 2014-2015 is shown in Figure 1 and an overview of the measurement programme and the responsible laboratories is given in Table 2. Totally 20 measurement sites are included in the list. For some of the sites, the data values are not included in this report since they are still regarded preliminary either due to data format technicalities or due to unresolved questions to the data quality and the data filtering (flagging local influences). Furthermore, VOC data reported to EBAS from sites not labeled as an EMEP VOC site have not been included in this report. This regards e.g. the sites Cape Verde (CV01) located outside the EMEP domain and the suburban site Sirta (FR20) at the outskirts of Paris.

NMHC monitoring are carried out at all sites except Lahemaa in Estonia while carbonyls are only measured regularly at a few sites; two sites in France, one in Spain and at the Estonian site Lahemaa which started reporting carbonyl data for 2014.

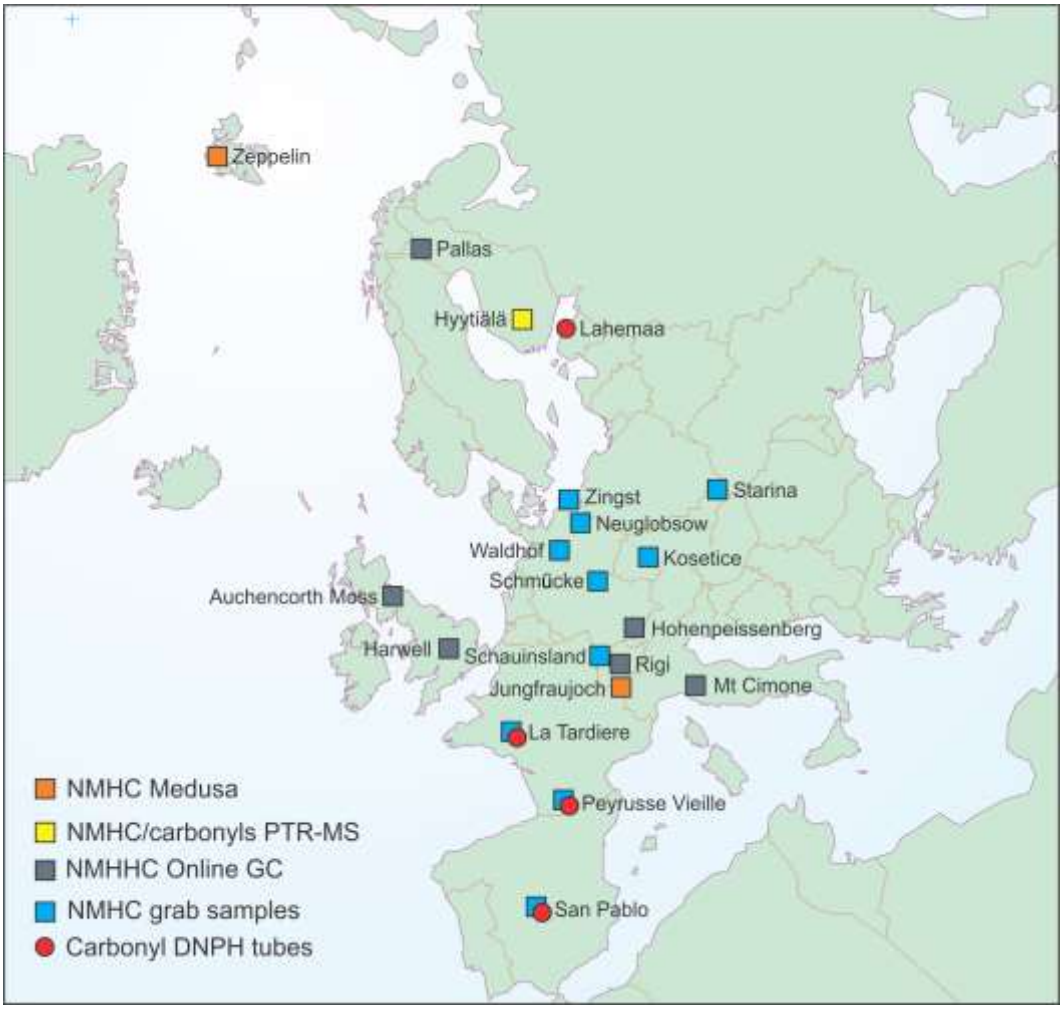

Figure 1: Monitoring sites for VOC in 2014-2015.

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 (carbonyls) the original method based on sampling in DNPH adsorption tubes with subsequent lab analyses is still the only method used at the reported EMEP sites.

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.

Furthermore, as seen by Table 2, based on data quality checks (described in the next chapter) data from some of the sites were classified as preliminary and unsettled and were not included in the present report. The reasons varied between the stations as explained briefly below.

UK data (Harwell (GB36) and Auchencorth Moss (GB48)): NMHC data for 2014 and 2015 have been received. However, a number of issues related to data values and the use of data flags remain unanswered and unsettled, and the datasets could therefore not be accepted as final, approved data when this report was written.

NMHC data from Slovakia (Starina, SK06): Data have been received for 2014 and 2015. Inspection of the data revealed substantial differences compared to the expected general levels and compared to other sites for some of the species. Furthermore, there have been known problems with the instrumentation at the site for some time. Until a more detailed evaluation and assessment of the data and the station is carried out, the data are considered preliminary. A station audit plus some kind of association with the extensive QA work provided within ACTRIS-2 is recommended.

NMHC data from Spain (San Pablo ES01): As for Slovakia, data have been received for 2014 and 2015, however the data have revealed significant differences compared to the expected general levels and compared to other sites for some of the species. There has been a dialogue between NILU and the data provider and the status of the data is still regarded unsettled.

Table 2: VOC monitoring at EMEP sites in 2014-2015. 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.	NO42	Cont.	NILU	-	MEDUSA	y	
Pallas	FI96	Cont.	FMI	-	GC/MS	y	
Hyytiälä	FI50	Cont	UHel	-	PTR-MS.	y	
Lahemaa	EE09	-	EERC	Reg.	DNPH tubes	n	Started reporting for the year 2014
Auchencorth Moss	GB48	Cont.	Ricardo	-	GC/MS	y	Prelim. data not included in the report
Harwell	GB36	Cont.	Ricardo	-	GC/MS	y	Prelim. data not included in the report
Waldhof	DE02	Reg.	UBA	-	Flask samples	y[3)]	Twice//week
Schauinsland	DE03	Reg.	UBA	-	"	y[3)]	"
Neuglobsow	DE07	Reg.	UBA	-	"	y[3)]	"
Schmücke	DE08	Reg.	UBA	-	"	y[3)]	"
Zingst	DE09	Reg.	UBA	-	"	y[3)]	"
Hohenpeissenberg	DE43	Daily	DWD	-	GC/FID	y	2/day (noon, midnight)
Košetice	CZ03	Reg.	CHMI	-	Flask samples	y	Twice/week
Starina	SK06	Scat.	SMHI	-	Flask samples	n	Prelim. data not included in the report
Jungfraujoch	CH01	Cont.	EMPA	-	MEDUSA	y	
Rigi	CH05	Cont.	EMPA	-	GC/MS	y	
Peyrusse Vieille	FR13	Reg.	EMD	Reg.	Flask/DNPH samples	y	NMHC 2/week. OVOC 1/week
La Tardière	FR15	Reg.	EMD	Reg.	"	y	"
Mt. Cimone	IT09	Cont.	UU	-	GC/MS	y	
San Pablo	ES01	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
AEA = AEA Technology
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 is 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 NMHC 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).

The ACTRIS consortium as mentioned above took a central role in the quality control of the laboratories' reported data from the regular monitoring for institutes participating in that project and a comprehensive Standard Operation Procedures (SOP) manual for VOCs has been developed in the project.

NMHC data from ACTRIS-stations were presented by the representatives and discussed in detail at dedicated workshops. Additionally, data from UBA, not formally partner of ACTRIS-2, were included. Then, based on certain statistical tools developed within ACTRIS, potential outliers and errors in the data were discussed, and recommendations for data base flagging were provided by the workshops. EMPA, the ACTRIS-2 work package leader for VOCs, had a key role in this process. In addition, detailed inspection of the data has been done by NILU in parallel with EMPA. Dialogues between EMPA, NILU and the data providers have been essential in this work and a web based issue tracker have been developed and used in the process.

The elaborate QA work on NMHC provided by ACTRIS-2 and EMPA in particular has meant a significant improvement for the general quality of the data. Hopefully, these procedures and routines will flow into the part 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.

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

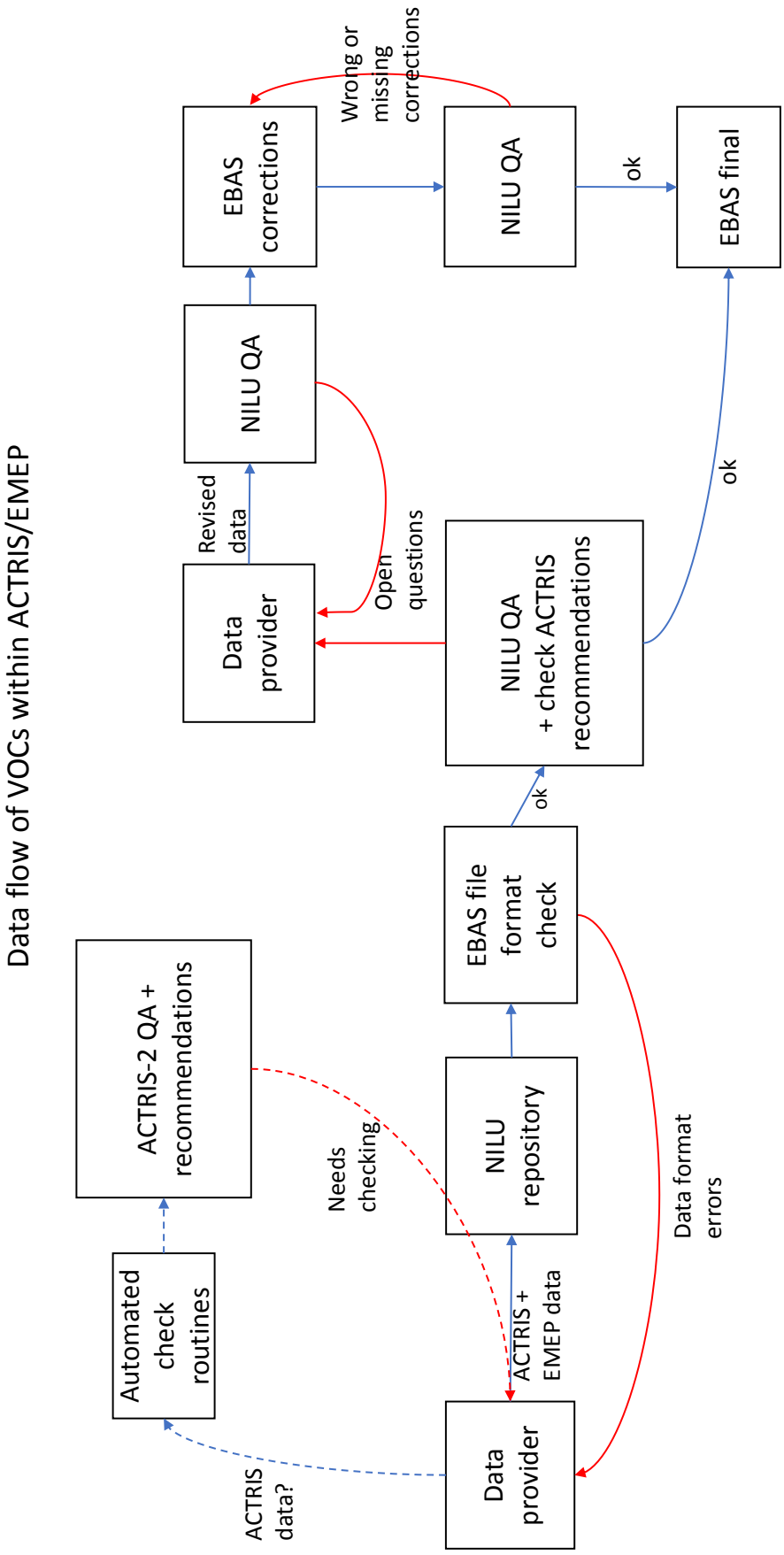

Figure 2: Flow diagram for the VOC data submitted within ACTRIS/EMEP.

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

Since then, various intercomparisons for VOCs have been carried out, e.g. through the projects 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) 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 were seen, indicating measurement difficulties with these compounds. The spread in the results were, however, much less for laboratories using a NPL standard for calibration (Aas et al., 2001). Thus, it 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.

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.

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) was distributed to all participating laboratories for analyses. For calibration, the laboratories were asked to use their own certified multicomponent standards, which should be traceable to the GAW scale. Three laboratories served as reference labs, analyzing the mother 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) turned out to be the most problematic species to measure. 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 reasonably good results although not among the best ones. 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.

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 2014-2015

3.1 General

Time series of the diurnal means of all compounds during 2014-2015 are given in the Appendix. A comparison of the seasonal mean and percentile concentrations of NMHC from the six months winter half year (Oct-Mar) is given in standard box and whisker plots in Figure 3. The stations are arranged from north to south. Considering that the sites span a wide area from the Arctic to Southern Europe, the hydrocarbon winter mean levels are fairly uniform. A systematic pattern is seen though, with generally lower levels in the north (Zeppelin, Pallas) and at the high mountain sites (Jungfraujoch and Mt Cimone).

Similar 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.

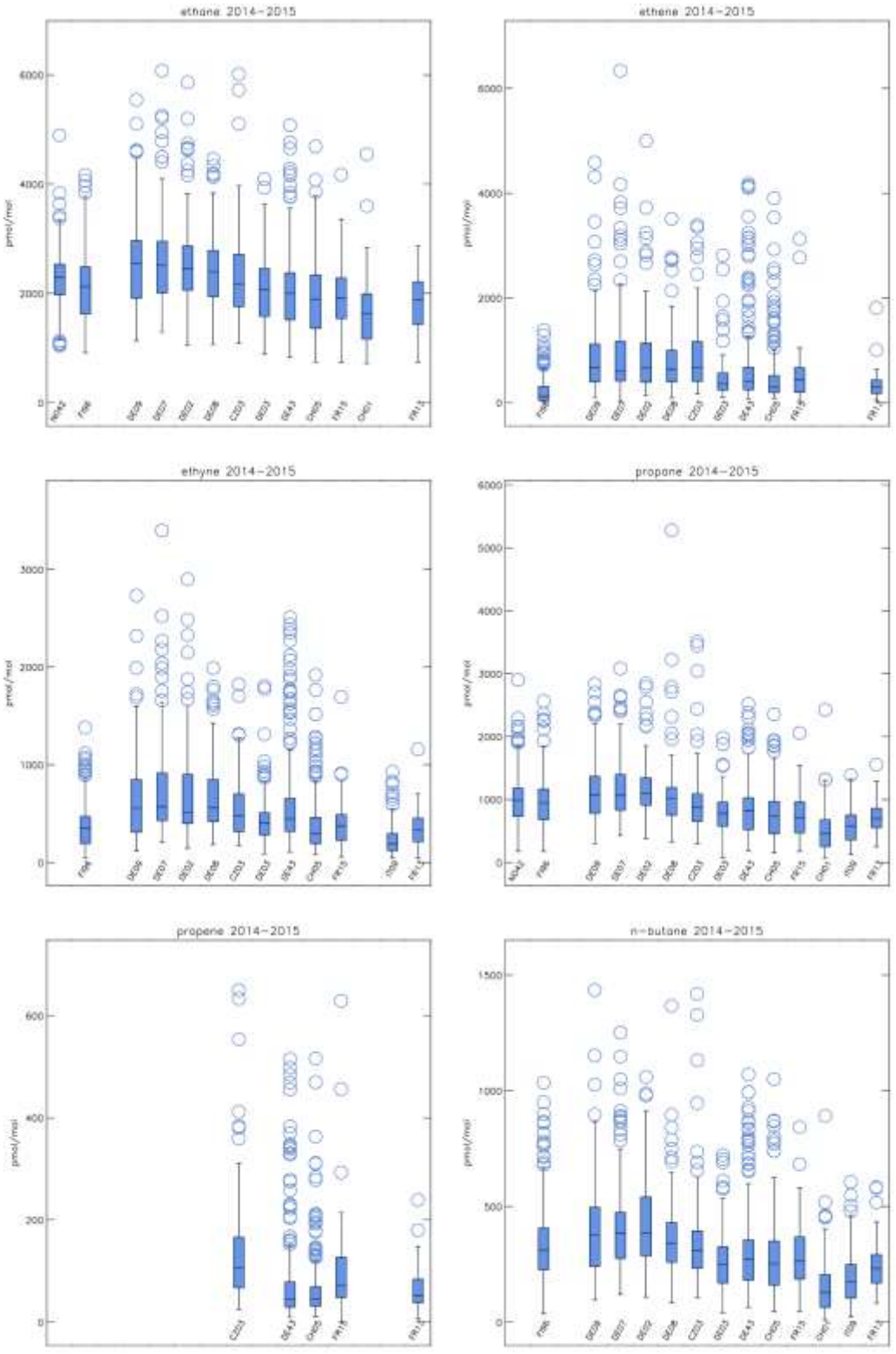

Figure 3: Box- and whisker-diagrams for hydrocarbons during the 6-months winter half years in 2014-2015 (Oct.-Dec.). The boxes enclose the 25- and 75-percentile with the median marked inside. The whiskers extend out to the max or min value, or to the 1.5 times of the 25p or 75p if there is data beyond this range. Outliers are identified with circles.

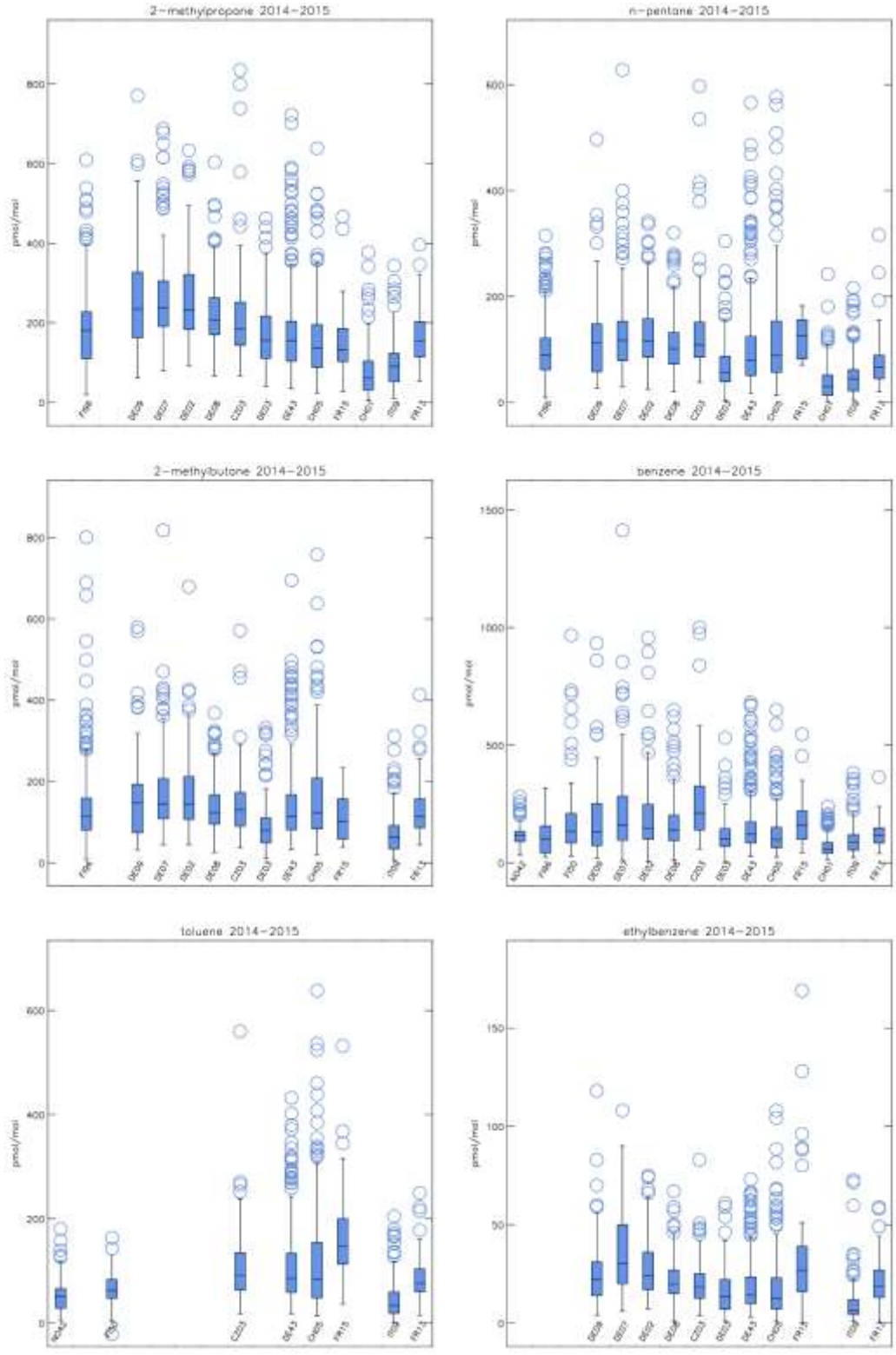

Figure 3. (contd)

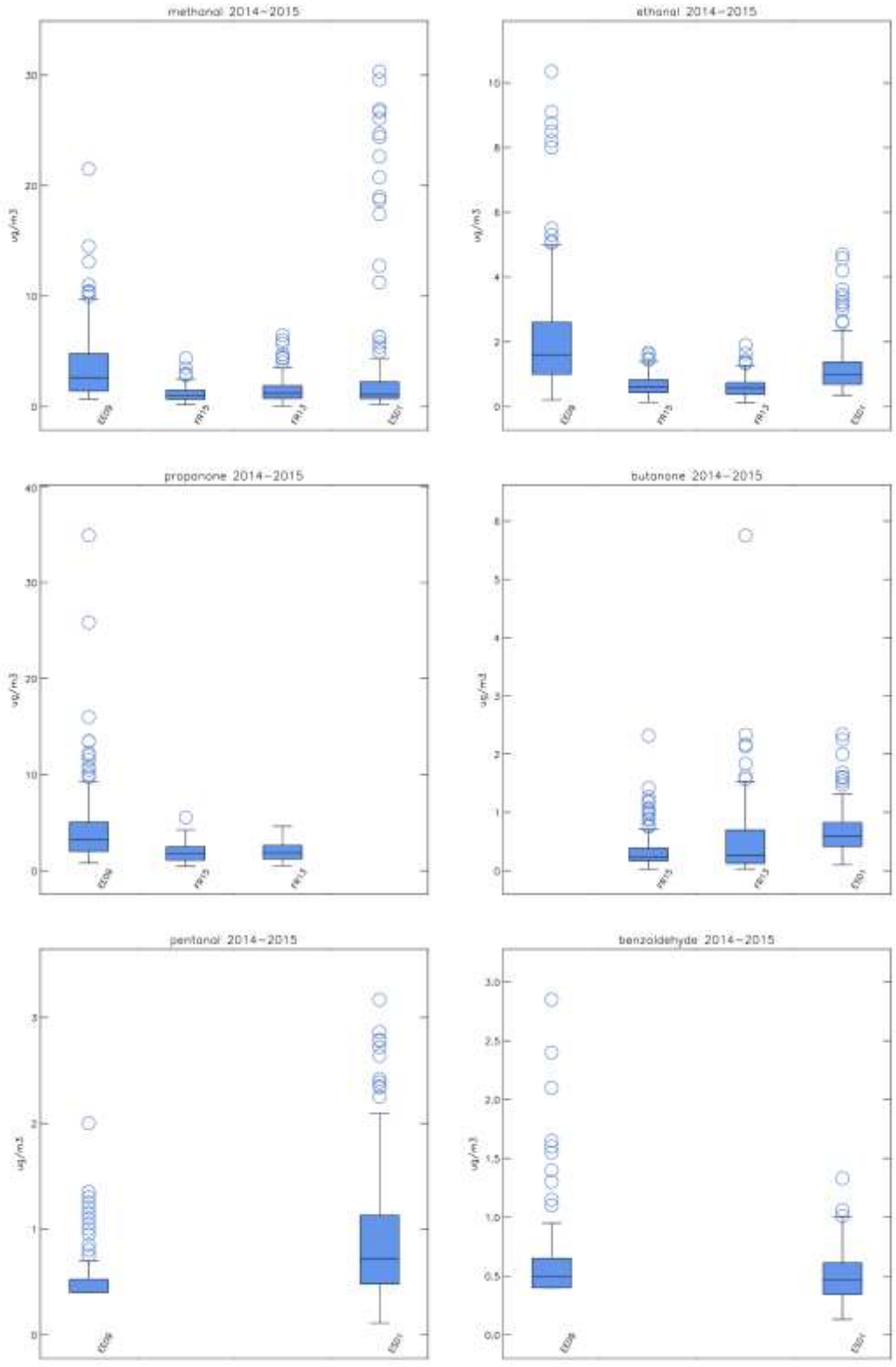

Figure 4: Box- and whisker-diagrams for carbonyls for the whole years 2014-2015. The boxes enclose the 25- and 75-percentile with the median marked inside. The whiskers extend out to the max or min value, or to the 1.5 times of the 25p or 75p if there is data beyond this range. Outliers are identified with circles.

3.2 Regional distribution of VOC

Figure 5 shows maps with the stations' median concentrations of light hydrocarbons for the winter half year (October-March) in 2014 and 2015 taken together. 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 at 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. For the period 2014-2015 data were only available for Lahemaa (EE09), Peyrusse Vieille (FR13), La Tardiere (FR15) and San Pablo (ES01).

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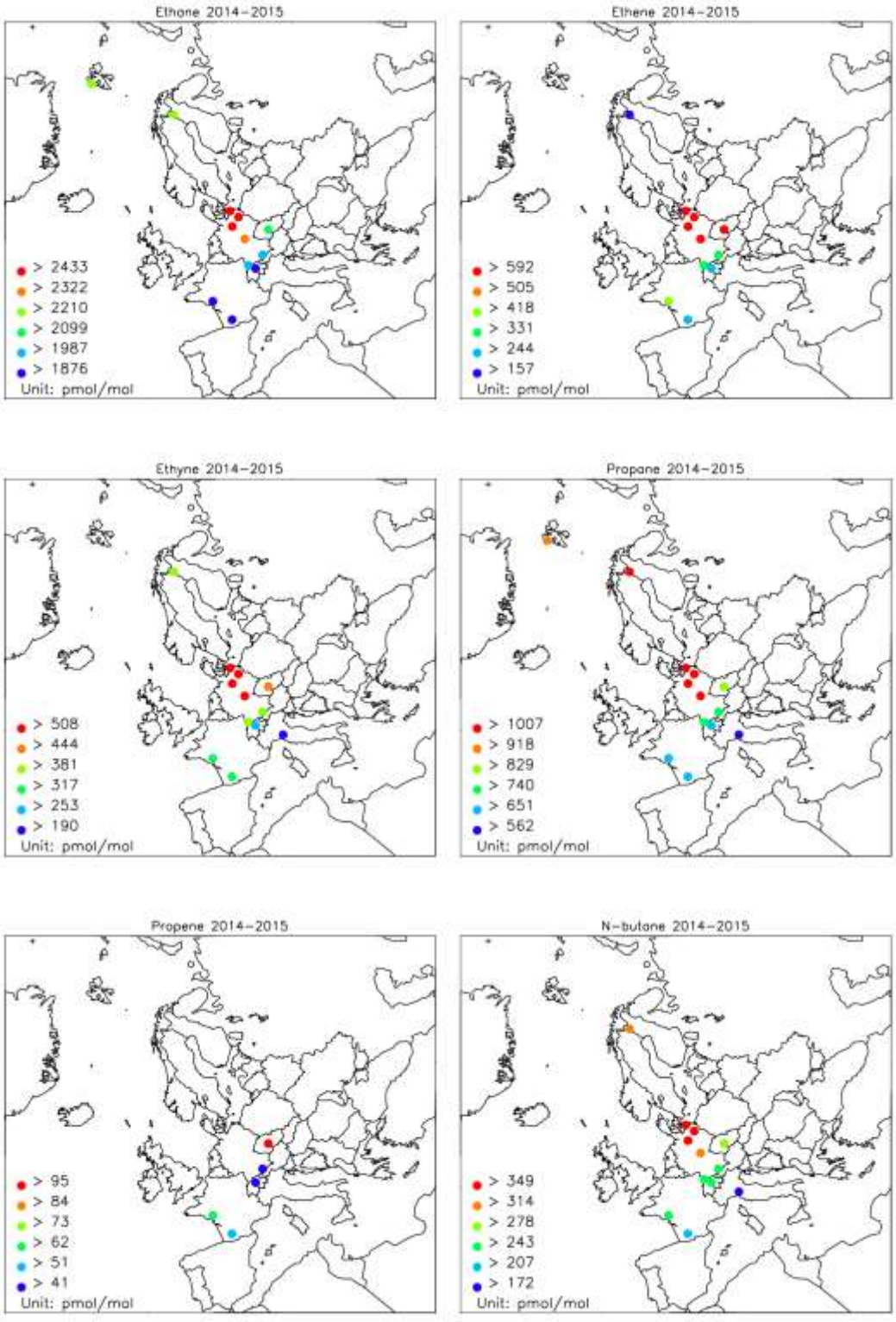

Figure 5: Median concentration of NMHCs in the winter half years (Oct-Mar) 2014-2015 taken together.

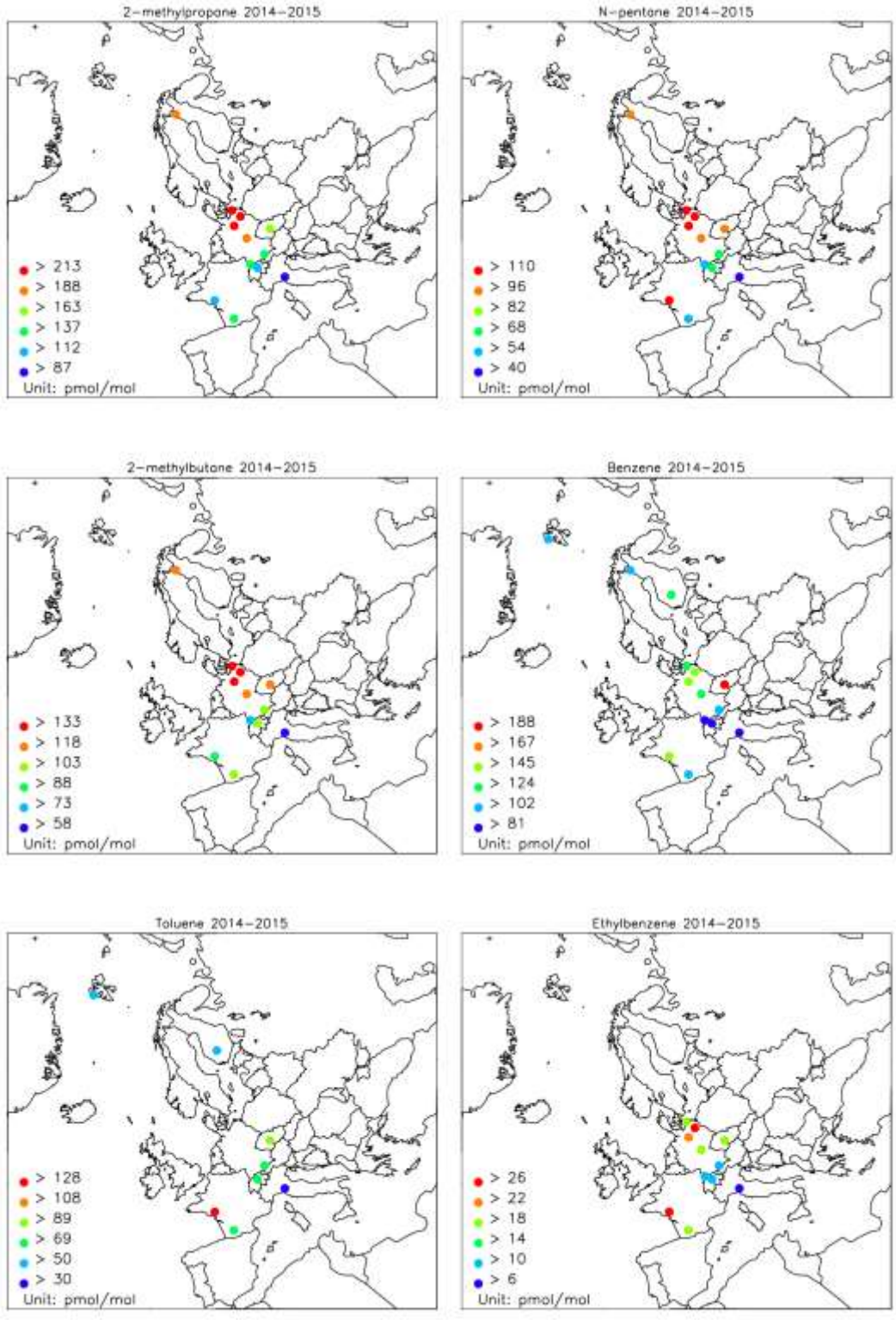

Figure 5 (contd.).

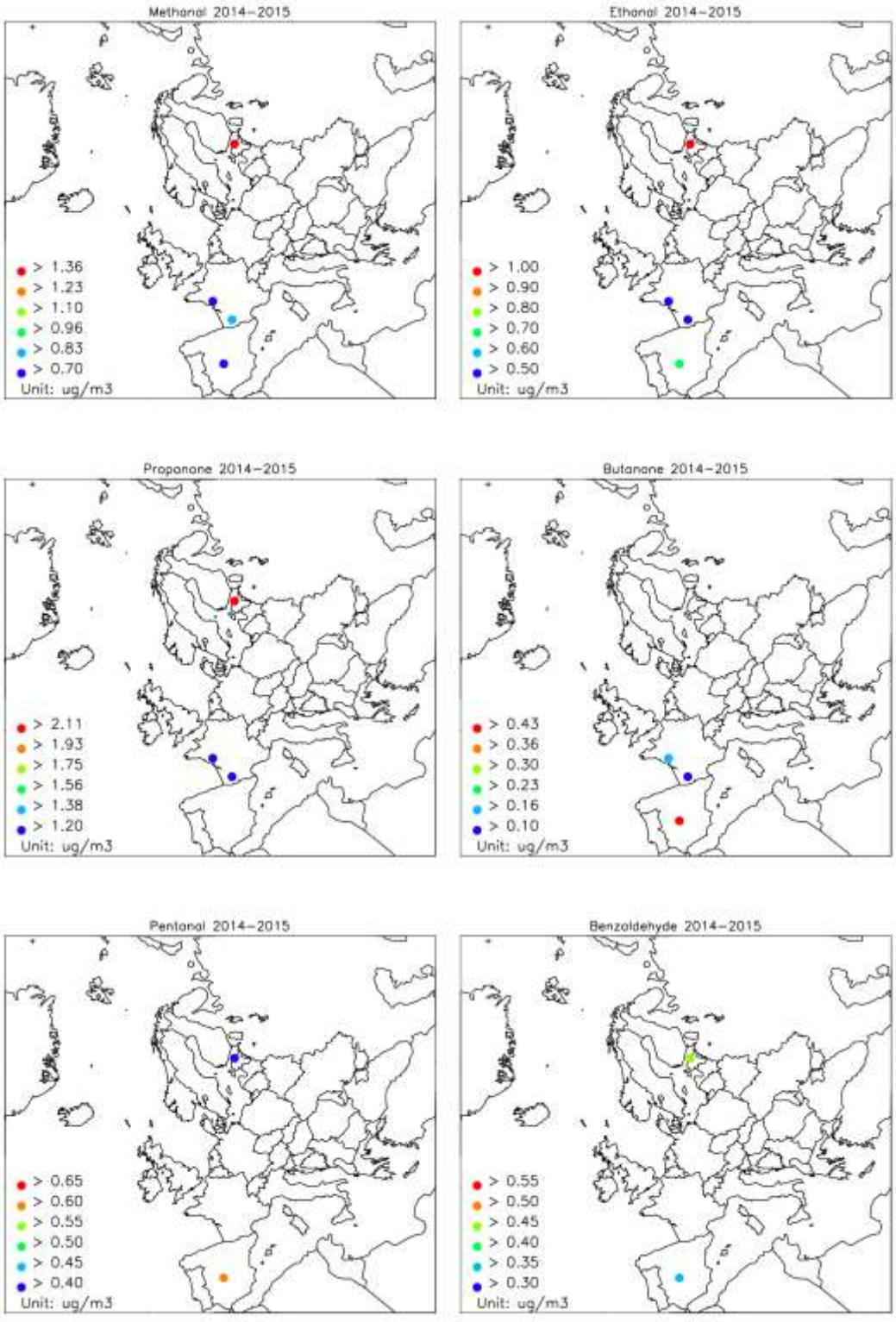

Figure 6: Median concentration of carbonyls over the whole year for 2014-2015 taken together.

4. Long-term trends in VOC

According to the official emission data, there have been substantial reductions in the anthropogenic emissions of VOCs 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).

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}. They also found a marked difference between ethane and propane on one hand, showing relatively stable levels and other alkanes showing pronounced declines on the other hand.

Long-term monitoring data from an urban network in Switzerland (Hüglin, pers. comm.) also shows strong declines in the concentration levels of NMHCs and OVOCs from the start of the 1990s to 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 was the main source region for high concentration levels at Pallas.

A simple 1:1 relationship between observed VOC concentrations at rural background sites and the overall European emission numbers are 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 three long-term running EMEP sites over the period 2000-2015. 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:

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

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 a number of iteration below 10 using a very strict tolerance criteria for the iteration. 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 these coefficients.

Eq 1 represents a simple seasonal cycle with a mean level a_0, amplitude a_1 and seasonal phase displacement a_2 that are changing with time according to a long-term exponential change with the a_3 coefficient defining the rate of either a growth ($a_3 > 0$), a decline ($a_3 < 0$) or no trend ($a_3 = 0$).

To assure positive solutions the least squares fit was applied to log-transformed data, i.e. 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.

Figure 7 shows the daily concentration values (blue marks) together with the fitted curve in red for the three sites Pallas (FI96), Hohenpeissenberg (DE43) and Peyrusse Vielle (FR13) which all have sufficiently long time series of NMHCs. The percentage change from the year 2000 to 2015 are given in the title whenever a significant trend was found. We assumed significant trends when a_3-2σ and a_3+2σ were either both negative (significant reduction) or both positive (significant increase), where σ is the confidence interval as mentioned above. For the significant trends we estimated the total percentage change from 2000 to 2015 by calculating the annual mean values in these two years based on the fitted curve.

The results in Figure 7 indicate clear differences in the long-term trends between the individual species. For ethane and propane none of the three sites show significant trends. For the other species (ethene, ethyne, n-butane and benzene) marked reductions are found at all sites except for n-butane at Peyrusse Vieille. Strongest relative trends are found for ethyne and benzene with total reductions over this period corresponding to a 30-60 % reduction. Smaller downward trend (still significant) are found for n-butane corresponding to a reduction of 17-18 % over this period.

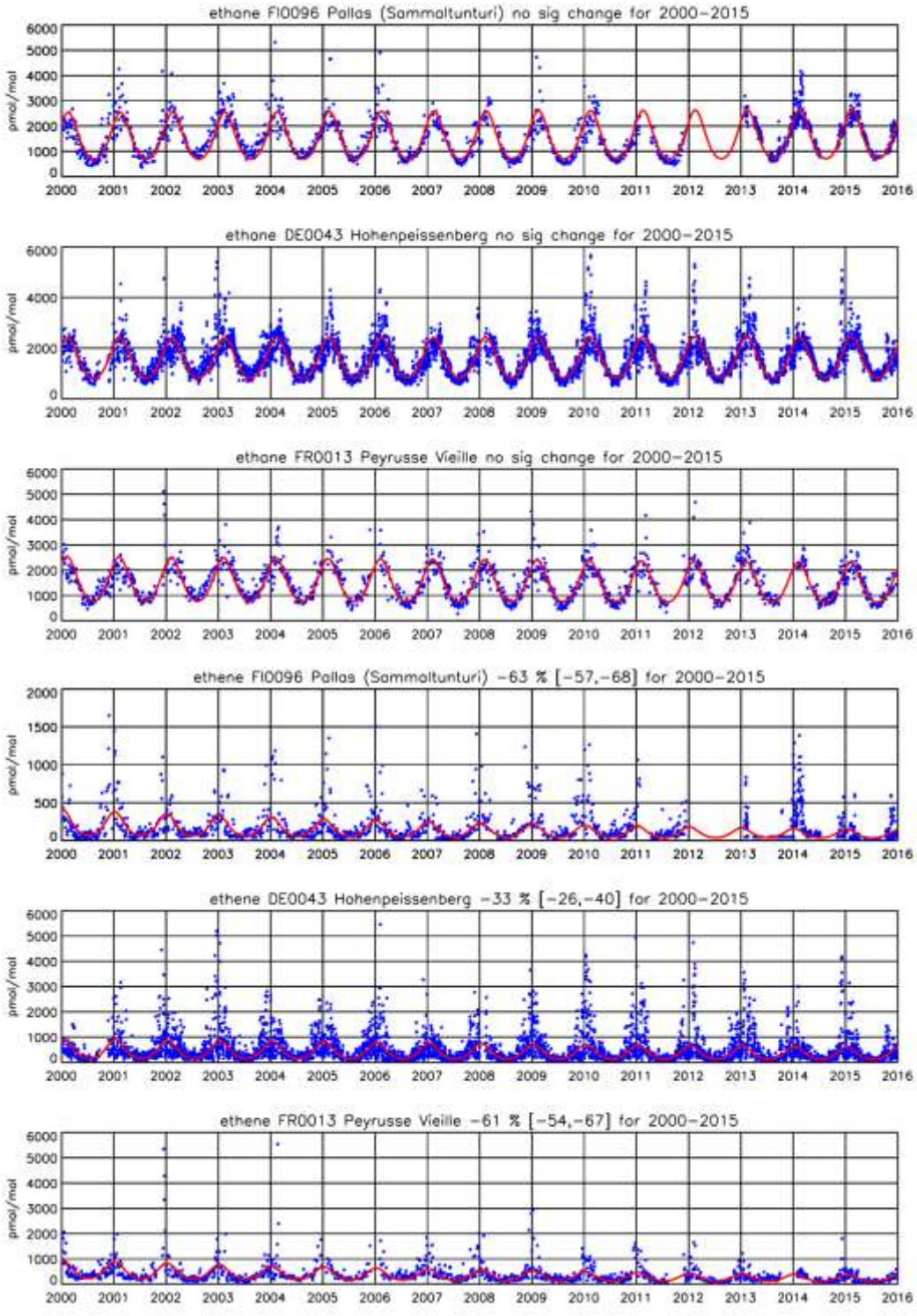

Figure 7: Daily averaged measured concentration of hydrocarbons (blue) at three EMEP sites during 2000-2015 together with the estimated seasonal trend curve in red (see text for explanation). Significant trends are given with confidence intervals in the header as percentage change from 2000 to 2015.

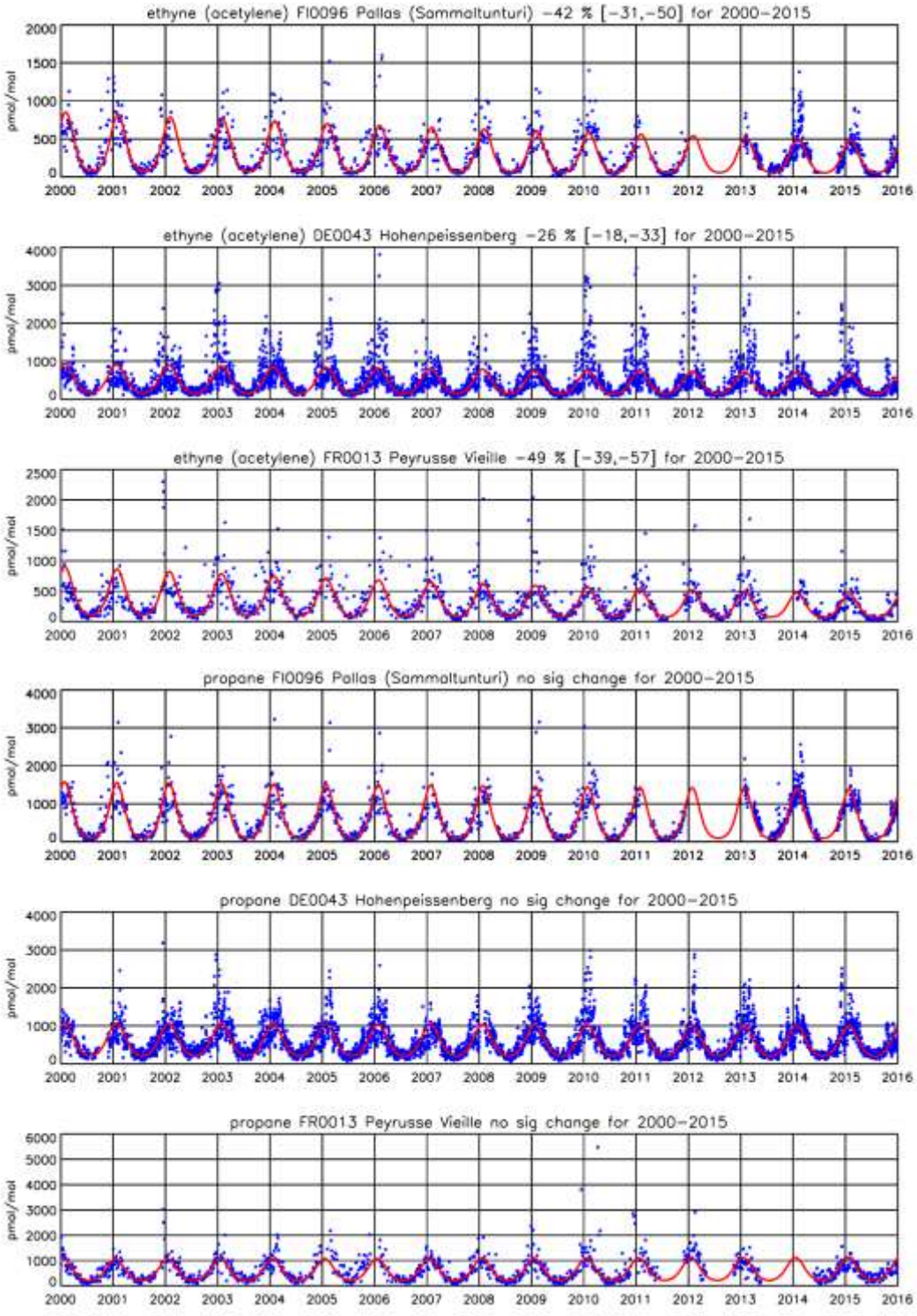

Figure 7 (contd.).

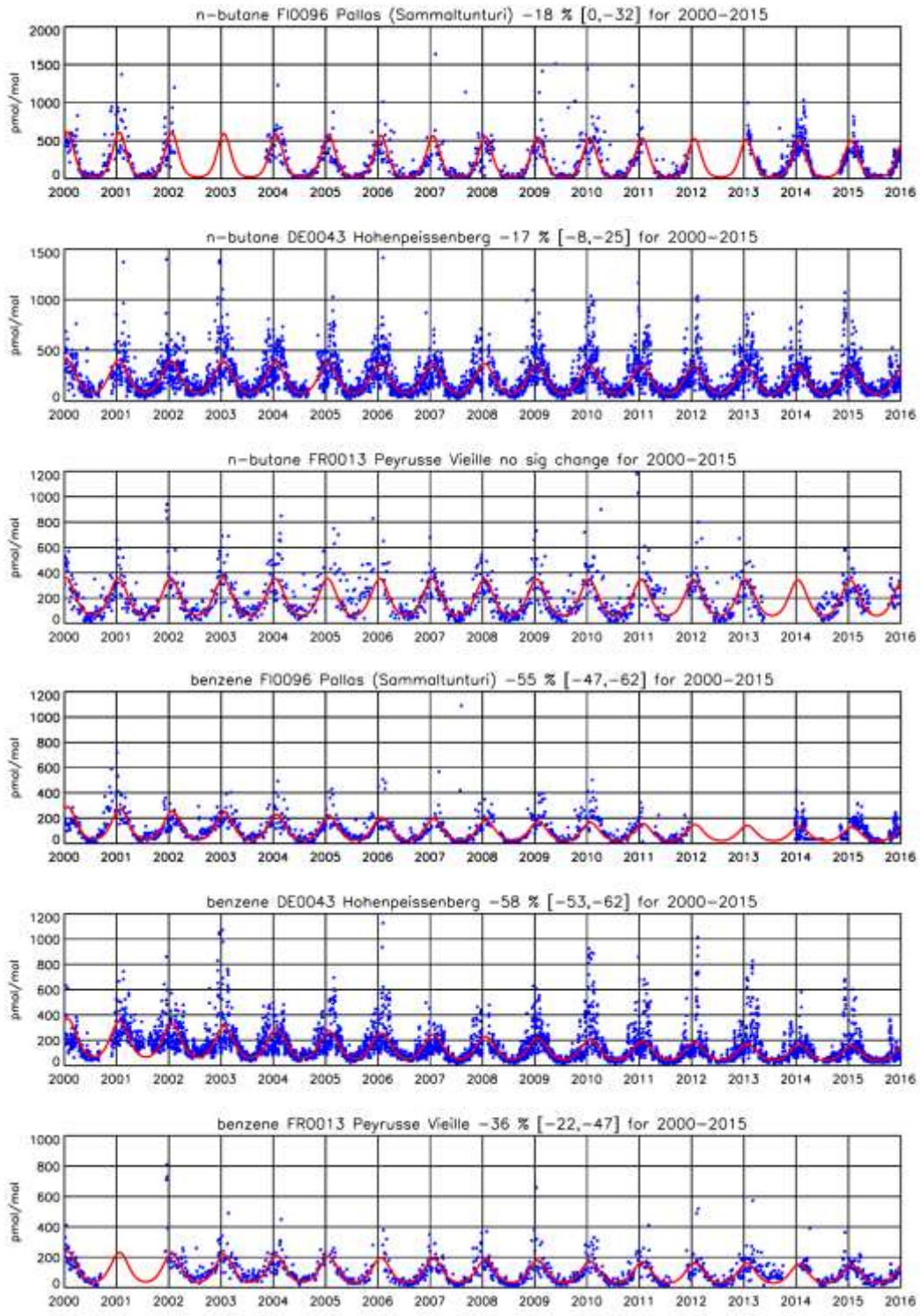

Figure 7 (contd.).

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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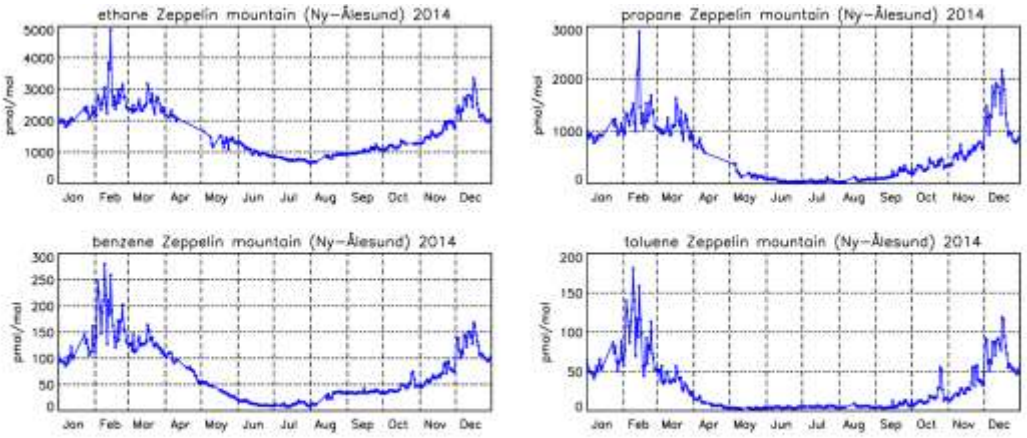
Appendix

Time series of daily means of VOC measured in 2014 and 2015 listed from north to south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propane Zeppelin mountain (Ny-Ålesund) 2014
benzene Zeppelin mountain (Ny-Ålesund) 2014
toluene Zeppelin mountain (Ny-Ålesund) 2014
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


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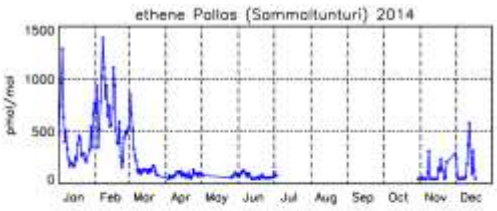
pmol/mol
0
1000
2000
3000
4000
5000
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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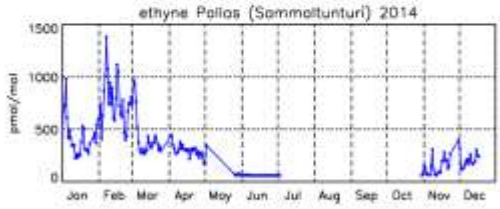
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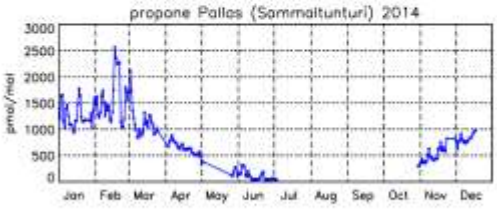
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500
1000
1500
2000
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3000
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


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800
1000
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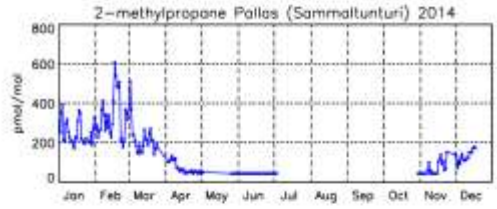
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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800
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


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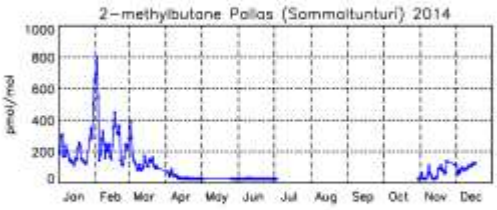
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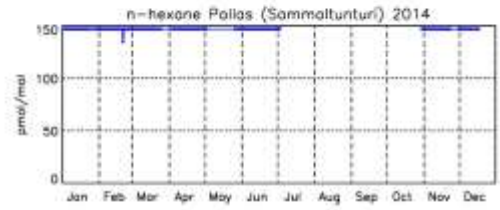
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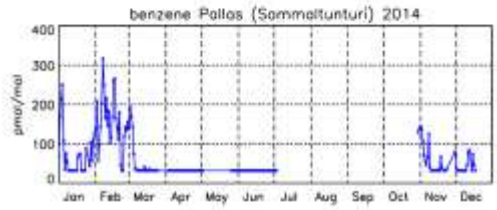
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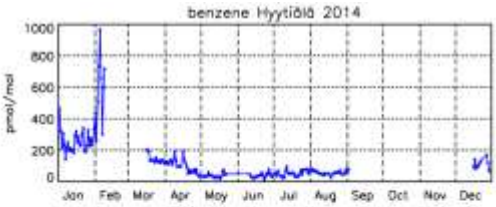
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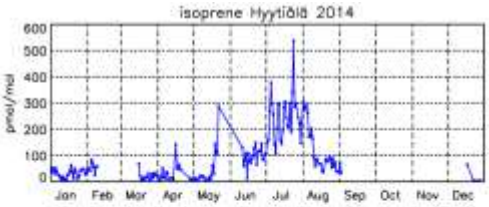
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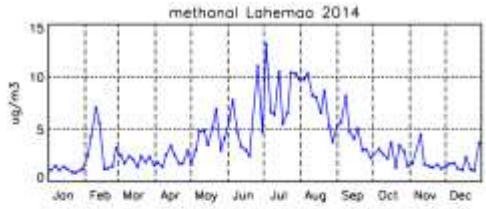
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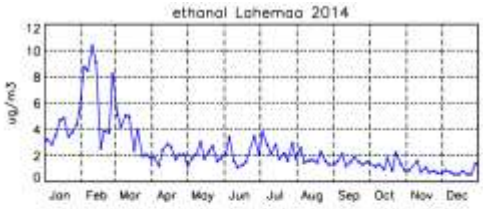
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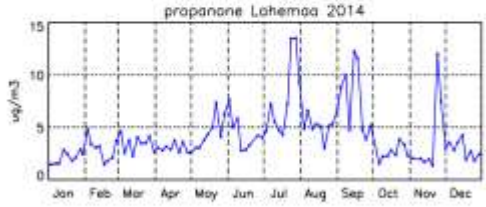
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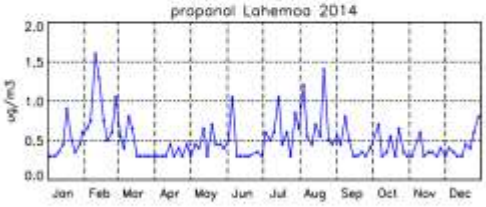
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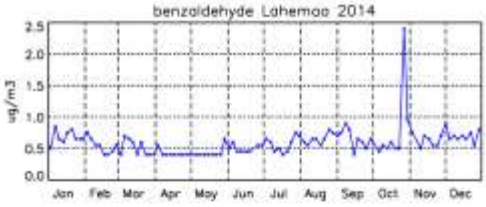
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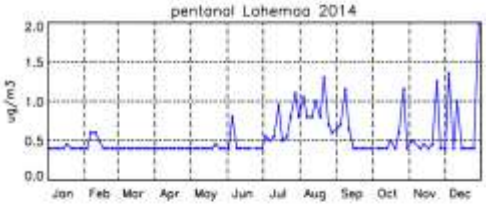
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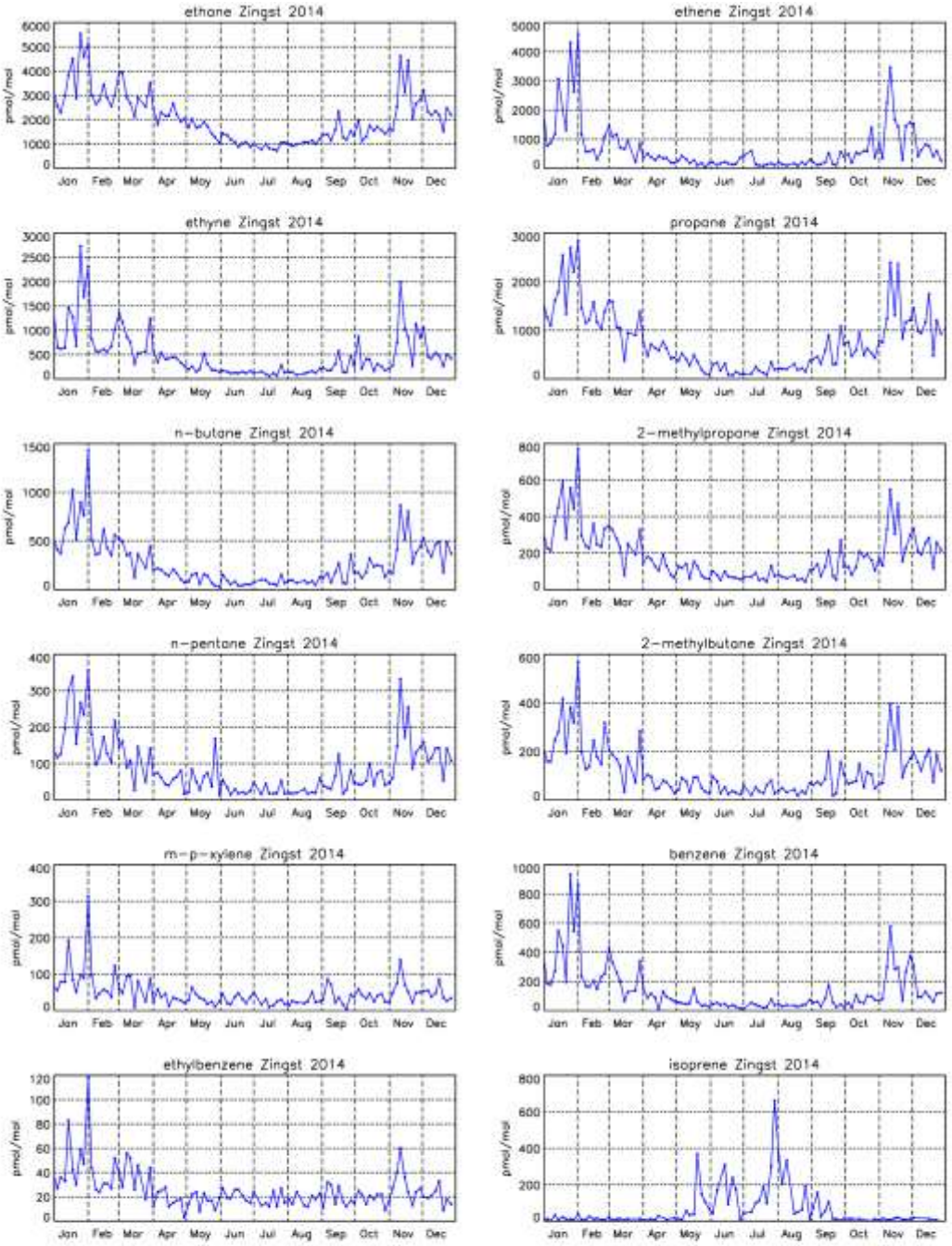
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benzene Zingst 2014
ethylbenzene Zingst 2014
isoprene Zingst 2014
pmol/mol
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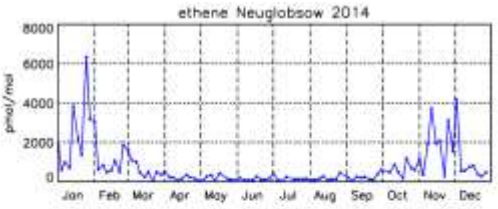
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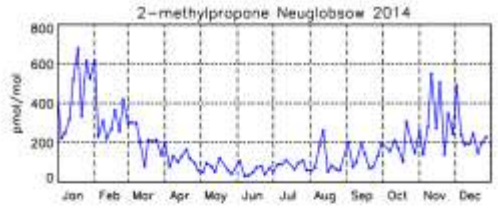
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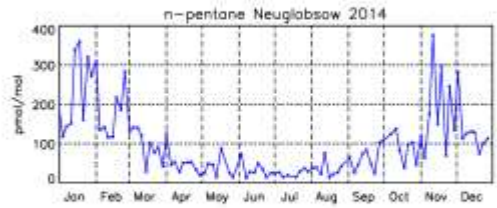
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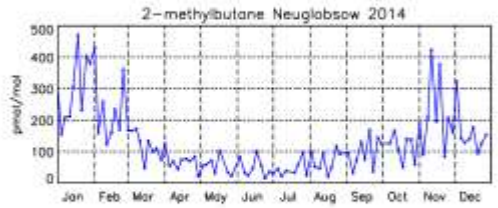
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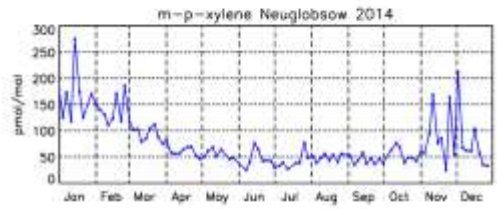
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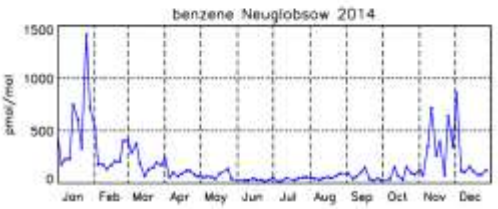
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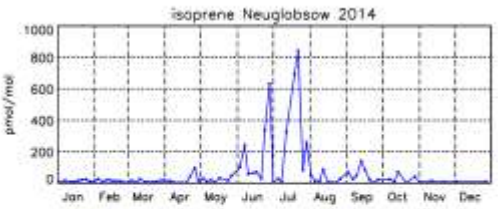
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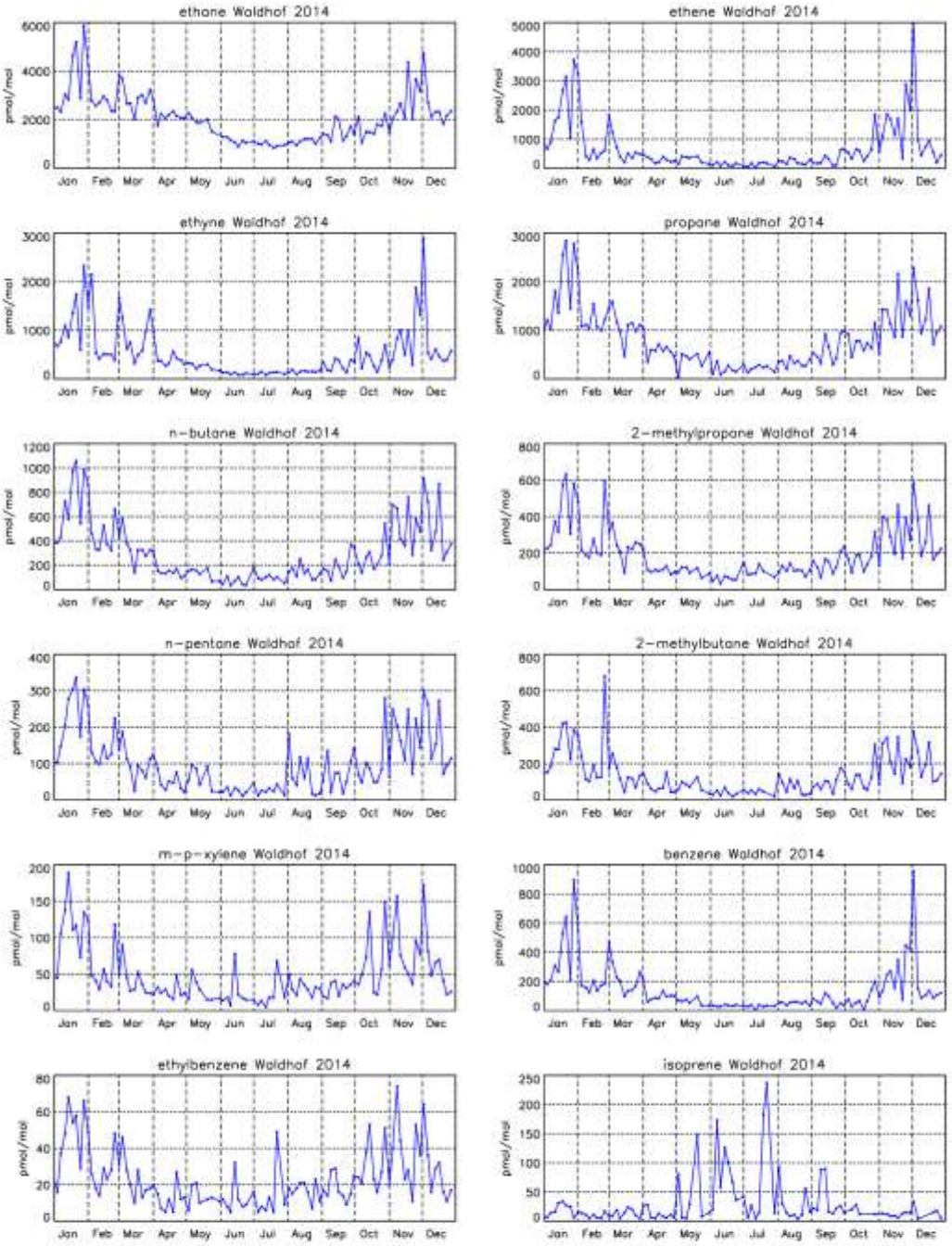
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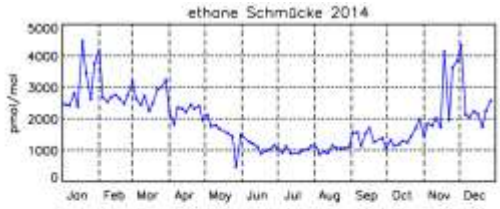
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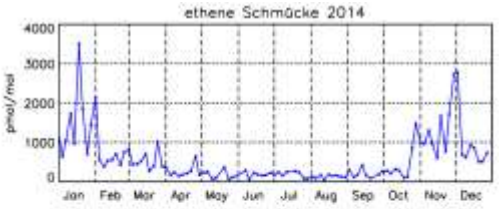
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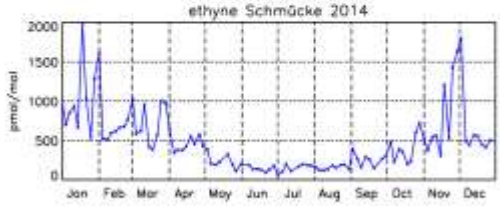
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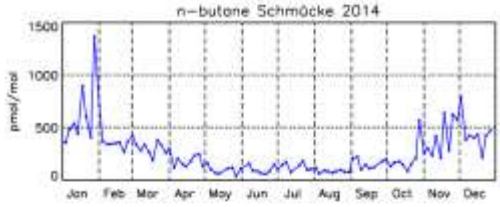
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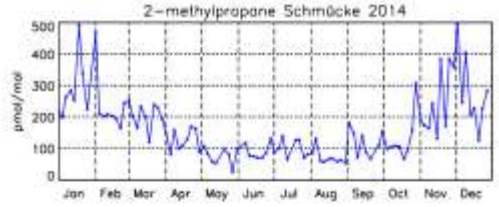
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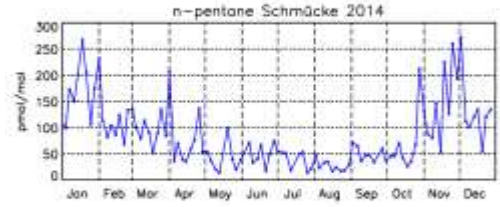
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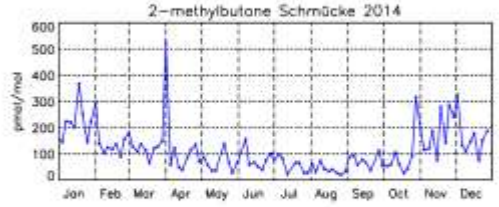
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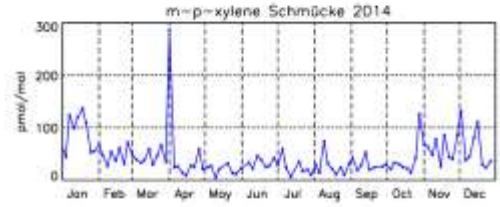
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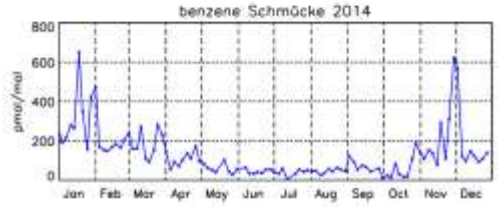
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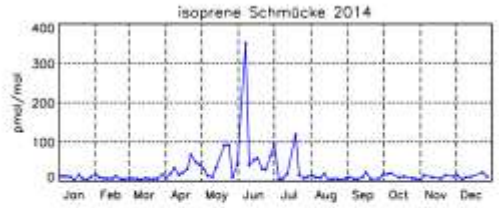
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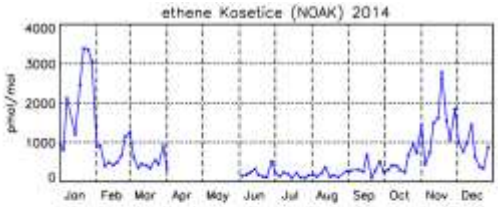
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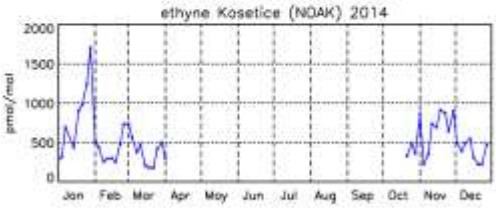
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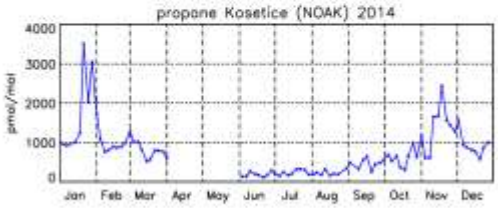
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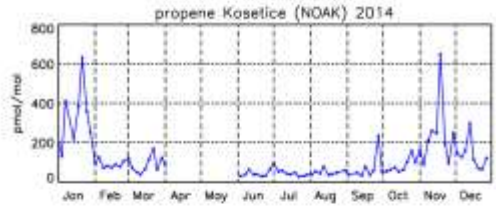
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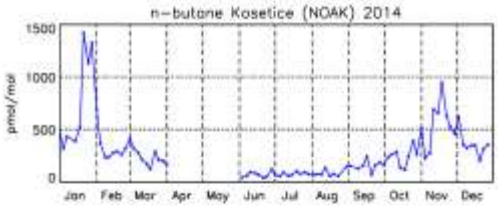
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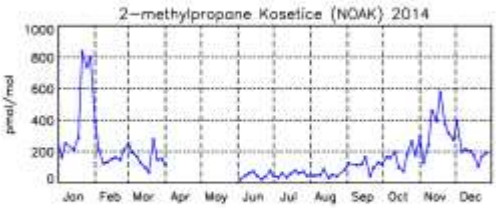
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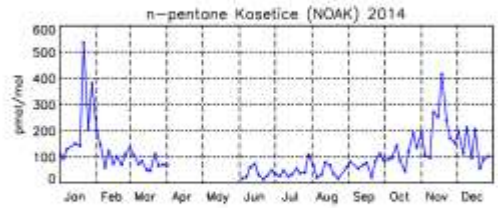
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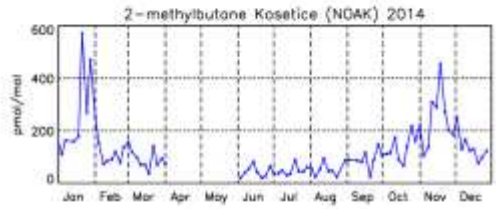
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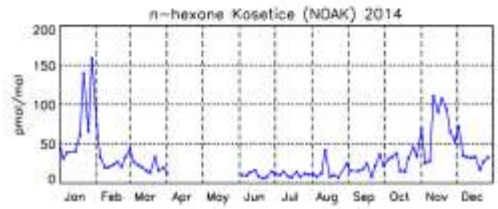
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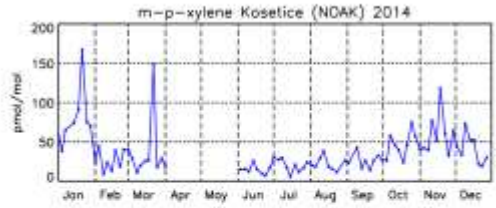
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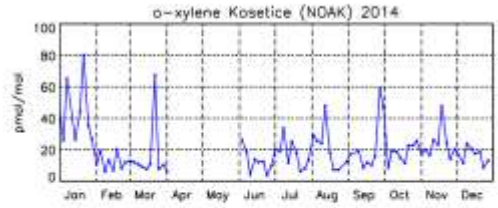
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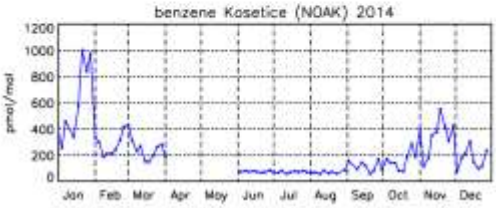
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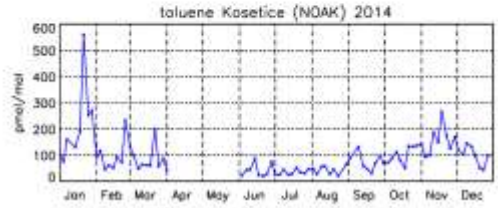
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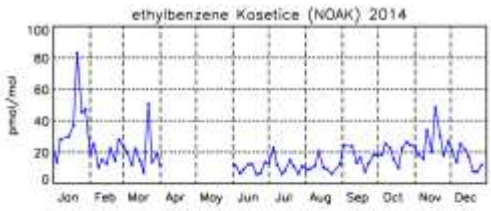
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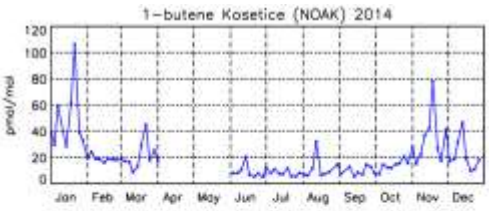
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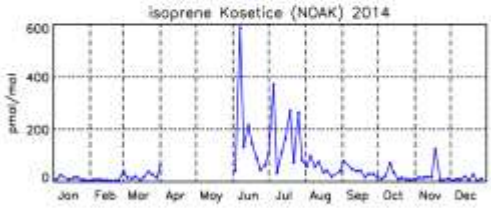
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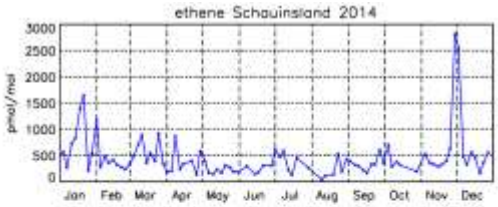
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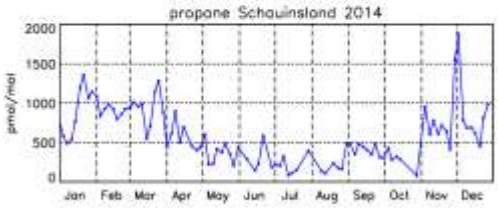
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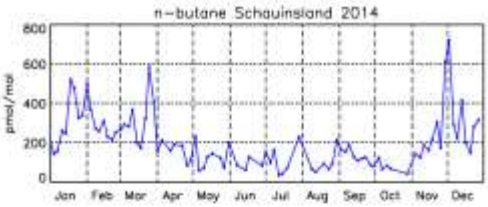
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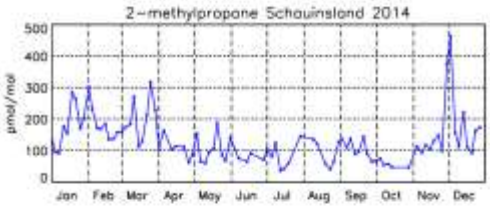
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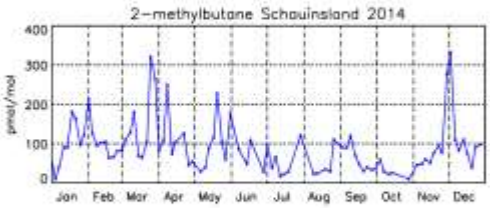
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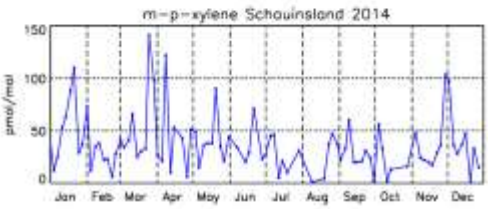
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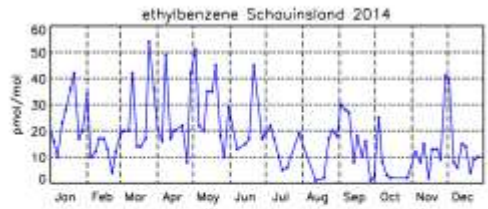
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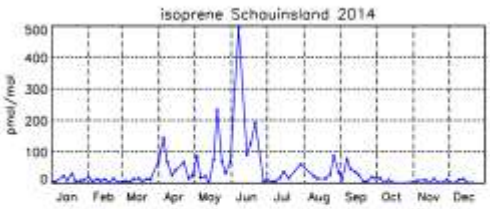
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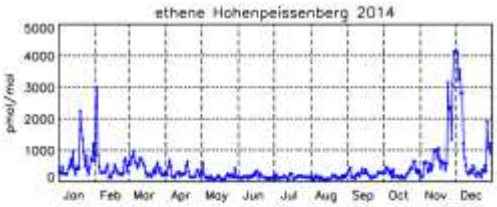
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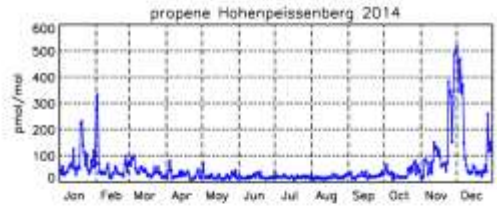
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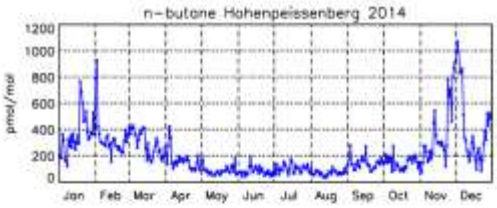
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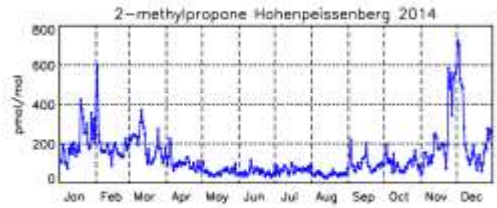
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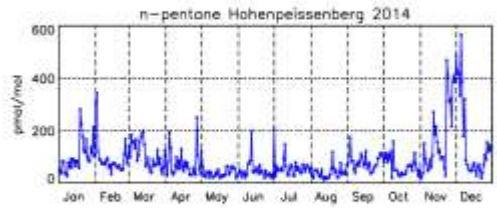
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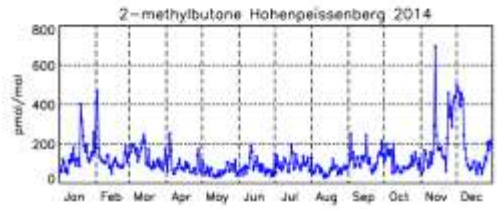
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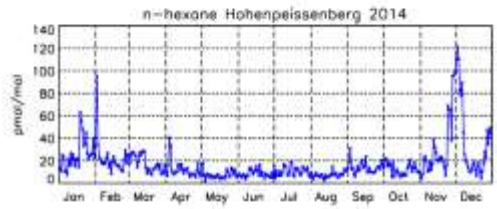
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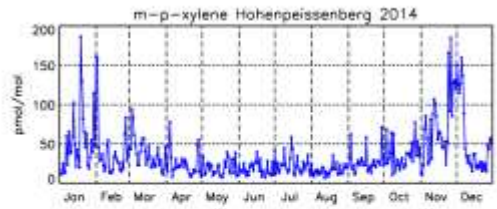
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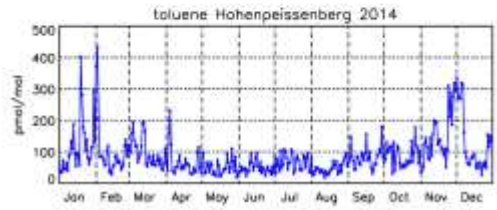
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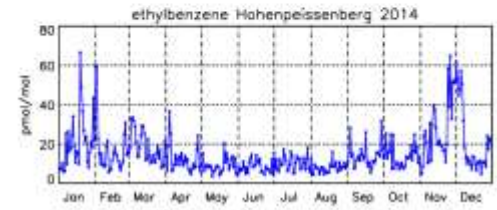
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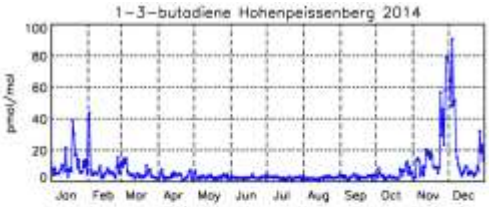
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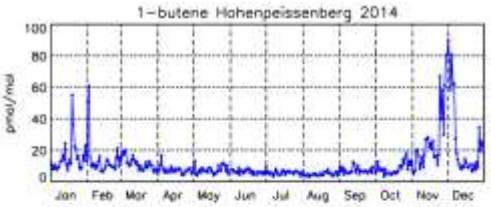
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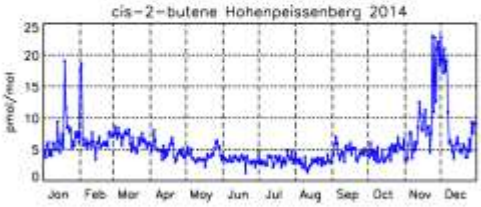
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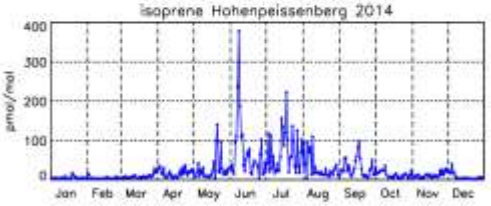
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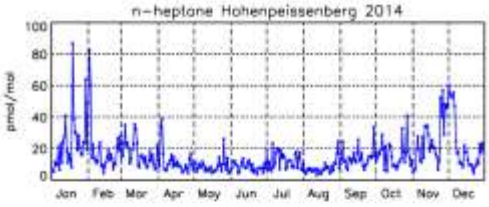
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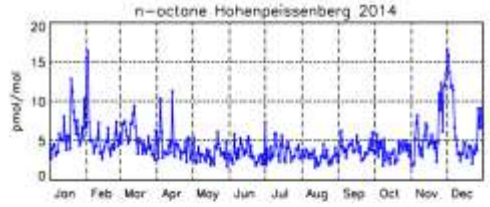
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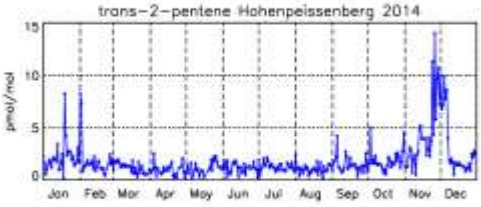
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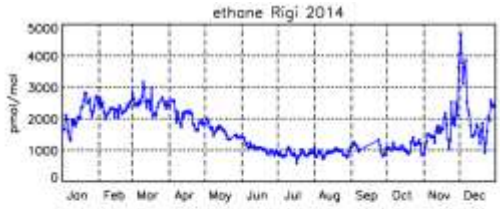
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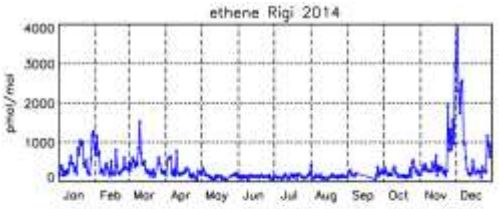
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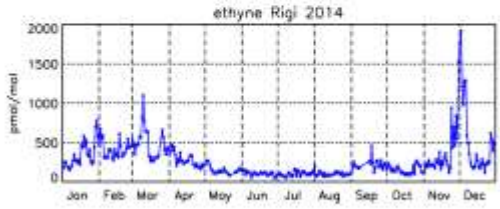
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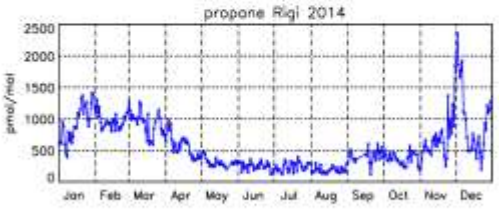
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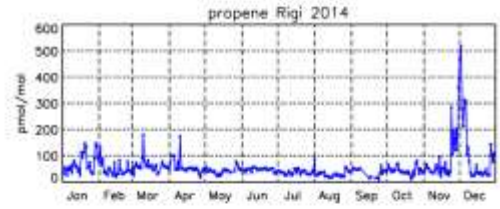
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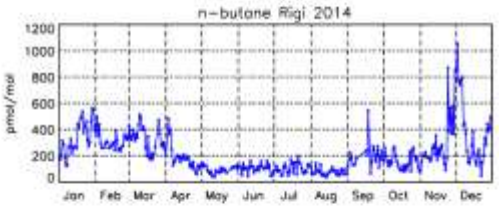
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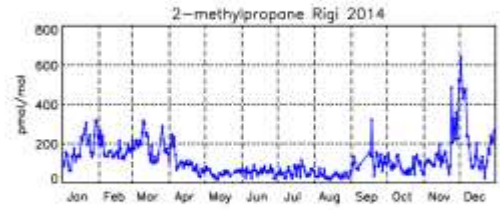
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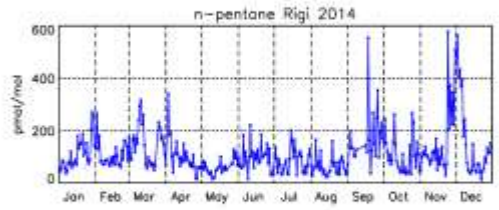
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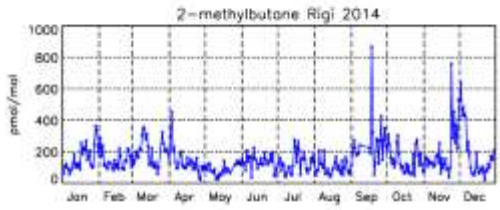
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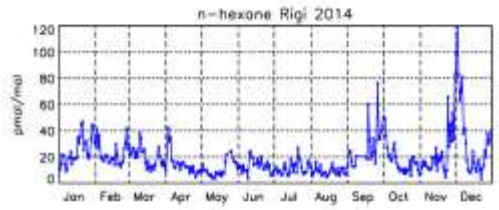
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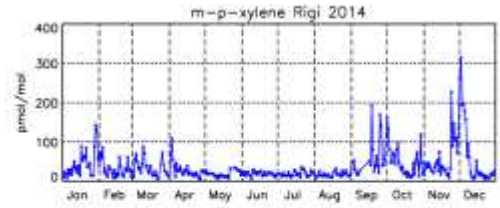
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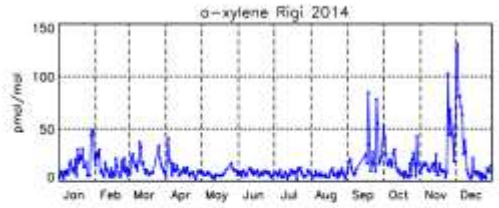
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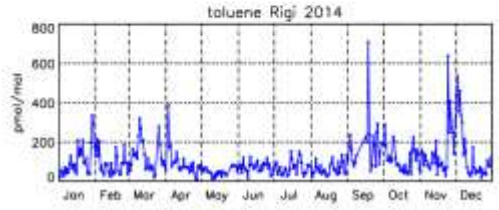
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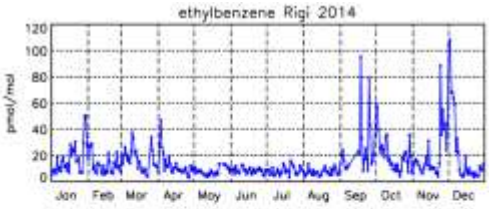
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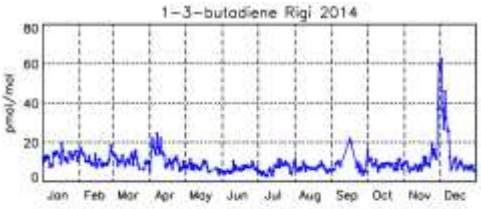
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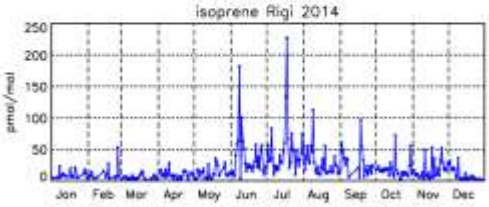
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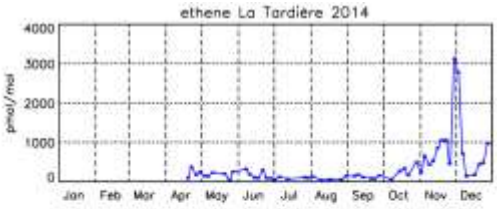
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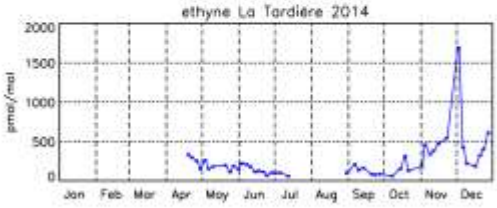
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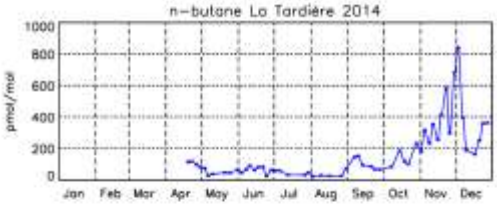
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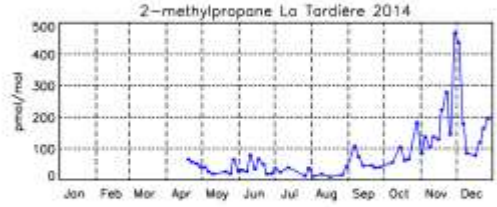
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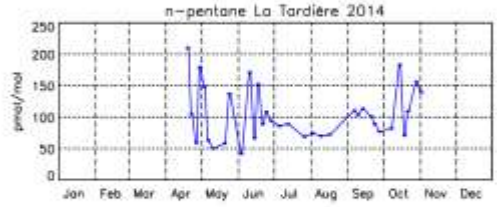
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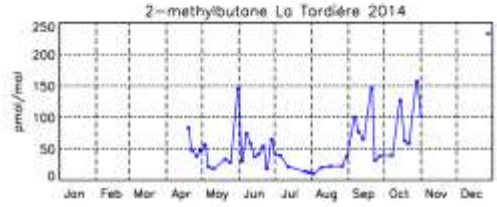
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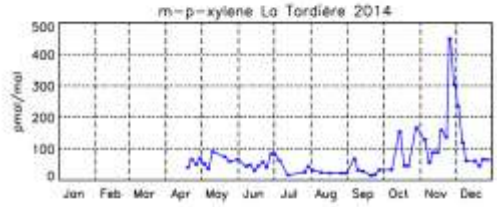
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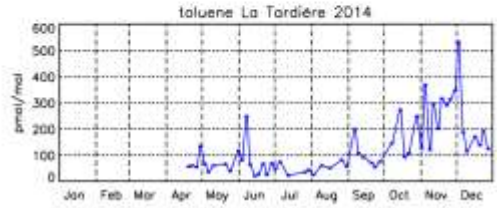
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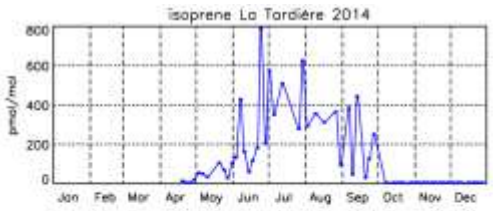
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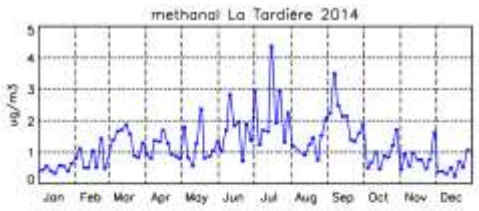
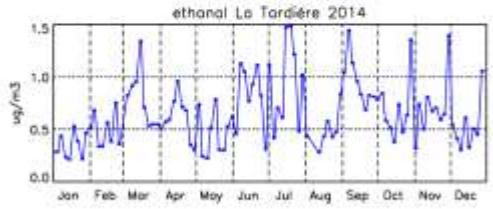
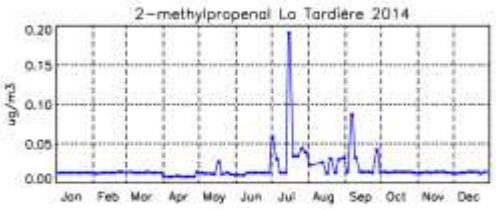

methanol La Tardière 2014
ug/m3
0
1
2
3
4
5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanal La Tardière 2014
ug/m3
0.0
0.5
1.0
1.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propanone La Tardière 2014
ug/m3
0
1
2
3
4
5
6
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propanal La Tardière 2014
ug/m3
0.0
0.2
0.4
0.6
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


butanone La Tardière 2014
ug/m3
0.0
0.5
1.0
1.5
2.0
2.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-methylpropenal La Tardière 2014
ug/m3
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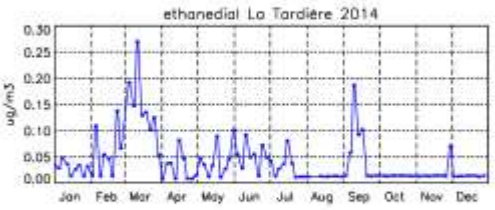
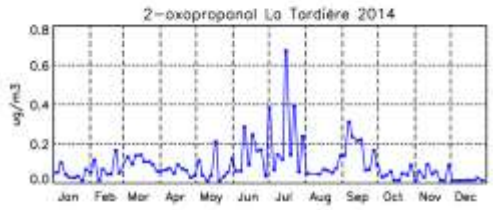
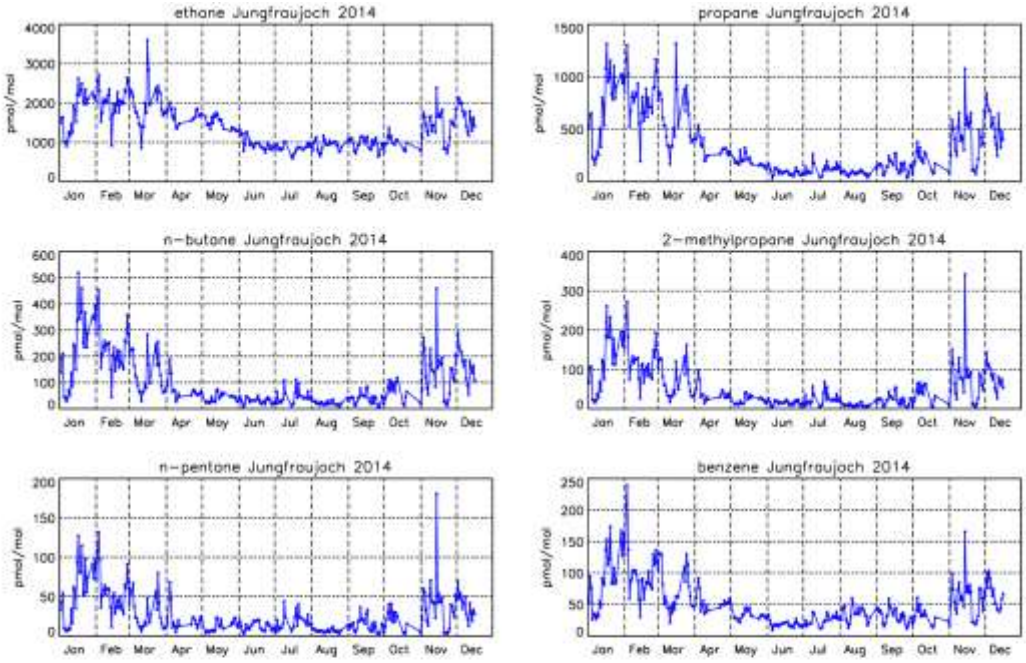
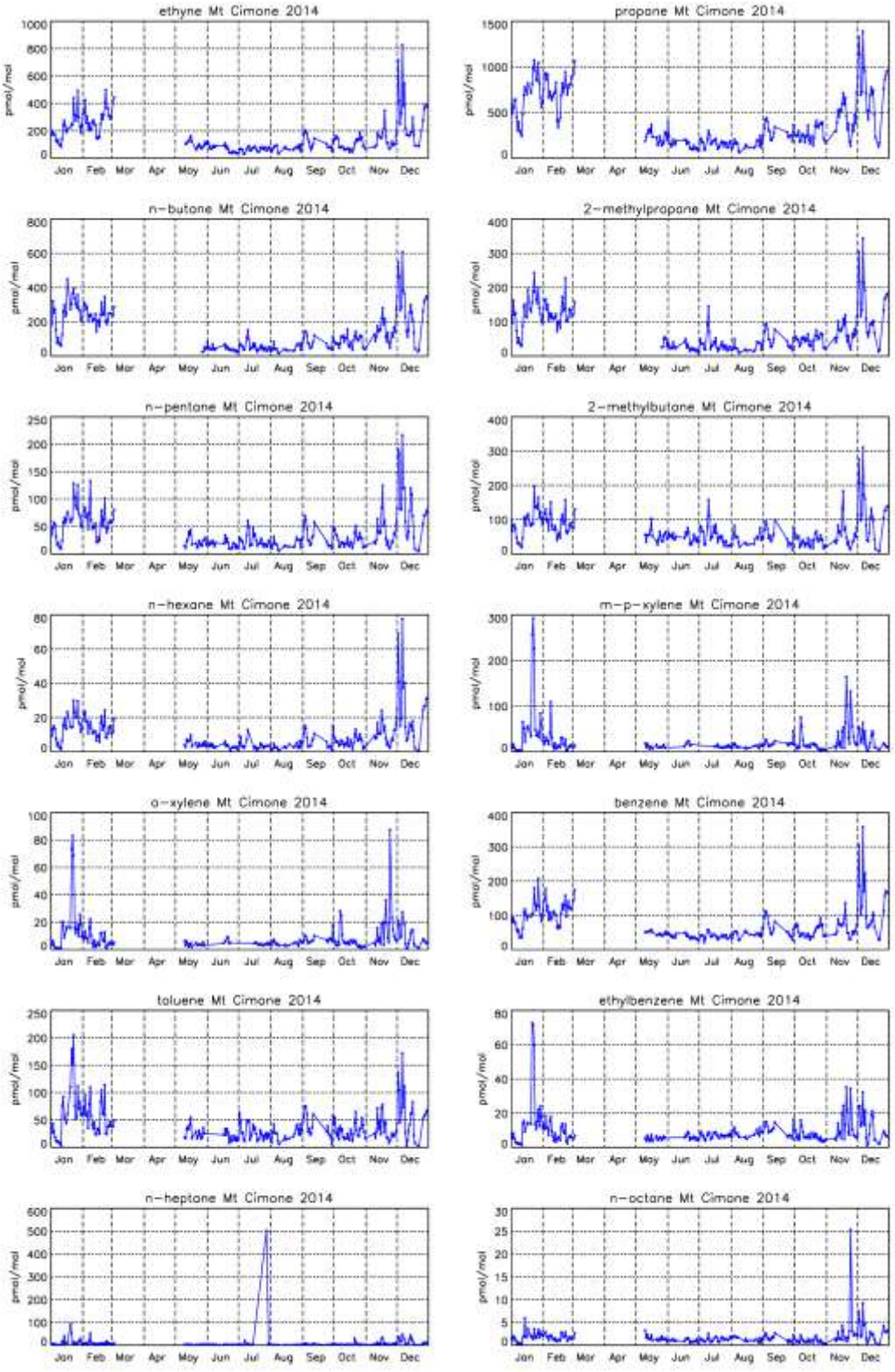
0.05
0.10
0.15
0.20
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanedial La Tardière 2014
ug/m3
0.00
0.05
0.10
0.15
0.20
0.25
0.30
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

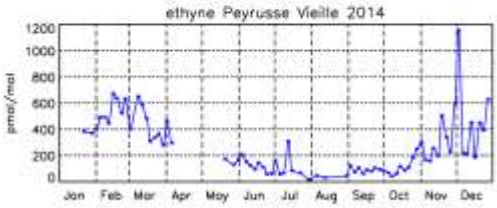

2-oxopropanal La Tardière 2014
ug/m3
0.0
0.2
0.4
0.6
0.8
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethane Jungfraujoch 2014
propane Jungfraujoch 2014
n-butane Jungfraujoch 2014
2-methylpropane Jungfraujoch 2014
n-pentane Jungfraujoch 2014
benzene Jungfraujoch 2014
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethyne Mt Cimone 2014
propane Mt Cimone 2014
n-butane Mt Cimone 2014
2-methylpropane Mt Cimone 2014
n-pentane Mt Cimone 2014
2-methylbutane Mt Cimone 2014
n-hexane Mt Cimone 2014
m-p-xylene Mt Cimone 2014
o-xylene Mt Cimone 2014
benzene Mt Cimone 2014
toluene Mt Cimone 2014
ethylbenzene Mt Cimone 2014
n-heptane Mt Cimone 2014
n-octane Mt Cimone 2014
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethane Peyrusse Vieille 2014
pmol/mol
0
1000
2000
3000
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethene Peyrusse Vieille 2014
pmol/mol
0
500
1000
1500
2000
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethyne Peyrusse Vieille 2014
pmol/mol
0
200
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600
800
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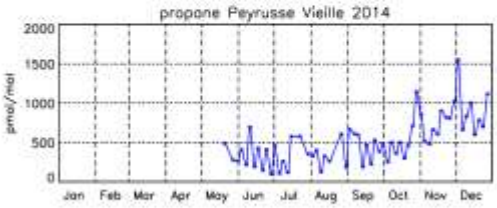
1200
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propane Peyrusse Vieille 2014
pmol/mol
0
500
1000
1500
2000
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propene Peyrusse Vieille 2014
pmol/mol
0
50
100
150
200
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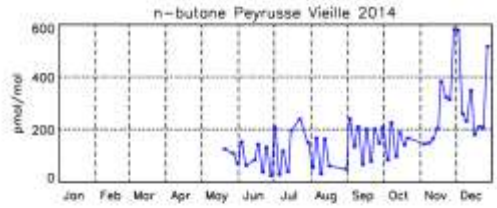
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-butane Peyrusse Vieille 2014
pmol/mol
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200
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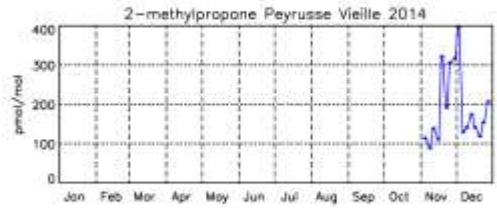
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-methylpropane Peyrusse Vieille 2014
pmol/mol
0
100
200
300
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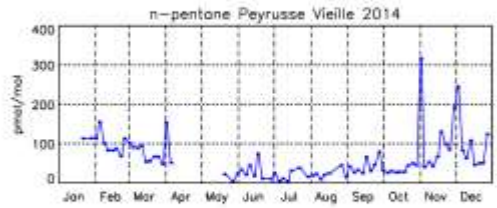
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-pentane Peyrusse Vieille 2014
pmol/mol
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200
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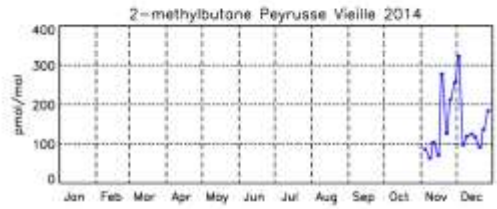
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-methylbutane Peyrusse Vieille 2014
pmol/mol
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100
200
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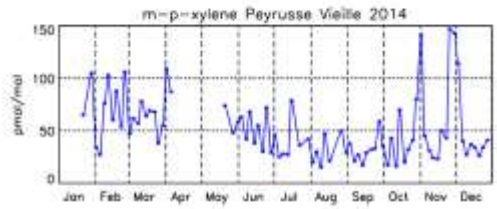
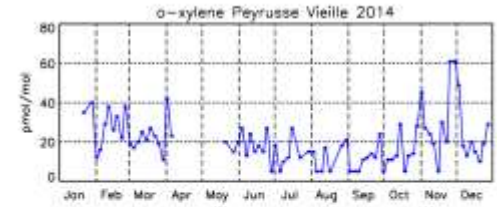
400
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


m-p-xylene Peyrusse Vieille 2014
pmol/mol
0
50
100
150
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


o-xylene Peyrusse Vieille 2014
pmol/mol
0
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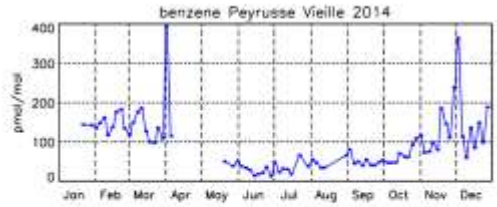
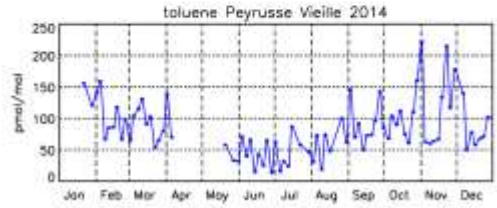
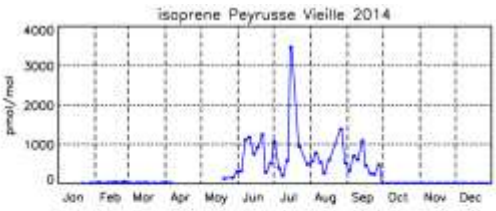
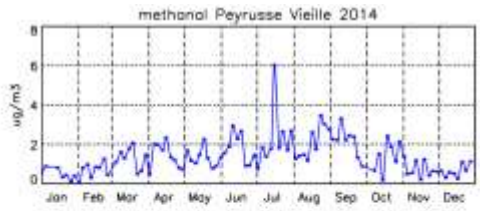
40
60
80
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


benzene Peyrusse Vieille 2014
pmol/mol
0
100
200
300
400
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


toluene Peyrusse Vieille 2014
pmol/mol
0
50
100
150
200
250
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethylbenzene Peyrusse Vieille 2014
pmol/mol
0
20
40
60
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


isoprene Peyrusse Vieille 2014
pmol/mol
4000
3000
2000
1000
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


methanol Peyrusse Vieille 2014
ug/m3
8
6
4
2
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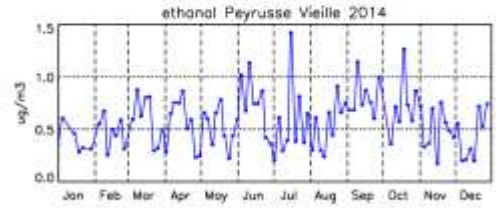
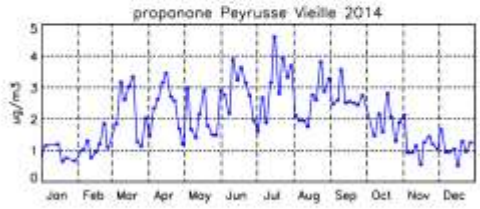
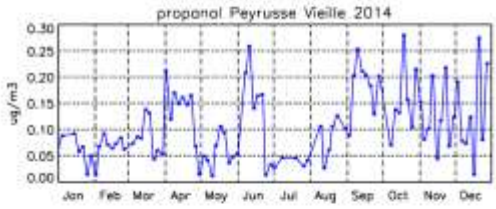
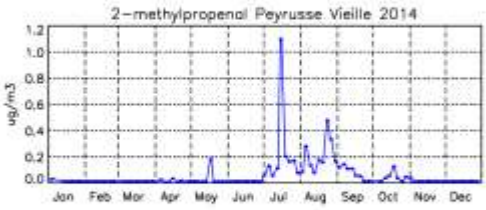
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

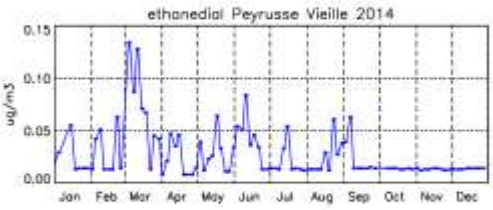

ethanol Peyrusse Vieille 2014
ug/m3
1.5
1.0
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

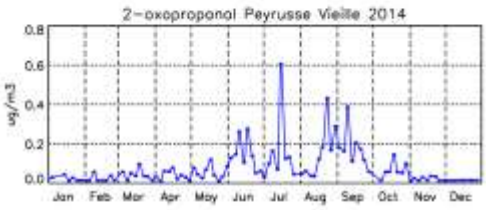

propanone Peyrusse Vieille 2014
ug/m3
5
4
3
2
1
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propanol Peyrusse Vieille 2014
ug/m3
0.30
0.25
0.20
0.15
0.10
0.05
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


butanone Peyrusse Vieille 2014
ug/m3
6
4
2
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-methylpropenal Peyrusse Vieille 2014
ug/m3
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

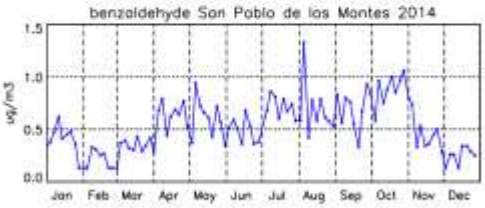

ethanedial Peyrusse Vieille 2014
ug/m3
0.15
0.10
0.05
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-oxopropanal Peyrusse Vieille 2014
ug/m3
0.8
0.6
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


methanol San Pablo de los Montes 2014
ug/m3
0
10
20
30
40
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

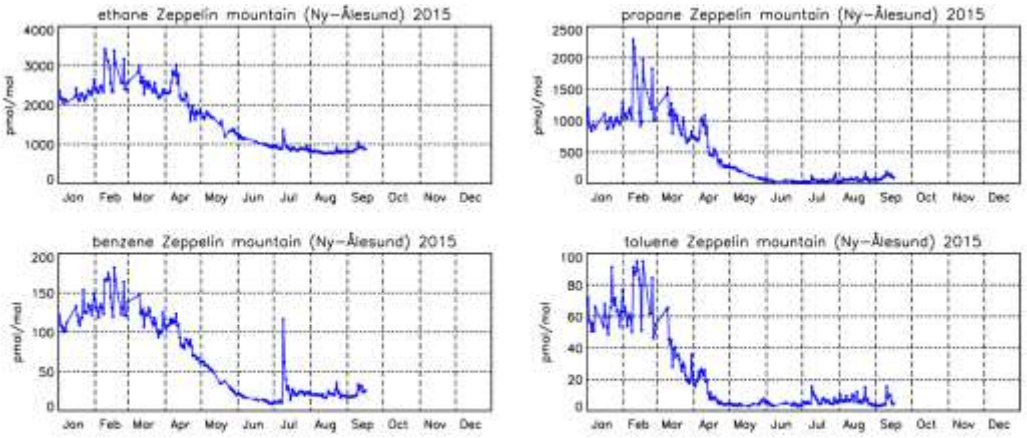

ethanol San Pablo de los Montes 2014
ug/m3
0
1
2
3
4
5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

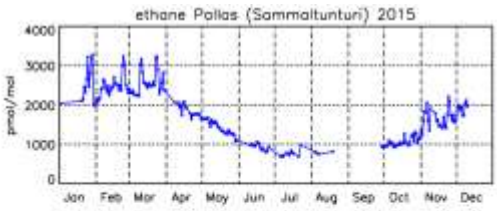

butanone San Pablo de los Montes 2014
ug/m3
0.0
0.5
1.0
1.5
2.0
2.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

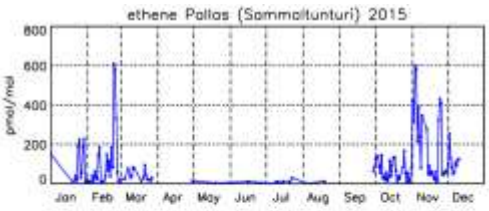

benzaldehyde San Pablo de los Montes 2014
ug/m3
0.0
0.5
1.0
1.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

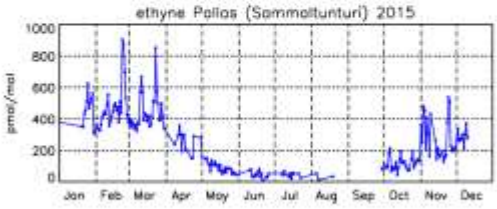

pentanol San Pablo de los Montes 2014
ug/m3
0.0
0.5
1.0
1.5
2.0
2.5
3.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

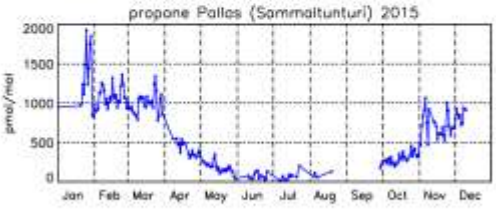

hexanal San Pablo de los Montes 2014
ug/m3
0.0
0.5
1.0
1.5
2.0
2.5
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethane Zeppelin mountain (Ny-Ålesund) 2015
propane Zeppelin mountain (Ny-Ålesund) 2015
benzene Zeppelin mountain (Ny-Ålesund) 2015
toluene Zeppelin mountain (Ny-Ålesund) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

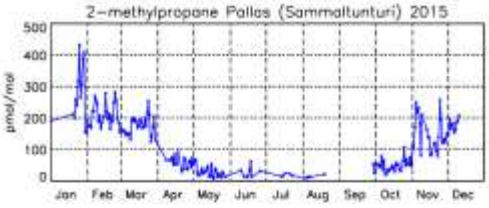

ethane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

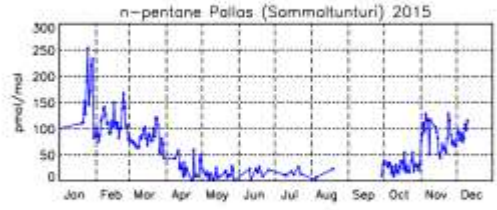

ethene Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

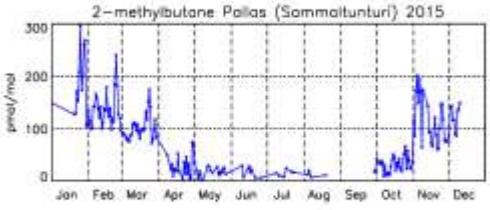

ethyne Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

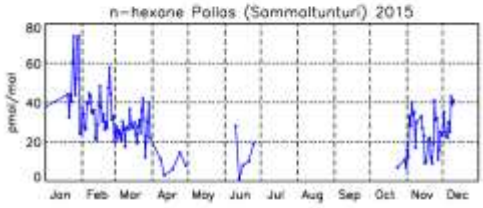

propane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-butane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

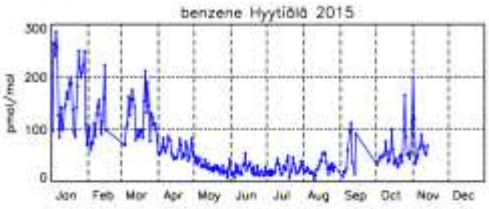

2-methylpropane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-pentane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-methylbutane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-hexane Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


benzene Pallas (Sammaltunturi) 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


benzene Hyytiälä 2015
pmol/mol
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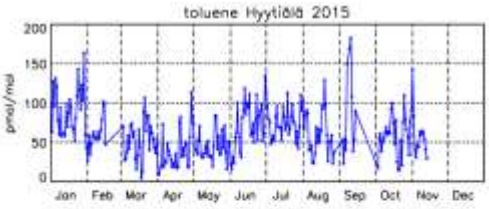
100
200
300
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


toluene Hyytiälä 2015
pmol/mol
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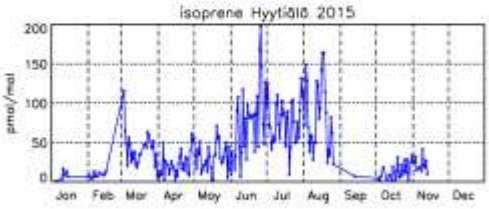
100
150
200
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


Isoprene Hyytiälä 2015
pmol/mol
0
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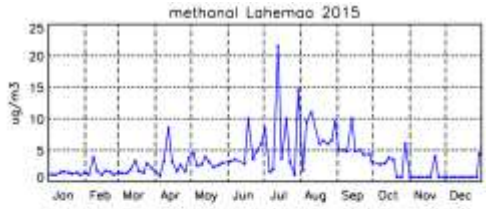
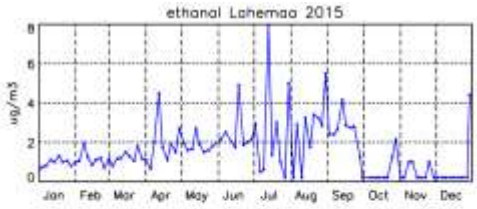
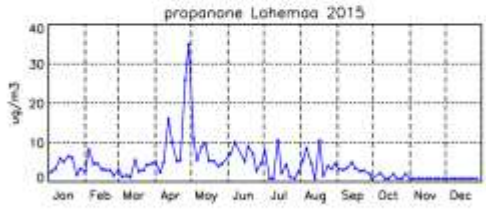
100
150
200
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


methanol Lahemaa 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanol Lahemaa 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec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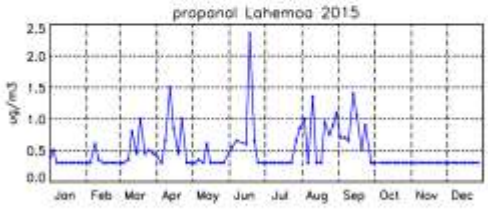
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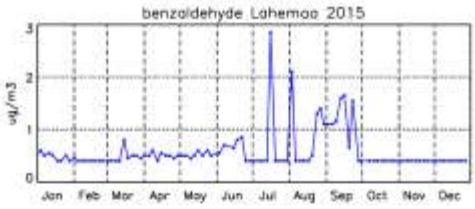
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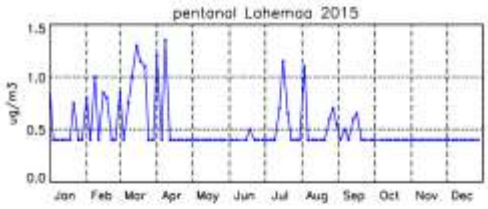
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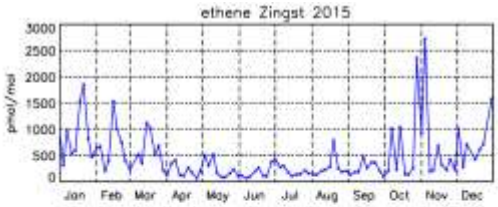
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propane Zingst 2015
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n-butane Zingst 2015
pmol/mol
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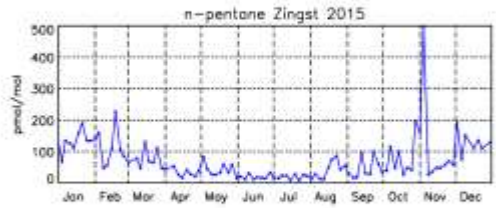
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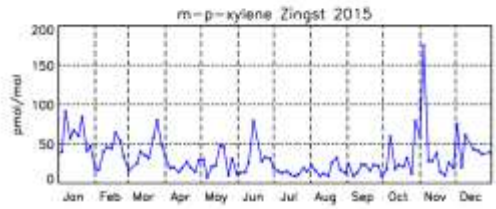
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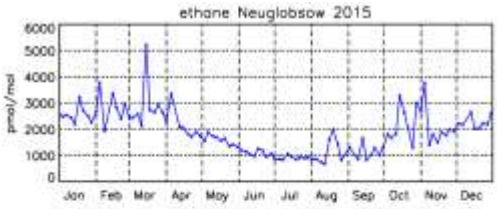
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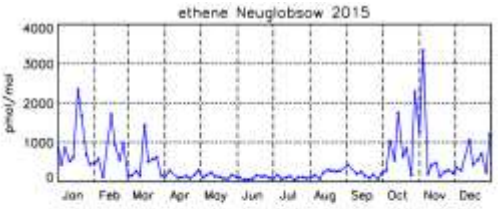
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propane Neuglobsow 2015
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n-butane Neuglobsow 2015
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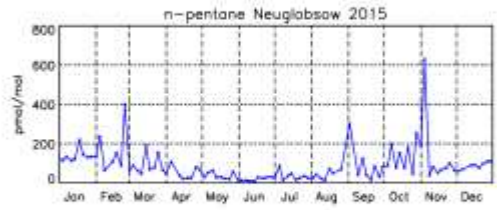
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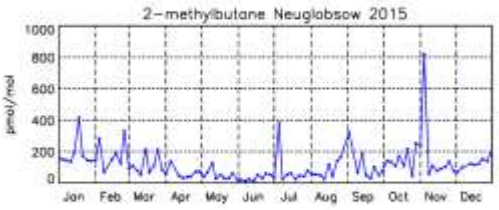
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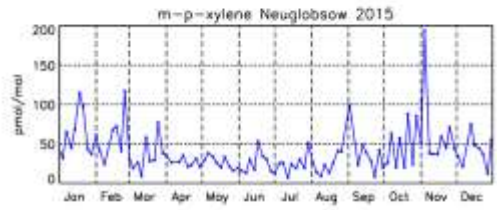
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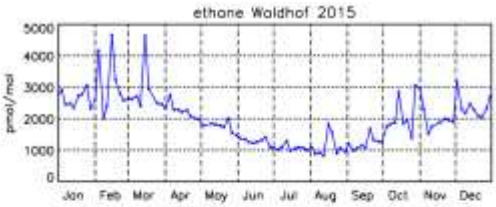
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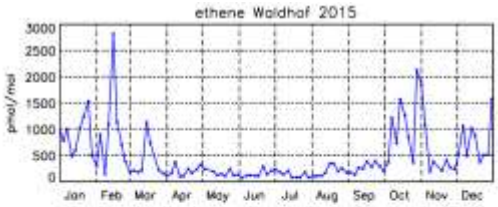
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propane Waldhof 2015
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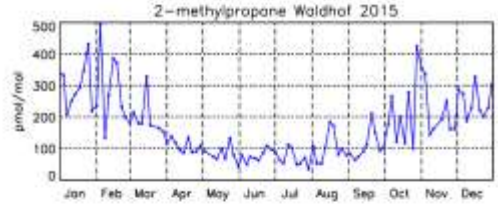
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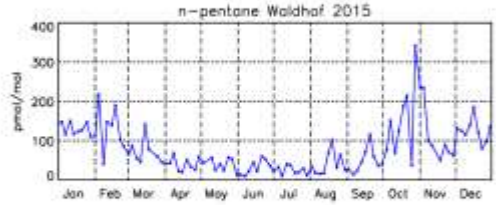
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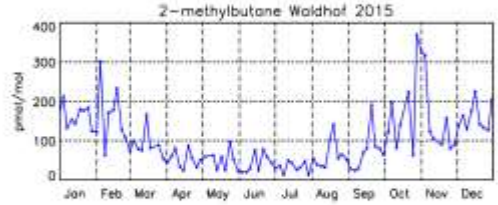
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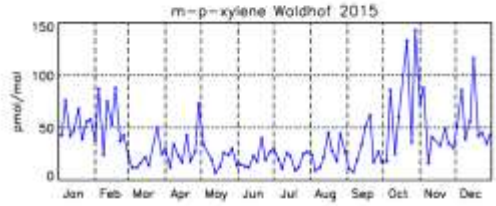
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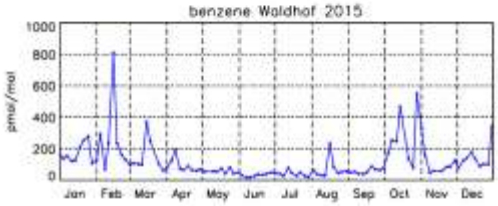
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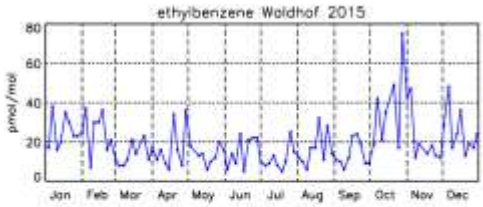
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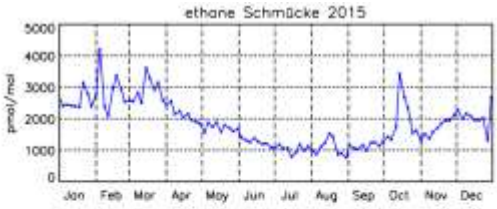
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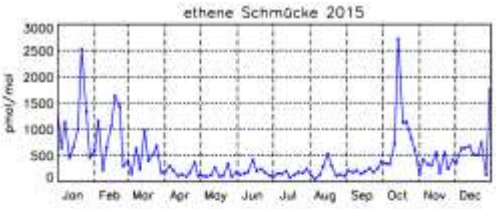
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propane Schmücke 2015
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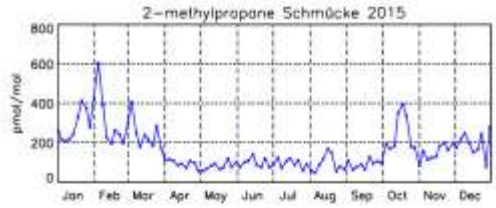
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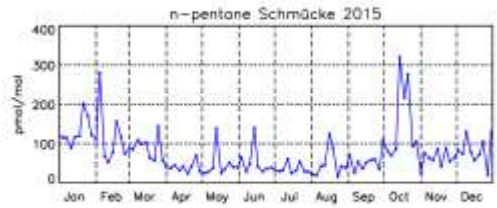
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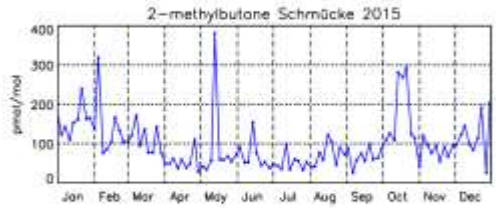
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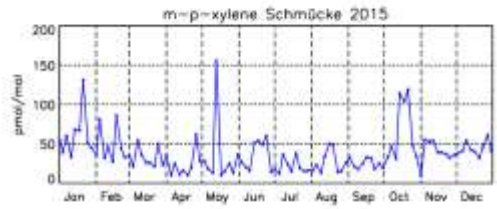
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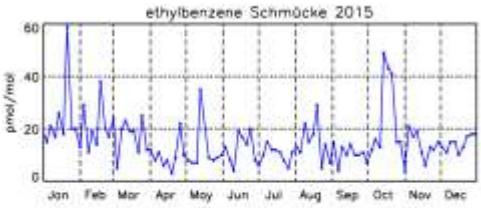
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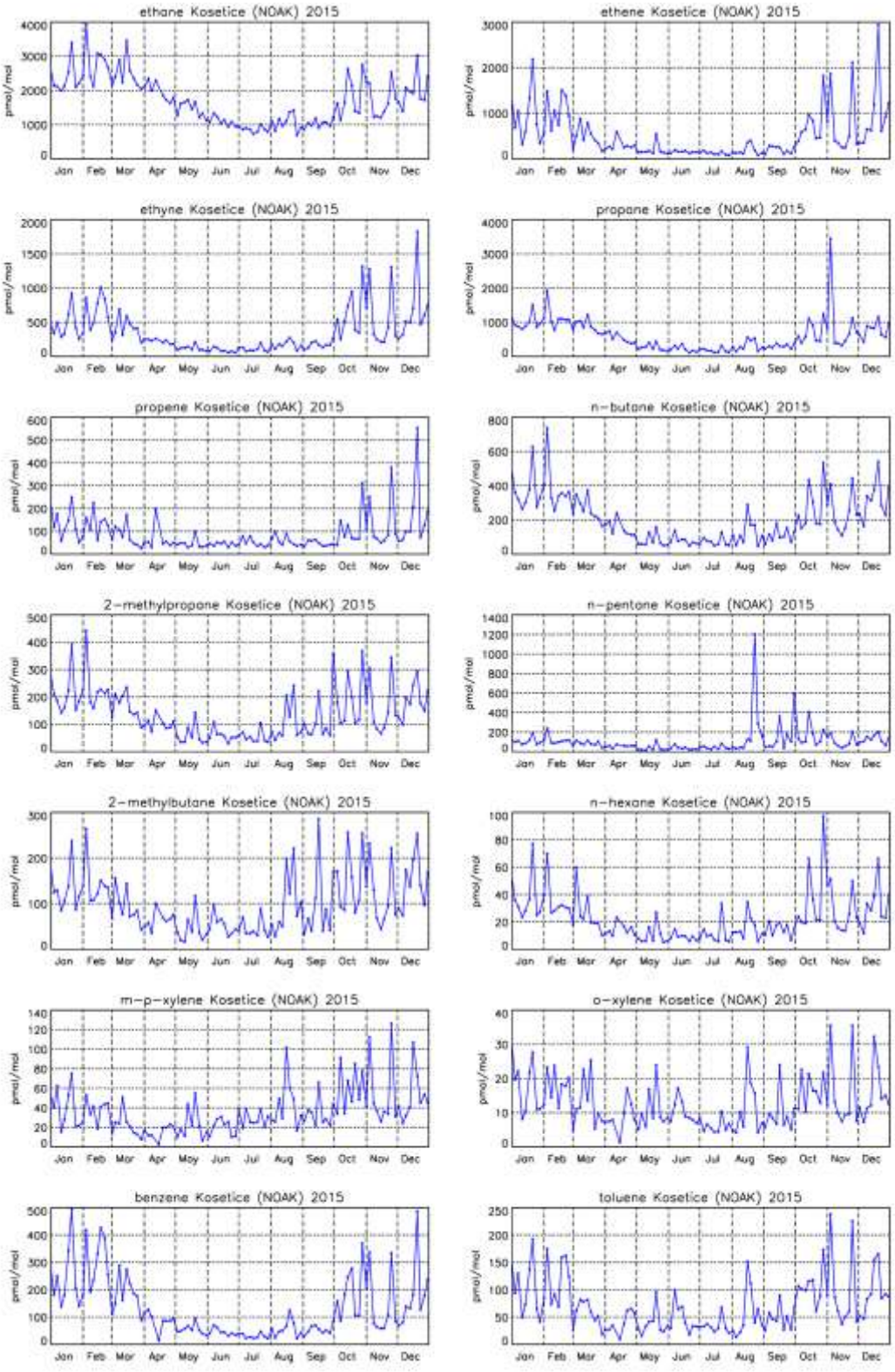
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propane Kosetice (NOAK) 2015
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2-methylpropane Kosetice (NOAK) 2015
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2-methylbutane Kosetice (NOAK) 2015
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m-p-xylene Kosetice (NOAK) 2015
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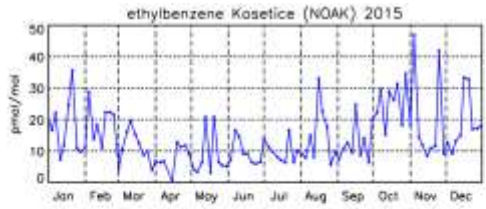
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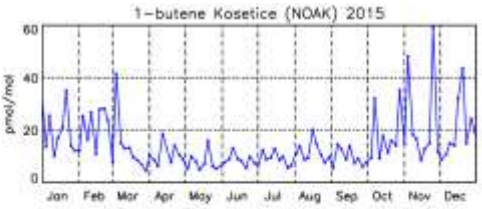
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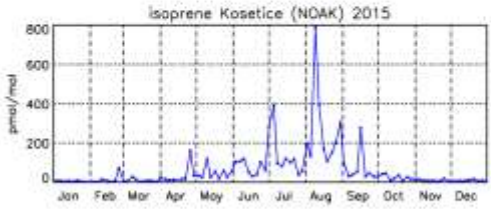
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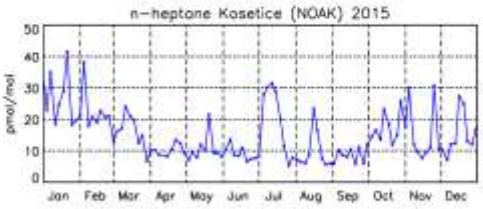
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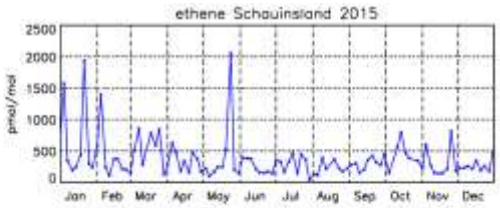
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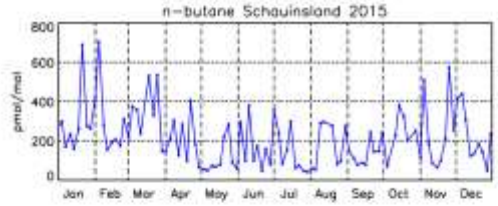
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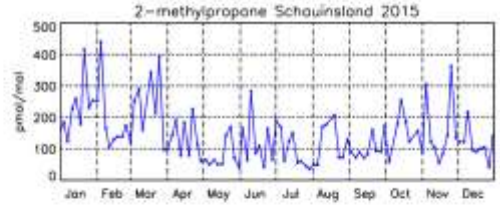
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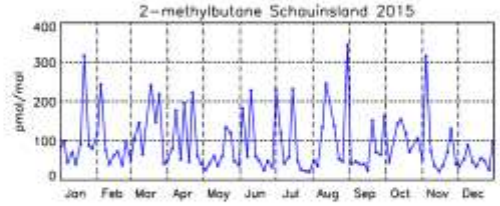
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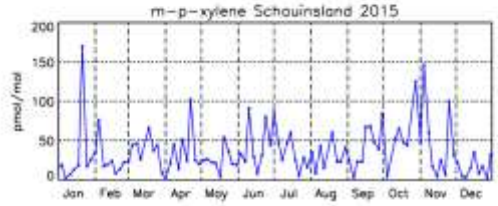
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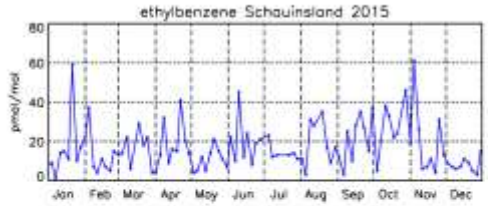
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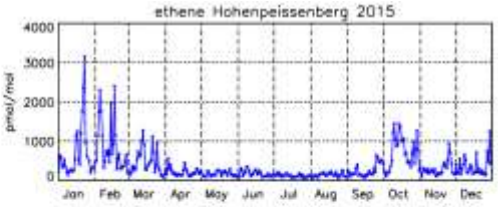
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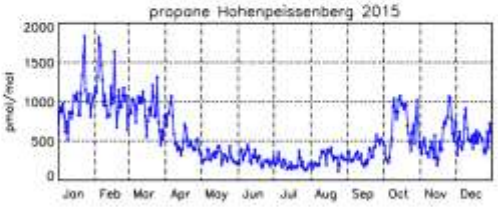
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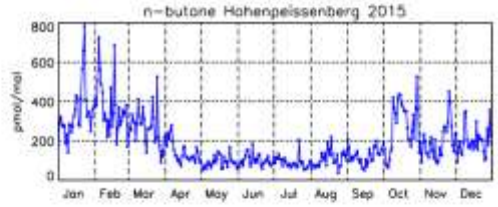
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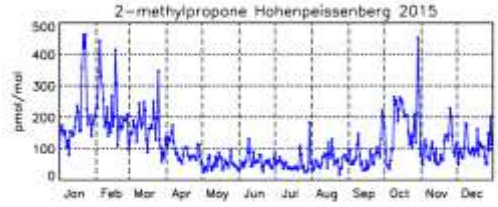
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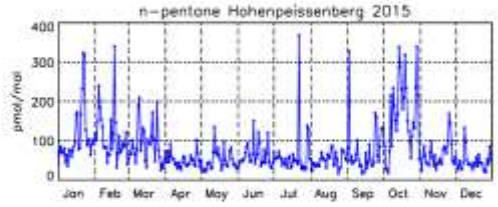
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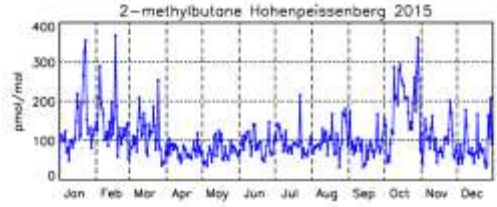
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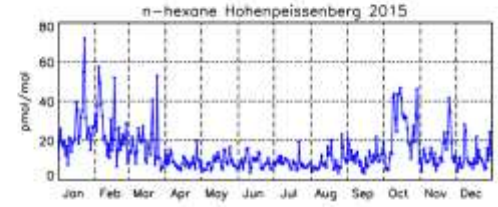
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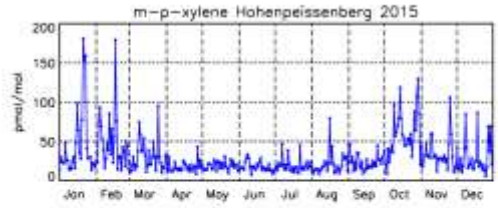
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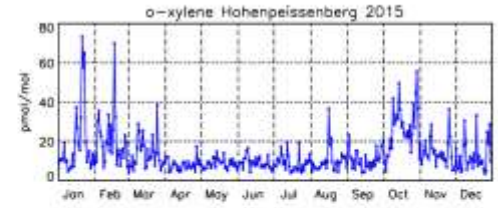
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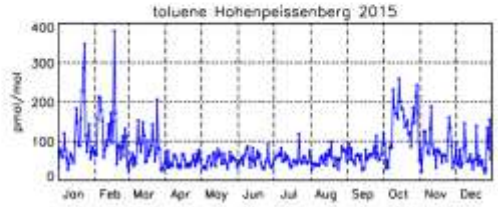
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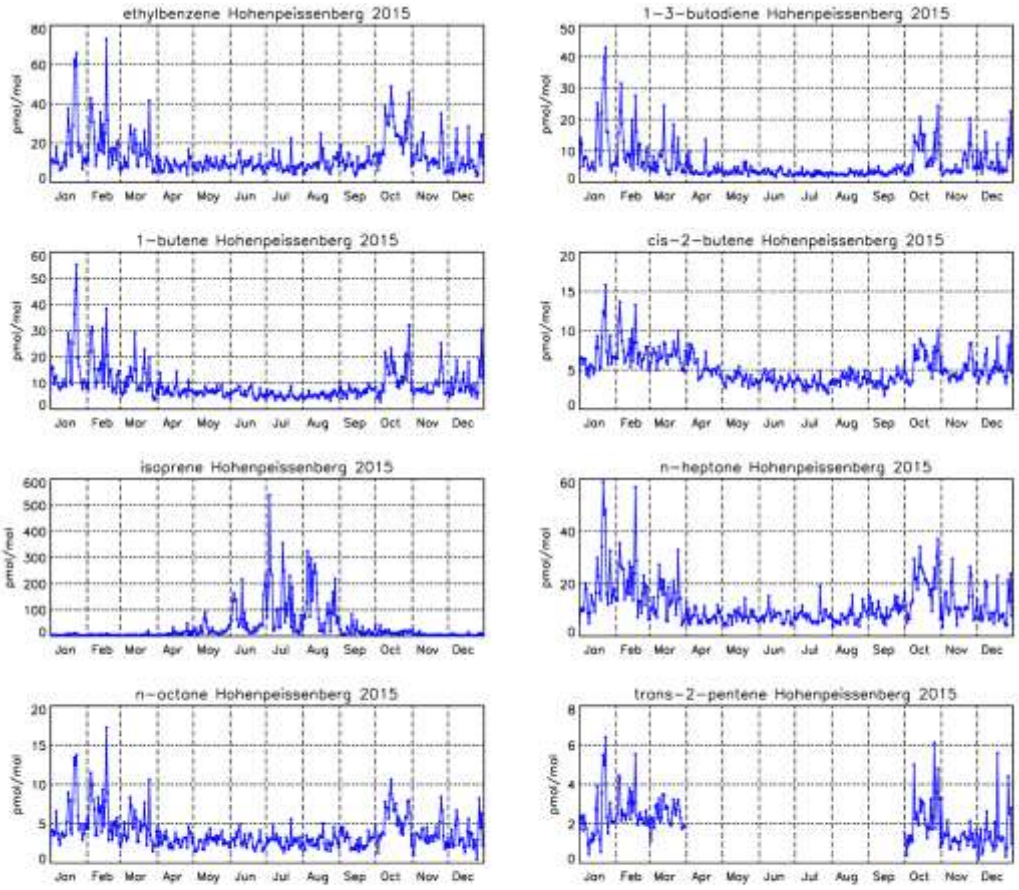
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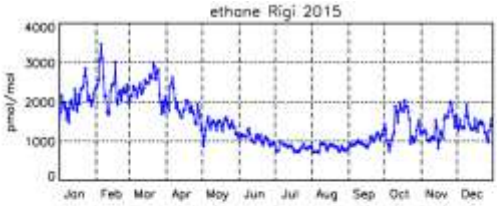
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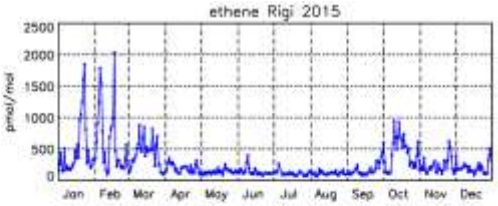
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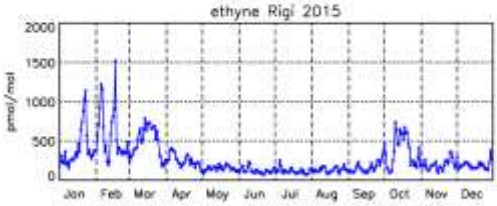
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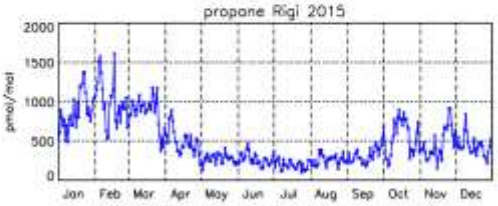
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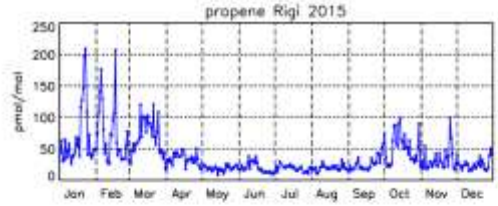
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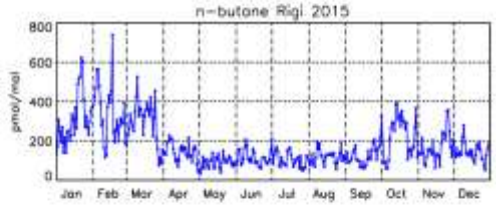
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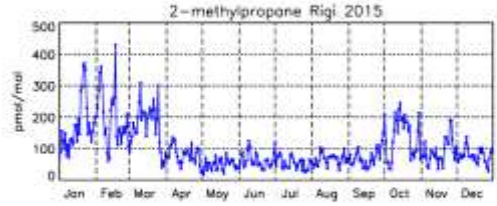
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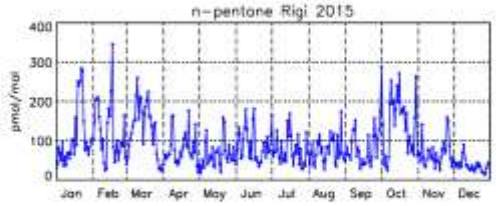
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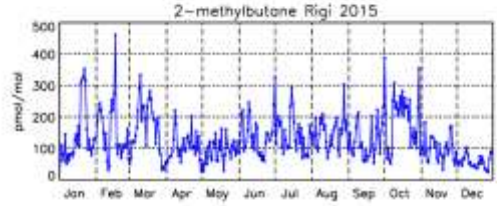
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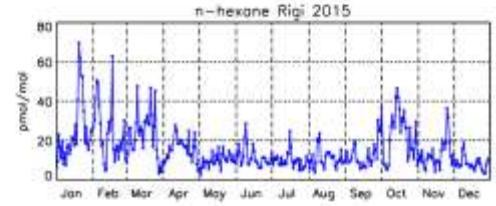
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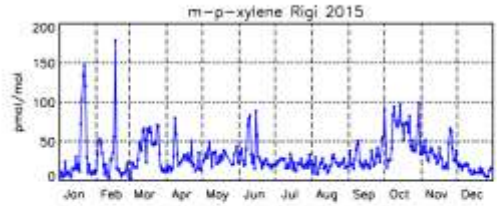
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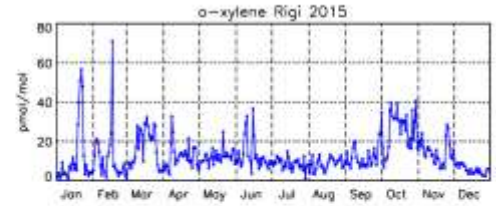
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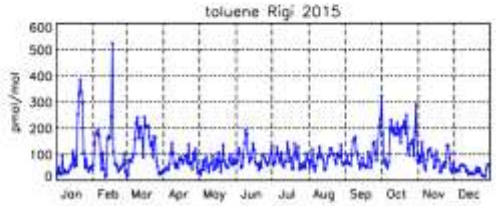
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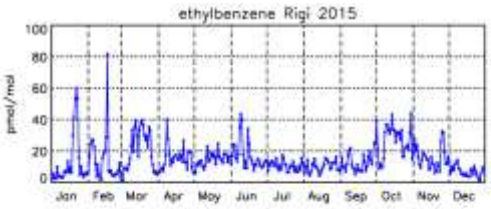
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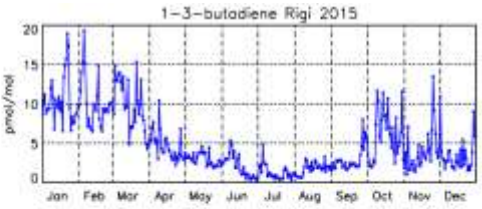
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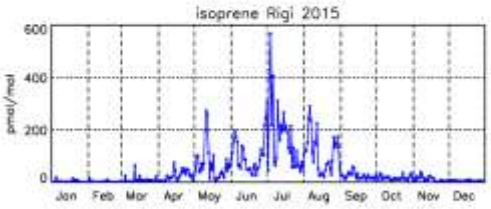
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ethane La Tardière 2015
pmol/mol
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ethene La Tardière 2015
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ethyne La Tardière 2015
pmol/mol
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pmol/mol
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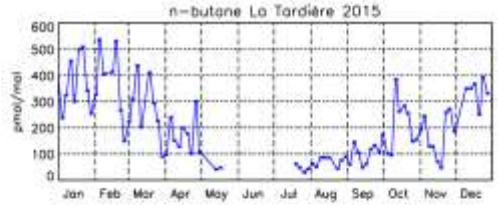
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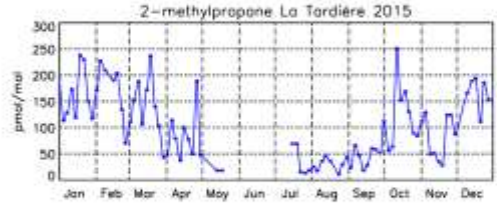
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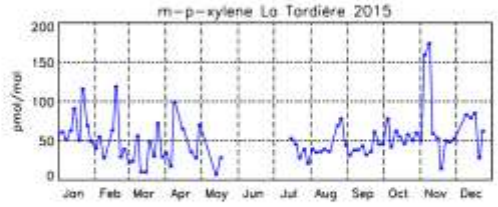
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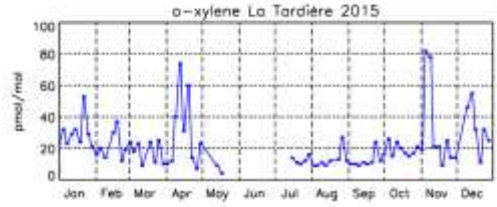
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benzene La Tardière 2015
pmol/mol
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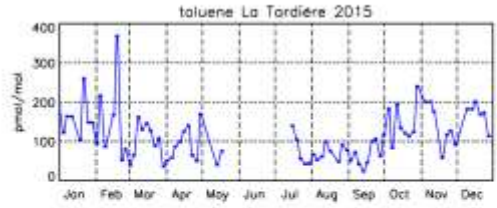
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ethylbenzene La Tardière 2015
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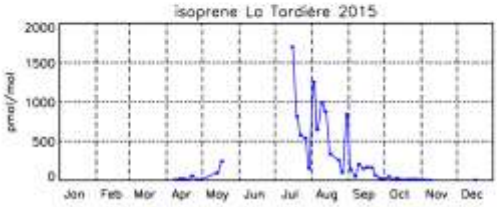
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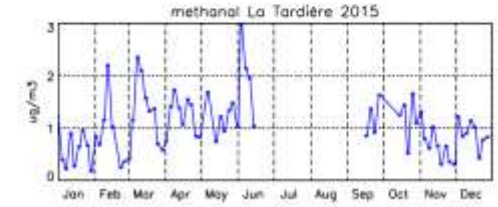
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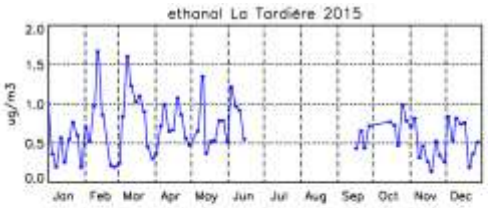
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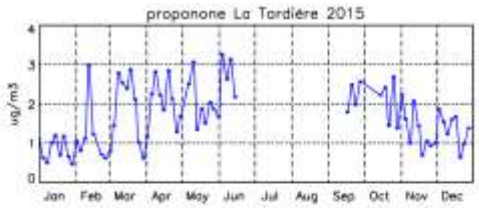
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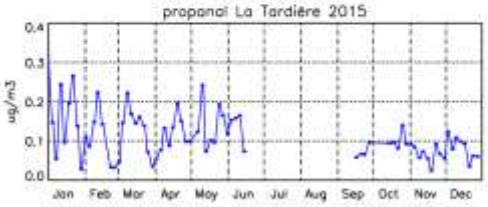
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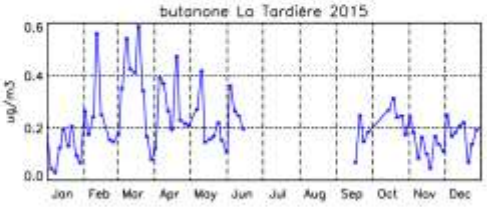
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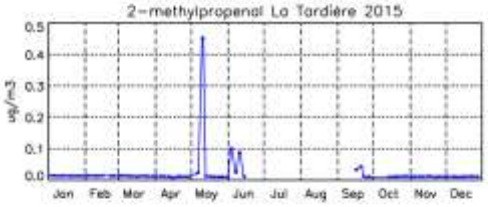
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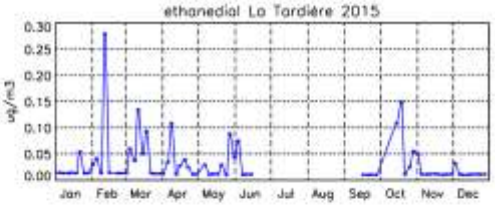
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2-oxopropanol La Tardière 2015
ug/m3
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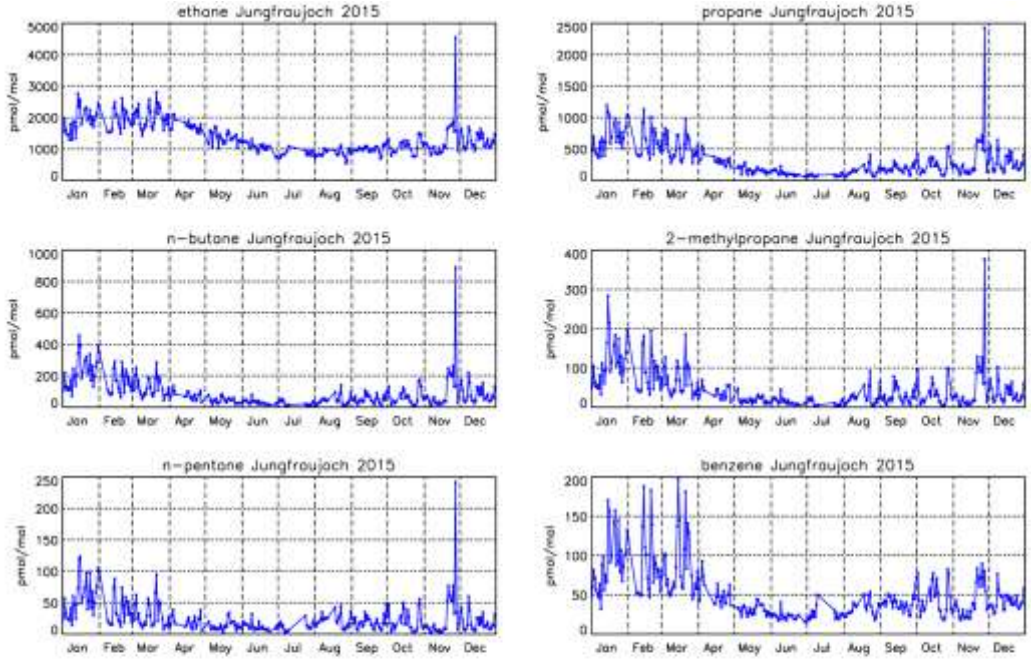
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pmol/mol
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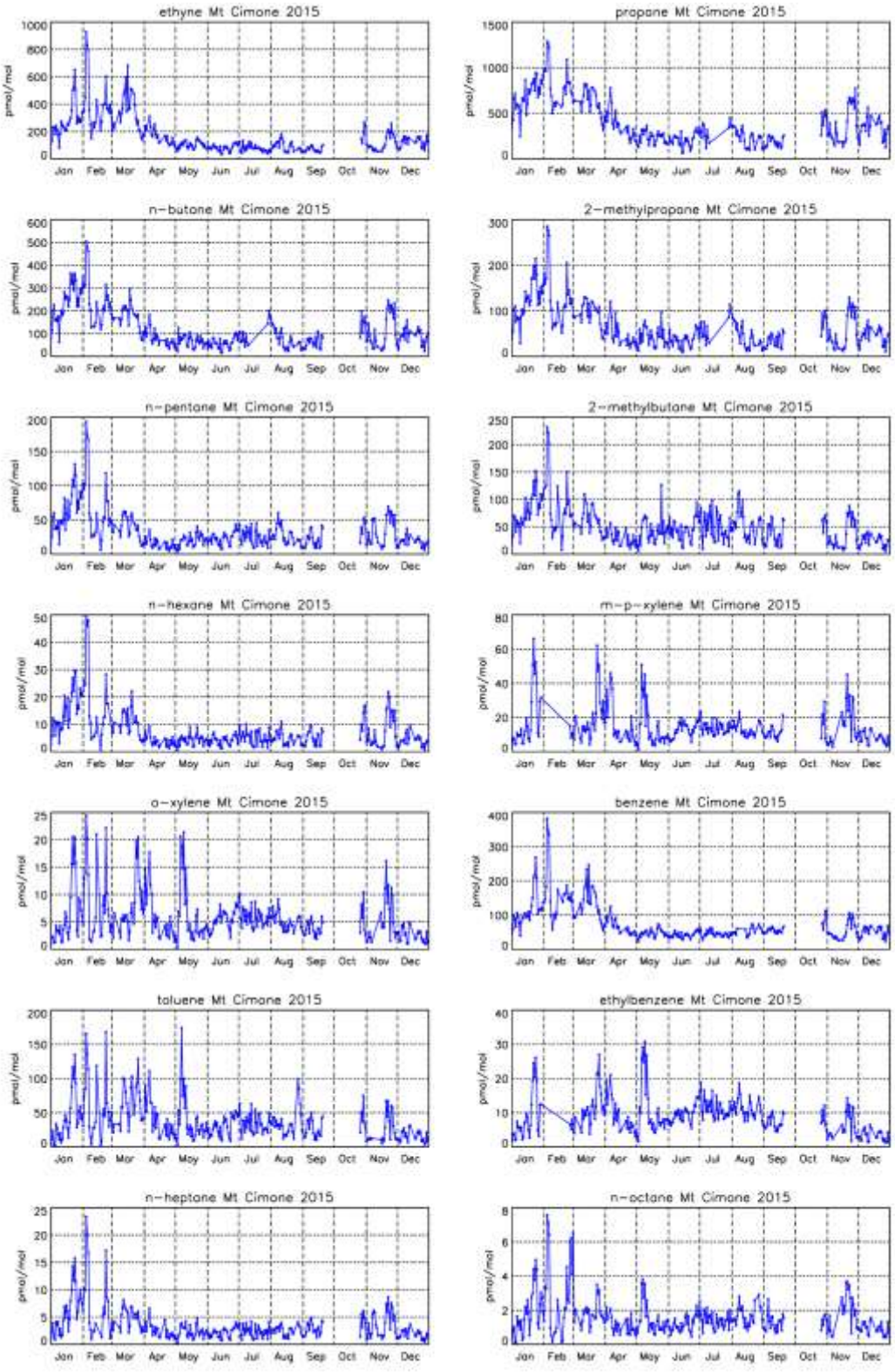
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o−xylene Mt Cimone 2015
benzene Mt Cimone 2015
toluene Mt Cimone 2015
ethylbenzene Mt Cimone 2015
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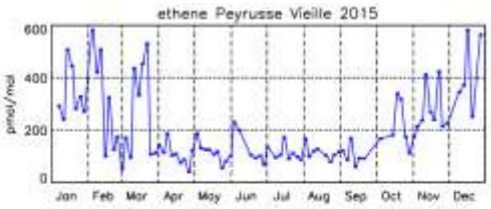
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pmol/mol
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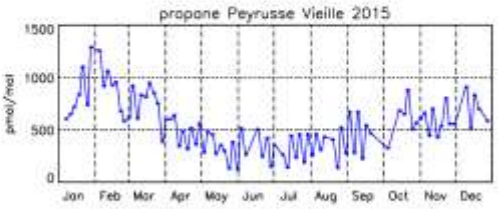
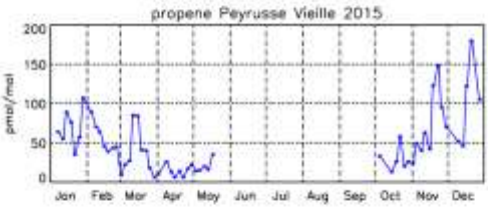
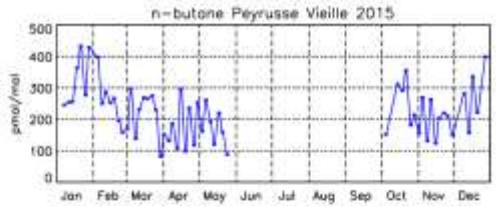
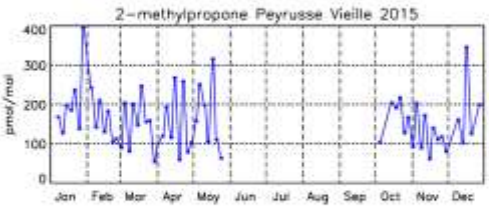
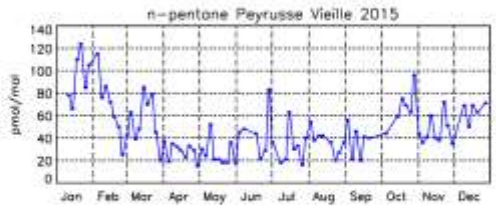
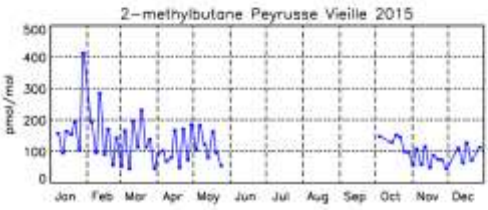

propane Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

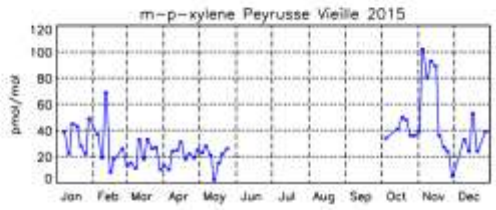

propene Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


n-butane Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

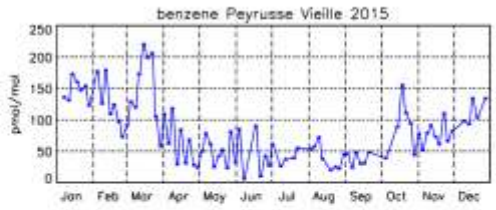

2-methylpropane Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

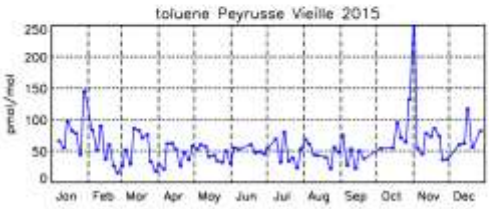

n-pentane Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

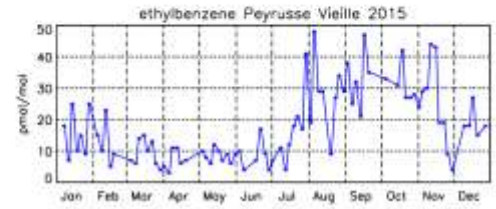

2-methylbutane Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

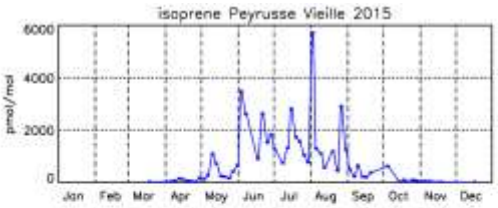

m-p-xylene Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

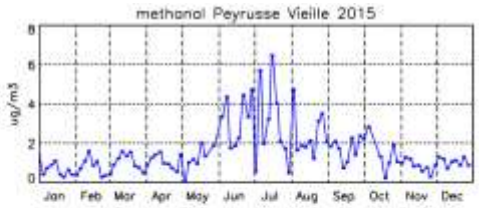

o-xylene Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

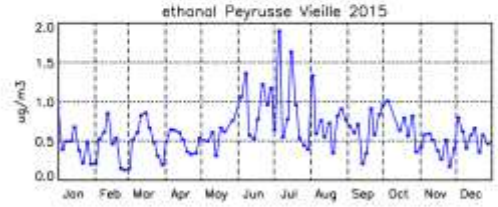

benzene Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

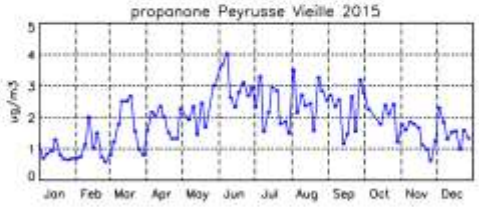

toluene Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

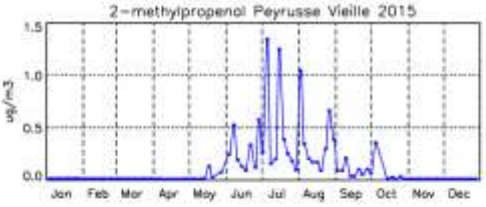

ethylbenzene Peyrusse Vieille 2015
pmol/mol
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

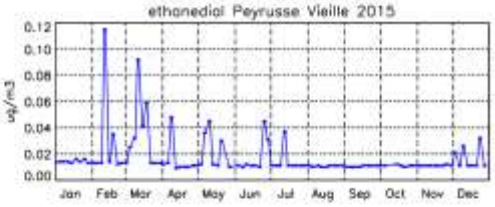

isoprene Peyrusse Vieille 2015
pmol/mol
6000
4000
2000
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

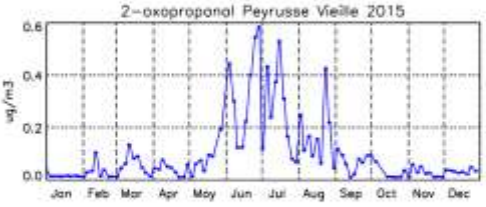

methanol Peyrusse Vieille 2015
ug/m3
8
6
4
2
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanol Peyrusse Vieille 2015
ug/m3
2.0
1.5
1.0
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


propanone Peyrusse Vieille 2015
ug/m3
5
4
3
2
1
0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-methylpropenol Peyrusse Vieille 2015
ug/m3
1.5
1.0
0.5
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

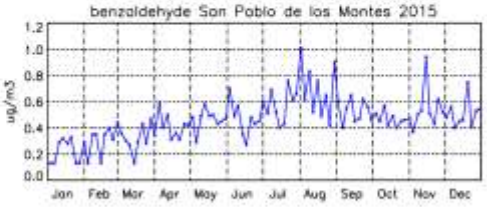

ethanedial Peyrusse Vieille 2015
ug/m3
0.12
0.10
0.08
0.06
0.04
0.02
0.00
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


2-oxopropanol Peyrusse Vieille 2015
ug/m3
0.6
0.4
0.2
0.0
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

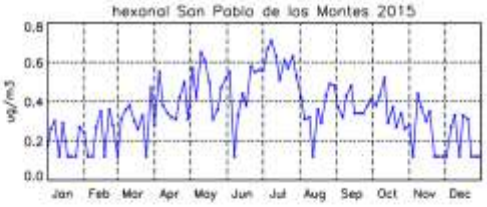

methanol San Pablo de los Montes 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


ethanol San Pablo de los Montes 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


butanone San Pablo de los Montes 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


benzaldehyde San Pablo de los Montes 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


pentanal San Pablo de los Montes 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec


hexanal San Pablo de los Montes 2015
ug/m3
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec