

Operation of EMEP 'supersites' in the United Kingdom

Annual Report for 2008

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Ecology & Hydrology
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This report was written by J N Cape, CEH Edinburgh (jnc@ceh.ac.uk)
with contributions from:

W Bealey
M Coyle
C DiMarco
J Kentisbeer
D Leaver
H Malcolm
E Nemitz
I Simmons
M Twigg
N van Dijk

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Executive Summary

As part of its commitment to the UN-ECE Convention on Long-range Transboundary Air Pollution the United Kingdom operates two 'supersites' reporting data to the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP).

This report provides the annual summary for 2008, the second full calendar year of operation of the first EMEP 'supersite' to be established in the United Kingdom. Detailed operational reports have been submitted to Defra every 3 months, with unratified data. This annual report contains a summary of the ratified data for 2008.

The EMEP 'supersite' is located in central southern Scotland at Auchencorth (3.2°W, 55.8°N), a remote rural moorland site ~20 km south-west of Edinburgh. Monitoring operations started formally on 1 June 2006.

In addition to measurements made specifically under this contract, the Centre for Ecology & Hydrology also acts as local site operator for measurements made under other UK monitoring networks: the Automated Urban and Rural Network (AURN), the UK Eutrophication and Acidification Network (UKEAP), the UK Hydrocarbons Network, and the UK Heavy Metals Rural Network. Some measurements were also made under the auspices of the 'Air Pollution Deposition Processes' contract. All these associated networks are funded by Defra.

This report summarises the measurements made between January and December 2008, and presents summary statistics on average concentrations.

The site is dominated by winds from the south-west, but wind direction data highlight potential sources of airborne pollutants (power stations, conurbations).

The average diurnal patterns of gases and particles are consistent with those expected for a remote rural site.

The frequency distributions are presented for data where there was good data capture throughout the whole period. Some components (e.g. black carbon) show log-normal frequency distributions, while other components (e.g. ozone) have more nearly normal frequency distributions.

A case study is presented for a period in June 2008, showing the influence of regional air pollutants at this remote rural site.

All the data reported under the contract are shown graphically in the Appendix.

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1. Overview

This project covers the operation of one of two 'supersites' in the UK for monitoring air pollutants. The project is part of the UK's contribution to a Europe-wide programme of air monitoring under the UN Economic Commission for Europe's Convention on Long Range Transboundary Air Pollution. The monitoring is done under the auspices of EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe), and contributes data to a central European database. The results are used to evaluate the levels of air pollution across Europe, to identify trends, and for comparison with computer model outputs.

The measurements under this project were started in June 2006 at Auchencorth Moss, approximately 20 km south of Edinburgh, in east central Scotland. The site is remote from sources of air pollutants, and provides a large flat area of uniform vegetation (moss, grass and heather) which is ideally suited to long-term monitoring. The site is classified as remote rural.

Measurements included a range of trace gases (ozone, nitrogen oxides, sulphur dioxide, nitric acid, ammonia, volatile organic compounds), and particles in two size fractions: diameters up to 2.5 micrometre (PM_{2.5}) and up to 10 micrometre (PM₁₀). Particle mass in the two size fractions was measured, as well as water-soluble particulate material (salts) and black carbon (soot). Occasional 'intensive' monitoring activity, coordinated at several sites across Europe to provide a more detailed picture of air quality for two months out of every year, is reported separately.

Data collected under this project are archived and made available via the internet through a central database.

2. Background

2.1 Policy context

The UNECE Convention on Long Range Transboundary Air Pollutants operates a number of measurement and monitoring programmes, including EMEP – the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (see http://www.emep.int/index_facts.html).

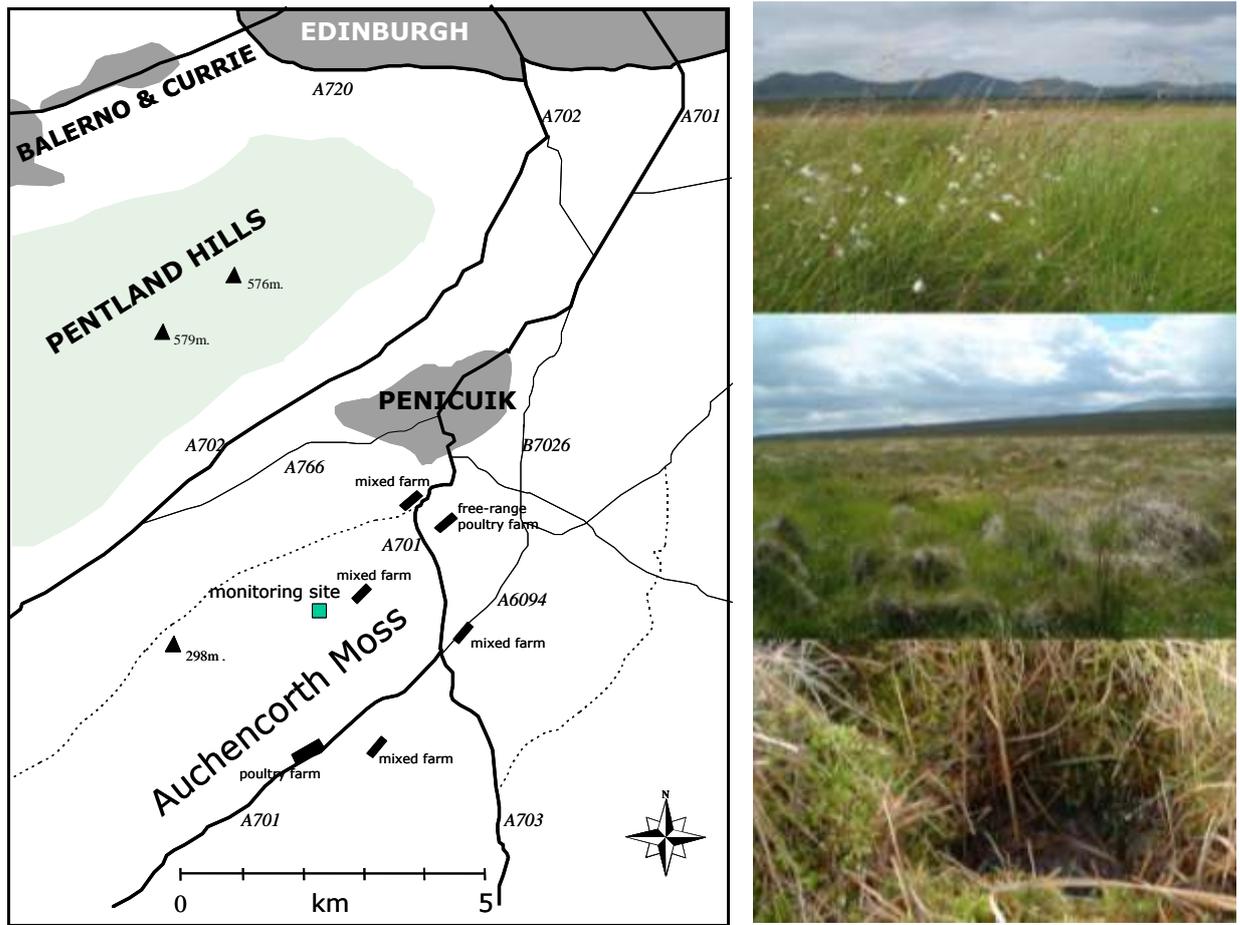
The Norwegian Institute for Air Research (NILU) arranged an EMEP Task Force on Measurements and Modelling (TFMM) workshop on the implementation of the EMEP monitoring strategy (EB.AIR/GE.1/2004/5) in Oslo on 22-24 November 2004 (see also <http://www.nilu.no/projects/ccc/reports/cccr9-2003.pdf>). The objective of the workshop was to discuss methodologies and technical requirements needed to implement the level 2 and level 3 activities defined by the EMEP monitoring strategy. Subsequently a meeting was held in Zagreb, Croatia in April 2005 (<http://www.nilu.no/projects/ccc/tfmm/index.html>) which also considered implementation of the EMEP monitoring strategy. Details of the EMEP monitoring strategy and measurement programme for 2004-9 are available at http://www.unece.org/env/emep/Monitoring%20Strategy_full.pdf.

The United Kingdom contribution to the EMEP monitoring strategy is based on the creation of two Level 2 ‘supersites’, one in the north of the UK and one in the south, at which additional measurements are, or can be, made to qualify as an EMEP Level 3 site. Data from these sites at hourly or daily frequency will be supplemented by long-term integrating measurements from existing Defra-funded monitoring networks across the UK (<http://www.nilu.no/projects/ccc/network/index.html>). The northern site is identified as Auchencorth Moss in eastern Scotland (UK OS grid reference NT220562; http://www.heavymetals.ceh.ac.uk/sites/site_auc.htm) which has been used by the Centre for Ecology & Hydrology for several years as an intensive monitoring site for trace gas and particle concentrations and fluxes. The site is based in an area of upland peat, with heather and grass cover, and has an extensive fetch to the south-west. The southern site is at Harwell, in Oxfordshire (OS grid reference SU468860), operated by Netcen over many years for sampling trace gases and aerosols.

The Auchencorth site was inaugurated as a ‘supersite’ in June 2006, although it had been used by CEH for 10 years previously for gas concentration and flux measurements. It has been operating continuously since then for continuous monitoring activities and the ‘intensive’ periods of more detailed measurements which are coordinated by EMEP across Europe for two months each year. The first ‘intensive’ period was in June 2006, with the second ‘intensive’ period in January/February 2007 and the third in September/October 2008. The suite of measurements includes data which are currently being collected as part of existing Defra UK monitoring networks. However, all data from the site are recorded and stored on a single database at CEH Edinburgh, so that the measurements from the different UK networks operating at the site can be compared.

2.2 Site details

The Auchencorth site is located ~20 km south-west of Edinburgh on open moorland at 255 m asl, 3.2 °W, 55.8°N (OS grid reference NT220562). There are no major sources of pollution nearby, although there are some large towns to the north-east and intensive farming to the south and south-east, as indicated on the maps in Figure 1a. There is an extensive uniform fetch of blanket bog to the south, west and north (Figure 1b) comprising mixed grass species, heather and substantial areas of moss species *Sphagnum spp.* and *Polytrichum spp.* (Figure 1c).



a.
b.

c.



Figure 1: a. Map showing the location of Auchencorth Moss, b view of the fetch from the SW to NW (panorama generated from several individual photos hence some discontinuities and distortion), c. the vegetation during summer months.

2.3 Outline of work at Auchencorth

The measurements made during 2008 are listed below. A summary overview is given in Table 1, which also shows the measurements that were being made as part of other research and monitoring programmes. Measurements made under this contract are shown in **bold** type. Additional measurements from the site with data reported here are shown in *italic* type.

Table 1. Summary of measurements made in 2008

Measurement	EMEP Level	Status (Funding)
<i>Meteorological data (30 min)</i>	I	CEH
Weekly bulk deposition of inorganic ions	I	Defra RMP2258– Acid Deposition Processes
<i>Daily wet-only deposition</i>	I	Defra/AEA Technology RMP2901 - Acid Deposition Monitoring Network
Monthly trace gases and particle concentrations	I	Defra/AEA Technology RMP2901 - Nitric Acid Network – Acid Deposition Monitoring
Trace gas and particle concentrations at PM_{2.5} and PM₁₀ (hourly)	II	This contract – MARGA
<i>Daily particle mass (PM_{2.5} and PM₁₀)</i>	I	Defra - AURN
Black carbon PM_{2.5} (30 min)	II	This contract - Aethalometer
Ammonia (monthly)	II	Defra RMP1906 -Ammonia in UK
<i>Ozone (hourly)</i>	I	Defra - AURN
<i>VOCs</i>	II	Defra RMP1833 - VOC network
<i>Heavy metals in precipitation (weekly)</i>	I	Defra CPEA32 - Heavy Metals
<i>Heavy metals in PM₁₀ (weekly)</i>	II	
<i>Mercury in precipitation (weekly)</i>	II	
<i>Speciated mercury in air (hourly)</i>	II	
Trace gas (O ₃ , NO _x , SO ₂) fluxes (hourly) (<i>concentrations only</i>)		Defra RMP2258 – Acid Deposition Processes
Monthly trace gas and particle fluxes		Defra RMP2258 – Acid Deposition Processes

2.4 Site modifications during 2008

There were no significant changes to the site infrastructure in 2008.

2.5 Site management and operation to meet EMEP Level II requirements

Effective operation of a ‘supersite’ over a period of several years requires that there is in place a management structure to ensure that access to the different instruments used for calibration, maintenance etc. does not compromise the operation of other measurements at the site. A designated site manager (Mr Robert Storeton-West) was appointed to oversee and coordinate all measurements at the site (including those made by CEH not under this or any other Defra contract). In most cases, routine maintenance of all the equipment, and standard calibrations and tests, was done by CEH staff. However, network protocols also required external maintenance and/or calibration visits for quality assurance, and such visits were coordinated as required.

In addition, the operation of a long-term site requires liaison with land owners and other users of the land, and ongoing maintenance of access routes and communications systems. This is particularly a problem at Auchencorth, where there is no road access. Heavy instruments, gas cylinders and water tanks (60 litres per week for the MARGA) must be transported to the site by an all-terrain vehicle. Health and safety requirements mean that lone working at the site is not usually permitted, particularly in winter.

CEH acted as local site operator for the following measurements (see also Table 1), which were operated under a separate Defra contract or as part of a UK monitoring network:

2.5.1 *Wet deposition*

The weekly bulk sampling of precipitation continued as part of the Defra-funded Acid Deposition Processes contract (RMP 2258).

Daily samples from a wet-only collector were collected every week and shipped to Netcen for analysis as part of the UK Acid Deposition Monitoring Project (RMP2901), under sub-contract agreement 14709935, following the protocols laid down for that network. Samples were analysed by ion chromatography for the concentrations of major anions and cations, pH and conductivity.

2.5.2 *Trace gas and aerosol concentrations*

The integrated monthly concentration measurements of NH_3 , SO_2 , HNO_3 , HCl gases and NH_4^+ , SO_4^{2-} , NO_3^- , Cl^- , Na^+ , Mg^{2+} , Ca^{2+} in aerosol continued under the Defra-funded Acid Deposition Monitoring contract with Netcen (RMP2901), sub-contracted to CEH as part of the Nitric Acid Network (Agreement 14709935). Measurements were made using coated denuders and filter packs (CEH DELTA samplers).

The integrated monthly flux measurements of NH_3 , SO_2 , HNO_3 , HCl gases and NH_4^+ , SO_4^{2-} , NO_3^- , Cl^- , Na^+ , Mg^{2+} , Ca^{2+} in aerosol also continued under the Defra-funded Acid Deposition Processes contract with CEH (RMP 2258). Measurements were made using the CEH COTAG (Conditional Time-Averaged Gradient) technique.

2.5.3 *Particle mass (PM)*

Automated particle samplers (Partisols) for measuring the daily integrated mass of both PM_{10} and $\text{PM}_{2.5}$ were already in operation as part of the Defra Particulate Monitoring Network, part of the Automated Urban and Rural Network (AURN). These were operated by CEH on behalf of the contractor Bureau Veritas (1/3/191). Exposed filters (daily integrated sampling) were removed two-weekly and shipped to the contractor for analysis.

Two TEOM/FDMS samplers ($\text{PM}_{2.5}$ and PM_{10}) were operated throughout the year. Data are collected by mobile phone by the AURN operators.

2.5.4 *Trace gases*

Measurements of vertical gradients of O_3 , NO_x and SO_2 by CEH are used to infer fluxes of these trace gases to/from the surface. Concentration measurements are cycled through 3 different heights, with 3 minutes at each measurement height. The

SO₂ measurements were part of the Acid Deposition Processes contract held by CEH (RMP2258). Data from the top height (2.5m) are reported here.

Ozone – a continuous UV-photometric instrument was operated by Bureau Veritas as part of the AURN (1/3/191). Data are recorded as hourly averages.

Volatile Organic Compounds (VOC) – an automated sampler/gas chromatograph was operated by AEA Technology at Auchencorth as part of the UK Hydrocarbons Monitoring Network (RMP1883). Data are recorded hourly.

Carbonyls (aldehydes and ketones) – automated samplers were constructed in 2006 for installation at Auchencorth and at Harwell. Following the EMEP protocol, two 8-hour integrated samples are scheduled to be taken in daylight hours every week. The exposed sampling tubes (commercially available packed cartridges containing dinitrophenylhydrazine, DNPH) are then shipped to AEA Technology weekly for analysis under the UK Hydrocarbons Monitoring Network (RMP1883). More frequent (manual) operation of the devices is possible during ‘intensive’ periods. Field testing of the samplers and analysis showed continuing problems; although earlier problems of contamination were overcome, the analytical sensitivity was inadequate to reliably characterise carbonyl concentrations at Auchencorth. The samplers were re-engineered in late 2008 to improve field operability. No data were collected.

2.5.5 *Heavy metals*

The Defra contract with CEH (CPEA32) includes Auchencorth as one of the UK Heavy Metal Network sites. Measurements include:

- Weekly sampling of precipitation for heavy metals (excluding mercury)
- Separate weekly sampling of precipitation for mercury
- Weekly integrated sampling of PM₁₀ aerosol for heavy metals
- Semi-continuous sampling of speciated airborne mercury (elemental mercury, reactive gaseous mercury, and particulate mercury) using an automated Tekran system. Data are reported as hourly averages every 2 hours; elemental mercury data are available every 5 minutes during every alternate hour, if required.

Precipitation and filter samples are sent weekly to CEH Lancaster for chemical analysis using ICP-MS (Inductively-coupled plasma mass spectrometry) for all heavy metals except mercury, and cold vapour atomic fluorescence for mercury. EMEP protocols are used for all heavy metal and mercury measurements.

3. **Operation of instruments under this contract**

Summaries of the data capture during 2008 are given in Table 2. Brief descriptions of the instrumentation and explanations of periods with missing data are given below.

3.1 *Meteorological measurements*

Measurements were made continuously of the following variables: wind speed, wind direction, air temperature, relative humidity, barometric pressure; precipitation amount, timing and duration. Data were stored as 30-minute averages. Wind direction data were averaged as vectors.

3.2 *Trace gas and aerosol concentrations (MARGA)*

The automated continuous-flow denuder and steam-jet aerosol sampler (MARGA) samples water-soluble trace gases and aerosol particles with hourly resolution, using a rotating wet denuder to remove gases from the sampled air stream before the residual particles (which pass through the denuder) are activated by steam into droplets, which are subsequently captured and analysed. The solutions of dissolved gases and dissolved particles are analysed on-line by ion chromatography, using parallel systems for cations and anions. A dual sampler was installed, with separate analysis of PM₁₀ and PM_{2.5} aerosols. Internal standards of Li⁺ (cations) and Br⁻ (anions) are used for calibration checks.

Data capture using the MARGA has been poor (around 40%), despite a dedicated operator and involvement of the manufacturer in advice and servicing. There has been no consistent type of problem, but a series of different and apparently unrelated issues:

January – problems with cation column and with air-flow consistency;

March – problems with anion column contamination;

May/June – failure of mass flow controller and long delay in replacement by manufacturer;

October/November – annual service by manufacturer, and repairs to mass flow controller;

December – contamination of sampling pre-concentrator, broken valve.

Repairs have been made and replacement parts have been installed as soon as possible after identification of the problem. The complexity of the instrument means that identifying the cause of any particular problem may take several days of investigation; repairs by the manufacturer typically have taken weeks.

3.3 *Particulate carbon (Aethalometer)*

The monitor for the continuous measurement of black carbon in air (aethalometer) was operated, recording data as 30-minute averages. The instrument had to be returned to the manufacturer for an upgrade and repair, so there are no data for February and a week of January and March. There was also a period without data in early November.

3.4 *Trace gases (ANNOX)*

Nitrogen oxides: a high-sensitivity automated NO/NO₂ analyzer (ANNOX) was installed and commissioned at the site in May 2007, but was subject to several initial problems, and ongoing problems with over-heating which were partially solved by installation of additional air-conditioning. The lack of a stable ozone analyzer (used for automatic calibration), and other calibration problems, meant that no data are reported for 2008. This chemiluminescent instrument uses UV photolysis to convert NO₂ to NO prior to analysis, as specified by the EMEP protocol. It is therefore specific for NO₂, and does not respond to other oxidised N species such as PAN and HNO₃, which are positive artefacts when using a thermal convertor.

3.5 *Wet-only sampler*

A daily wet-only sampler is operated as part of the UK Eutrophication and Acidification network (UKEAP), with chemical analysis done by Harwell Scientific under the UKEAP protocols. The sampler stopped working in mid-February and a

part had to be shipped to the manufacturer in Germany for repair, only becoming operational again in the last week of April.

Table 2. Data capture for instrumentation operated and reporting under this contract.

Instrument	% data capture for 2008
Wind speed and direction	94%
Air temperature	100%
Relative humidity	100%
Barometric pressure	100%
Surface wetness	99%
Precipitation (30-minute)	100%
Precipitation (daily)	80%
MARGA PM ₁₀	37% (at least one ion > l.o.d)
MARGA PM _{2.5}	39% (at least one ion > l.o.d)
MARGA gases	43% (at least one gas > l.o.d)
Aethalometer	80%
VOCs	56% (at least one VOC > l.o.d.)
Ozone (CEH)	99%
Sulphur dioxide	100%
Nitrogen oxides	99%
Daily PM ₁₀ and PM _{2.5}	91% (PM ₁₀), 82% (PM _{2.5})
Ozone (AURN)	98%
Hourly PM ₁₀ and PM _{2.5}	96% (PM ₁₀), 88% (PM _{2.5})
Mercury: Hg ⁰ , RGM, part	81%, 66%, 47%

4. Data collation and QA/QC

During 2007 a broadband link was established which permits most of the analytical instruments to be interrogated and controlled via the internet. This has led to improvements in detecting malfunctions, and a consequent improvement in data capture rates. All continuously-monitored data are recorded on a 'mirrored' server on site, and downloaded to a backed-up data store at CEH Edinburgh every night. Once data have been initially checked for instrument malfunction or obvious errors by dedicated CEH staff, the 'raw' data are uploaded to a web-accessible database, and available to any user with password access. Data are then ratified after identification of any long-term drifts in instrument response and recalibration (if necessary). The datasets are updated as required and marked in the database as 'ratified'. Data from other UK networks which sample at Auchencorth are downloaded (after ratification) from the Air Quality database (<http://www.airquality.co.uk/index.php>), or directly from the network operators, and uploaded into the 'Auchencorth EMEP' database at CEH Edinburgh. This means that all data collected at the site can be accessed simply, and can be reported to EMEP using the standard prescribed format.

The recommended maximum time scales for the release of non-quality-assured data that are collected as part of the operations of other networks or projects are as follows:

<u>Measurement</u>	<u>Data released within (period) after sampling</u>
Daily wet-only sample volumes	1 month
Daily wet-only ion concentrations	3 months
Monthly trace gas/aerosol concentrations (DELTA)	3 months
Monthly trace gas/aerosol concentrations (COTAG)	3 months
Hourly trace gas/aerosol concentrations (MARGA)	1 month
PM10/PM2.5 concentrations	1 month
Aethalometer	2 weeks
VOCs	2 weeks
Ozone, NO/NO ₂	2 weeks
Heavy metal concentrations (precipitation or air)	3 months
Mercury in precipitation	3 months
Mercury in air	2 weeks

Note that these data are to be used for quality assurance purposes only.

5. Reports on site operations and data

Formal reports are provided every 3 months to Defra, and take the form of short progress reports (form SID4), noting operational matters (e.g. data capture) and any problems with instrumentation or the site, and a summary of the data during the reporting period. This annual report brings together the information in the quarterly reports, and provides ratified data and summaries for the preceding calendar year (in this case 2008).

6. Summary data results

Plots for each of the data sets in Table 2 are presented in the Appendix. Statistical summary data are presented below in Table 3. These data should be read in conjunction with the % data capture in Table 2 to indicate how representative they are for the period. In all cases, the minimum concentrations recorded were at or below the limit of detection of the instrument. Note that for VOCs, only those compounds that exceeded the limit of detection (l.o.d.) are reported.

Data may be viewed and downloaded from the website <http://emepdata.ceh.ac.uk/> after application for a password from CEH (jnc@ceh.ac.uk).

Table 3. Summary data for the period 1 January to 31 December 2008

Instrument	mean	median	min/max
Wind speed, m s ⁻¹ (30 min)	4.15	3.55	0.01/31
Air temperature, °C (30 min)	7.3	7.3	-8.4/23.2
Relative humidity, % (30 min)	84.4	87.5	40/96
Bar. pressure, kPa (30 min)	97.9	98.0	93.0/101.0
Precipitation (mm/30 min)	0.069	0	0/8.5
Precipitation (daily wet-only)	2.58	0.01	0/28.8
Daily rain concentrations: H ⁺ (µequivalent L ⁻¹)	11.4	5.4	178
NH ₄ ⁺	21.6	15.1	149
Na ⁺	62.9	24.3	1436
K ⁺	2.2	1.4	32
Ca ²⁺	8.6	5.5	68
Mg ²⁺	13.8	5.7	315
Cl ⁻	69.9	26.9	1605
NO ₃ ⁻	15.8	8.6	132
SO ₄ ²⁻	21.8	15.3	166
Non-sea SO ₄ ²⁻	14.4	9.1	134
Conductivity (µS cm)	14.1	10.5	92
Daily rain deposition: H ⁺ (µequivalent m ⁻²)	66	19	605
NH ₄ ⁺	85	46	984
Na ⁺	264	68	4509
K ⁺	9	5	99
Ca ²⁺	38	21	333
Mg ²⁺	59	20	961
Cl ⁻	294	78	4954
NO ₃ ⁻	66	26	875
SO ₄ ²⁻	91	46	643
Non-sea SO ₄ ²⁻	59	26	489
Rain wtd. concentrations: H ⁺ (µequivalent L ⁻¹)	10.9		
NH ₄ ⁺	14.2		
Na ⁺	43.9		
K ⁺	1.5		
Ca ²⁺	6.3		
Mg ²⁺	9.8		
Cl ⁻	48.8		
NO ₃ ⁻	11.0		
SO ₄ ²⁻	15.1		
Non-sea SO ₄ ²⁻	9.9		
MARGA PM ₁₀ : NH ₄ ⁺ (µg m ⁻³)	0.57	0.20	14
Na ⁺	0.62	0.36	14
Ca ²⁺	0.12	0.08	1.3
Mg ²⁺	0.11	0.09	0.6
Cl ⁻	1.09	0.60	13
NO ₃ ⁻	1.18	0.38	24
SO ₄ ²⁻	1.01	0.68	12
MARGA PM _{2.5} : NH ₄ ⁺ (µg m ⁻³)	0.61	0.20	12
Na ⁺	0.43	0.27	8.5
Ca ²⁺	0.09	0.06	1.0

Instrument		mean	median	min/max
	Mg ²⁺	0.07	0.06	0.8
	Cl ⁻	0.68	0.38	8.9
	NO ₃ ⁻	0.98	0.30	21
	SO ₄ ²⁻	1.02	0.56	19
MARGA gases: (µg m ⁻³)	NH ₃	1.06	0.54	32
	HCl	0.11	0.06	4.6
	HNO ₃	0.10	0.06	4.4
	HNO ₂	0.17	0.14	1.9
	SO ₂	0.27	0.13	16
Aethalometer (µg C m ⁻³)		0.23	0.15	4.6
VOCs (µg m ⁻³)	1,3-butadiene	0.019	0.0	3.4
	1-butene	0.0013	0.0	0.4
	cis-2-butene	0.00035	0.0	1.1
	trans-2-butene	0.0035	0.0	17
	ethane	1.76	1.71	14
	ethene	0.19	0.14	4.0
	ethyne	0.16	0.19	2.2
	isoprene	0.011	0.0	2.3
	propane	1.34	1.04	74
	propene	0.07	0.0	2.0
	n-butane	0.75	0.51	78
	iso-butane	0.42	0.31	30
	n-pentane	0.24	0.15	28
	iso-pentane	0.32	0.21	26
	n-hexane	0.064	0.07	7.0
	n-heptane	0.024	0.0	23
	benzene	0.11	0.16	15
	toluene	0.11	0.15	17
	o-xylene	0.023	0.0	19
	m+p-xylene	0.053	0.0	39
	ethylbenzene	0.028	0.0	20
Ozone (CEH) (ppb)		25.1	24.9	62
Sulphur dioxide (ppb)		0.20	0.08	22
Nitrogen oxides:NO (ppb)		0.33	0.06	192
	NO ₂ (ppb)	2.1	1.2	34
Mass daily (µg m ⁻³)	PM _{2.5}	5.4	4	36
	PM ₁₀	8.5	7	39
Mass hourly (µg m ⁻³)	PM _{2.5}	3.3	2	44
	PM ₁₀	6.6	5	48
	Involatile PM _{2.5}	2.9	2	35
	Involatile PM ₁₀	5.5	4	39
	Volatile PM _{2.5}	0.41	0	13
	Volatile PM ₁₀	1.11	1	18
Ozone (AURN) (µg m ⁻³)		60.2	58	142
Mercury: Elemental (ng m ⁻³)		1.09	1.07	0.8/2.9
	Reactive gaseous (pg m ⁻³)	0.21	0	27
	Particulate (pg m ⁻³)	1.12	0	67
Heavy metals in air (weekly) (ng m ⁻³)		As	0.26	0.19
				1.0

EMEP Supersite report for 2008

Instrument		mean	median	min/max
	Cd	0.17	0.03	5.9
	Cr	0.66	0.47	6.9
	Cu	1.21	0.70	10.7
	Ni	0.42	0.17	3.2
	Pb	2.25	1.61	13
	Se	0.44	0.27	6.4
	V	0.84	0.40	7.1
	Zn	5.93	3.00	30
(two weekly)	Hg	1.28	1.26	2.3
Heavy metals in rain (weekly)				
(ng L ⁻¹)	As	0.10	0.08	0.36
	Cd	0.011	0.007	0.08
	Cr	0.09	0.06	0.35
	Cu	0.63	0.52	3.5
	Ni	0.21	0.14	1.6
	Pb	0.42	0.27	4.0
	Se	0.15	0.14	0.43
	V	0.24	0.15	1.2
	Zn	4.4	3.4	18
(monthly)	Hg	5.2	4.5	11

7. Discussion

7.1 Influence of wind direction

The site at Auchencorth is dominated by winds from the south-west, with less frequent winds from the north-east (Figure 2a). In contrast to earlier years there were easterly winds at the site for a significant proportion of the time. The strongest winds occur from the south-west (Figure 2b). The overall pattern for the year, however, disguises significantly different patterns from month to month, with some months dominated by more easterly flow (e.g. October - December, Figure 2c). The wind sector distribution of trace gases and particles gives a crude indication of potential sources that influence the air quality at the site. Examples are shown in Figures 3a-j below.

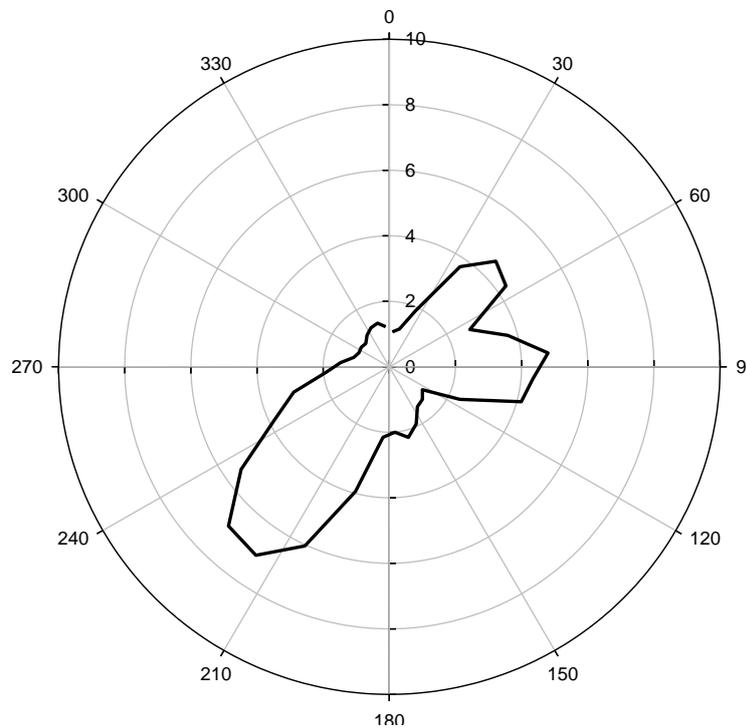


Figure 2: Distribution of wind direction frequency during 2008, expressed as % time (radial axis)

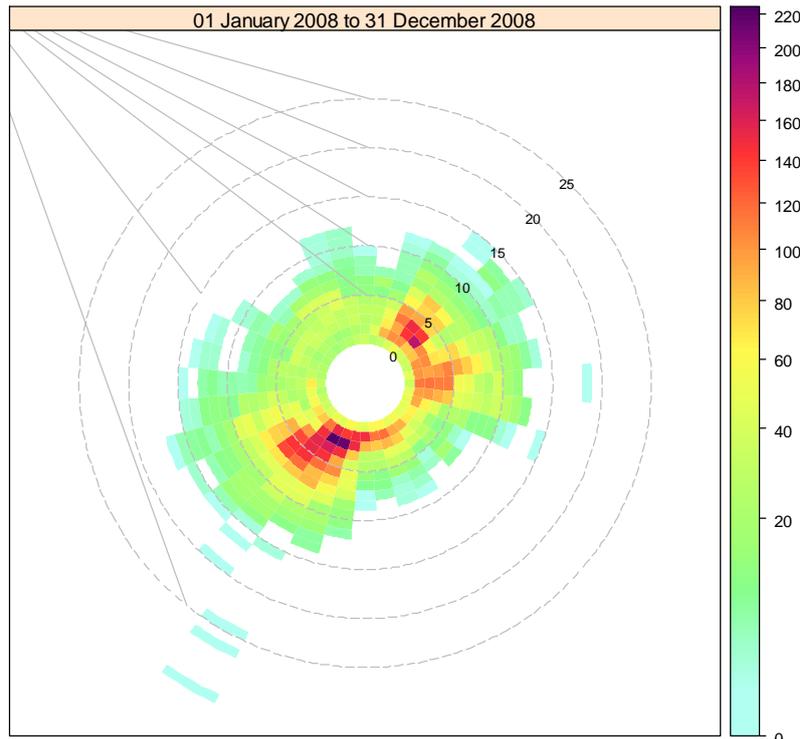


Figure 2b: Distribution of wind direction frequency (colour scale) during 2008, as a function of wind speed (m s^{-1}) on the radial scale. Note the very strong winds from the SW. Plot uses *openair* software (www.openair-project.org).

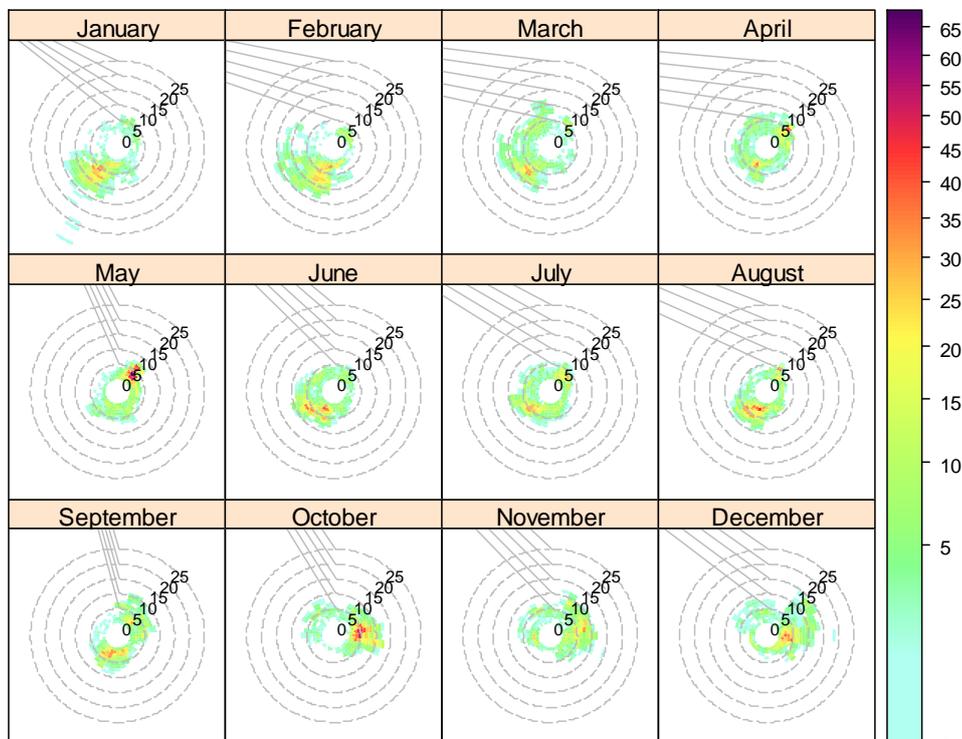


Figure 2c: Distribution of wind direction frequency (colour scale) during 2008, by month, as a function of wind speed (m s^{-1}) on the radial scale. Note the very strong winds from the SW in January and the preponderance of north-easterly flow in October-December. Plot uses *openair* software (www.openair-project.org).

The following plots were generated using ‘openair’ software (www.openair-project.org). The plots show the directional dependence of gas concentrations, with the colour showing the average concentrations for each sector. The annulus shows the dependence on time of day, with midnight in the inside of the ring, and midday at the mid-point of the ring. Spring is March, April, May; summer is June, July, August; autumn is September, October, November; and winter is December, January, February.

Concentrations of sulphur dioxide (SO_2) were low (median 0.08 ppb) and for much of the time close to the detection limit of the UV photometric analyzer deployed by CEH. Figure 3a shows the directional and seasonal dependence – the highest concentrations to the NW in summer probably originate from power stations at Grangemouth oil refinery and Longannet (coal-fired), while the peak to the N probably derives from the city of Edinburgh and the nearer small town of Penicuik. Peak concentrations in winter are seen in the middle of the night and the middle of the day, reflecting low wind speeds and inversion layers at night, and local transport in daytime. The afternoon peak to the NW in summer probably reflects better mixing during daytime and therefore longer-range transport, rather than patterns of emission. Concentrations from other wind directions are very low throughout the year.

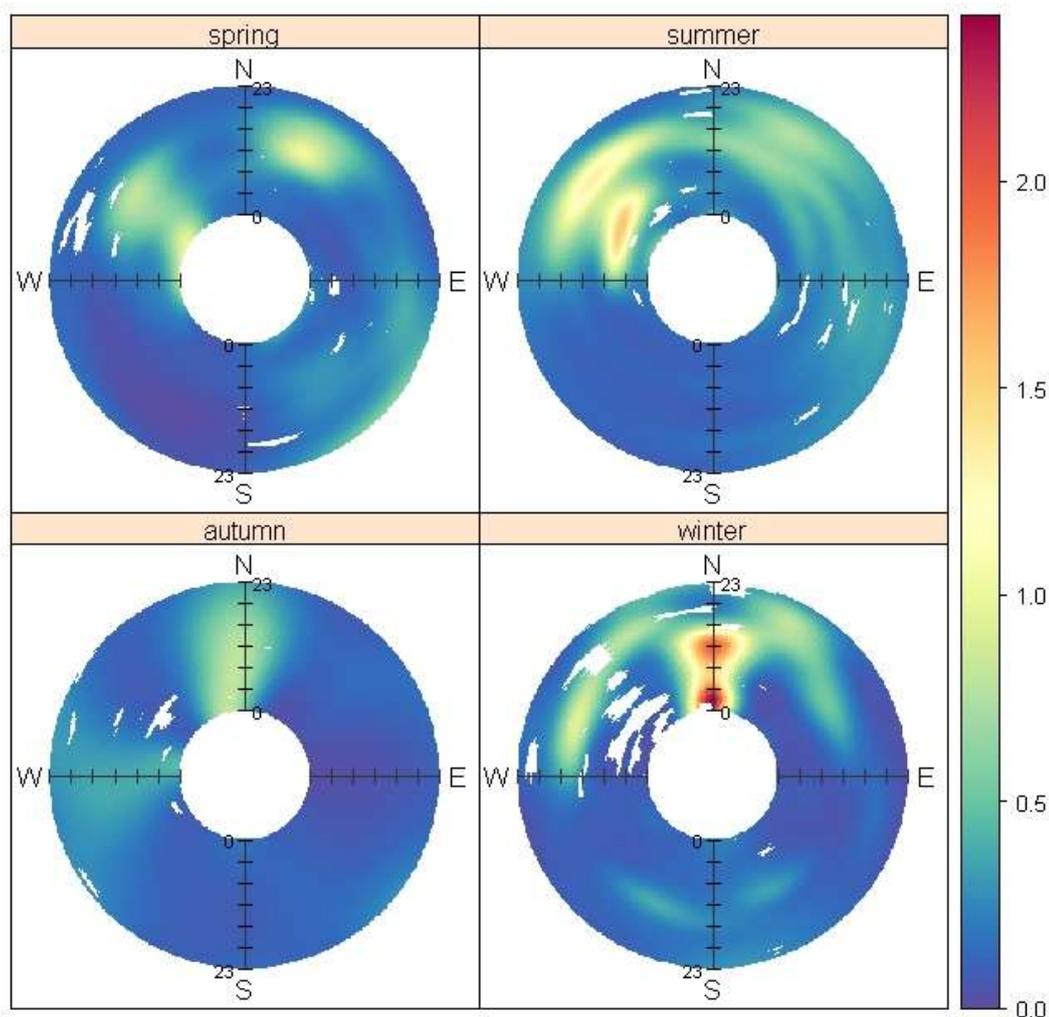


Figure 3a. Directional and diurnal dependence of SO_2 concentrations (ppb, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

The variation of ozone (O_3) concentrations (Figure 3b) shows the very strong seasonal cycle, with highest average concentrations in spring, and lowest in autumn and winter. The highest springtime concentrations are seen in south-easterly flow, typical of anticyclonic weather and transport of air from continental Europe. There is little evidence of episodic ozone production in summer, but the highest average concentrations occur in the evening, suggesting long-range transport of afternoon photochemical production. Low average concentrations (e.g. at night in autumn and winter) represent removal of O_3 below a nocturnal inversion, through titration with NO and deposition at the surface

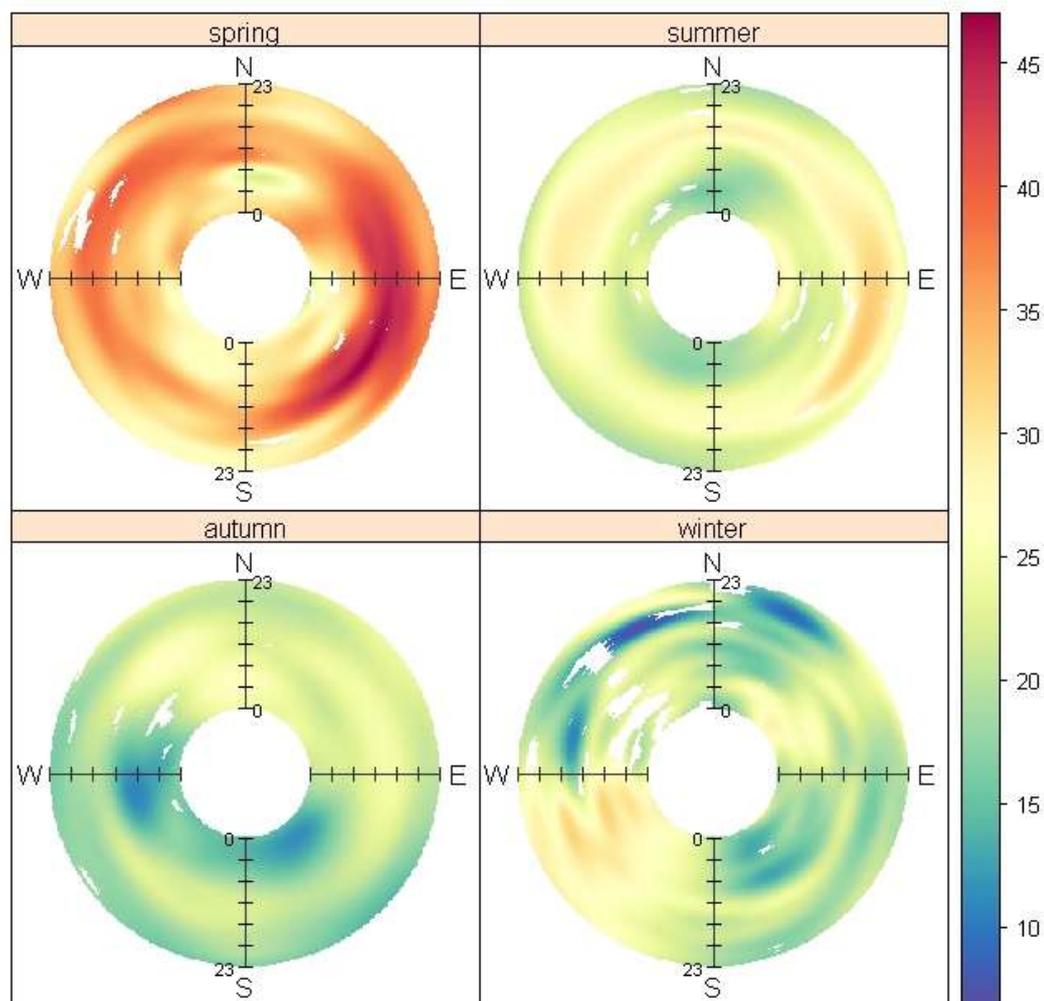


Figure 3b. Directional and diurnal dependence of O_3 concentrations (ppb, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

Nitric oxide (NO) concentrations were at or below the detection limit of the analyzer (0.5 ppb) for most of the time, so the directional data (Figure 3c) only show the effect of periods in winter when occasional high concentrations of NO were advected to the site from the urban areas to the north. The peak to the NNW is in line with a major motorway intersection and the Forth Road Bridge, but it is not clear why the peak in concentrations should occur during the evening rather than earlier in the day. The generally very low concentrations at night reflect the removal of NO by reaction with

O₃ during transport to the site, with small concentrations in day-time in summer and autumn arising from photolysis of NO₂.

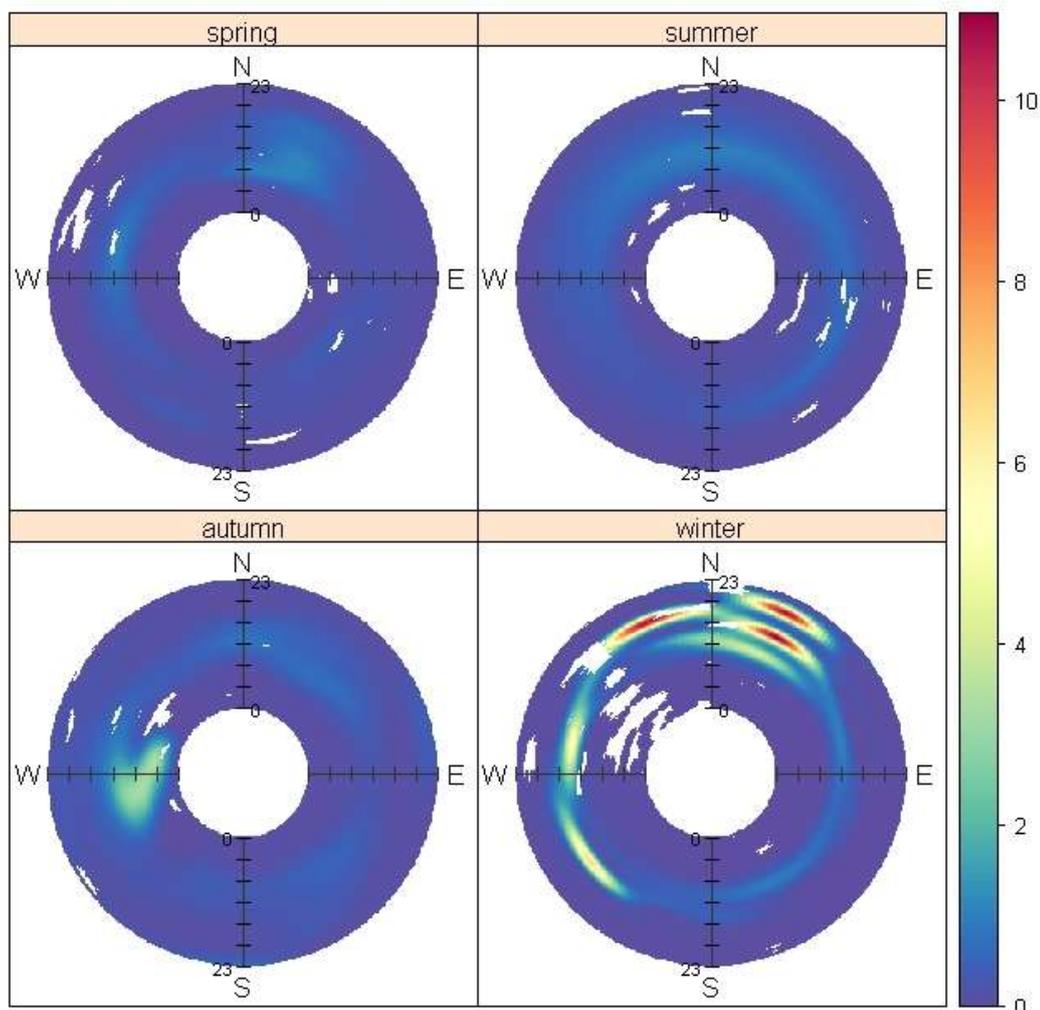


Figure 3c. Directional and diurnal dependence of NO concentrations (ppb, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

Concentrations of NO₂ were usually well above the instrument's detection limit, with a median for 2008 of 1.2 ppb. The overall patterns (Figure 3d) are similar to those for NO, with maximum concentrations in winter derived from relatively local emission sources from traffic and coal combustion. Even in spring and summer the effect of sources to the NNE of the site are evident, with also a diurnal pattern suggesting traffic sources from morning and evening rush-hours. The origin of the higher concentrations in westerly flow in autumn is not clear, but could reflect traffic emissions from the Glasgow conurbation, which is around 60 km due west of the site, or local emissions from the road A702 which passes through the village of Carlops about 6 km west of the site. The latter is possibly more likely, given the generally low wind speeds from the west during autumn (data not shown).

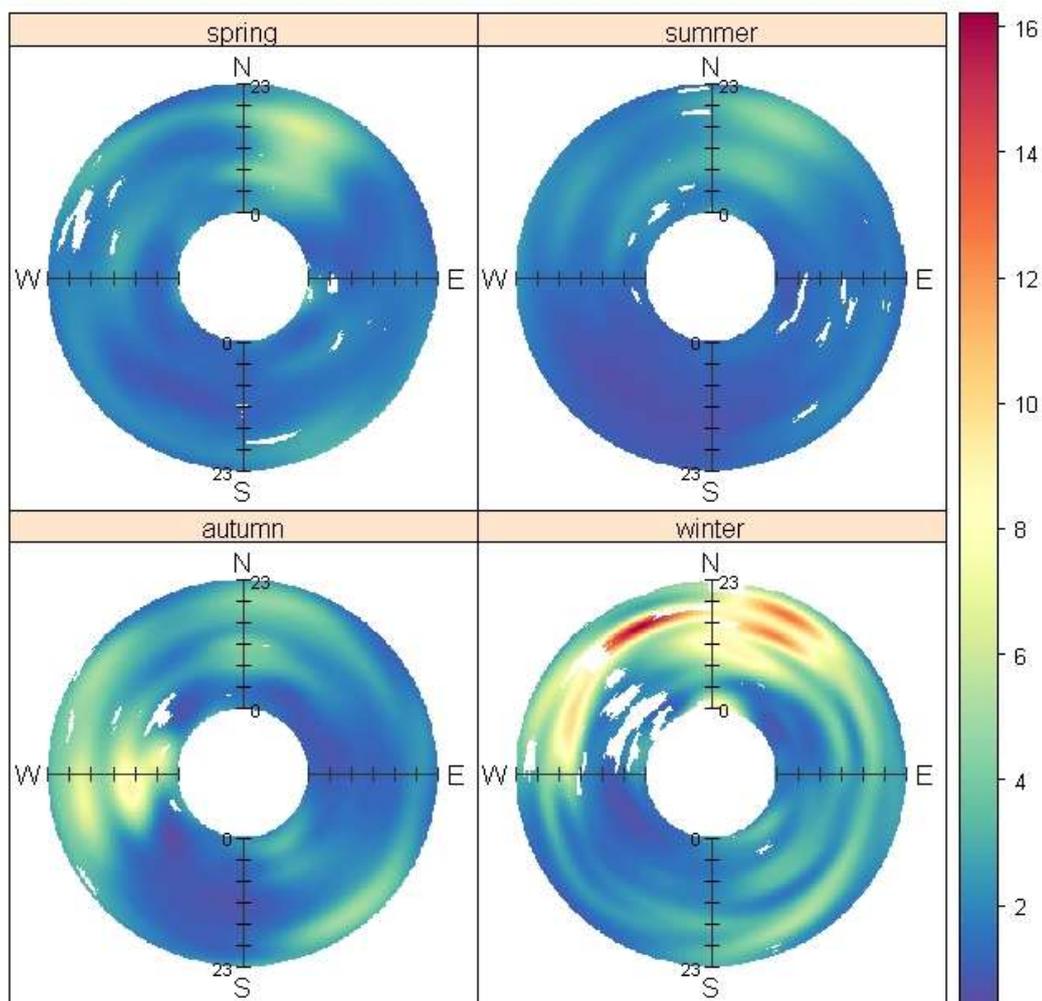


Figure 3d. Directional and diurnal dependence of NO₂ concentrations (ppb, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

Black carbon, as measured by the aethalometer, would be expected to show a similar pattern to combustion-related gases such as NO₂ and SO₂. In practice, however, although the urban areas to the NNE of the site show daytime and evening maxima, the largest average concentrations were observed in winter (excluding February, when no data are available), just to the N of west (Figure 3e), and possibly reflect local coal or wood burning for domestic heating.

The directional and diurnal patterns for short-chain hydrocarbons such as ethane and propane are dominated by high concentrations in springtime, which dictate the overall annual pattern (Figure 3f, propane). The pattern for benzene is different, with a less distinct maximum to the NNW and higher concentrations also associated with SE flow (Figure 3g). The spatial pattern for isoprene is different again, this time with a peak (again dominated by springtime) closer to NW, possibly from the moorland itself, or from the more distant refinery complex at Grangemouth (Figure 3h), which is presumably the source of the ethene measured at the site (Figure 3i).

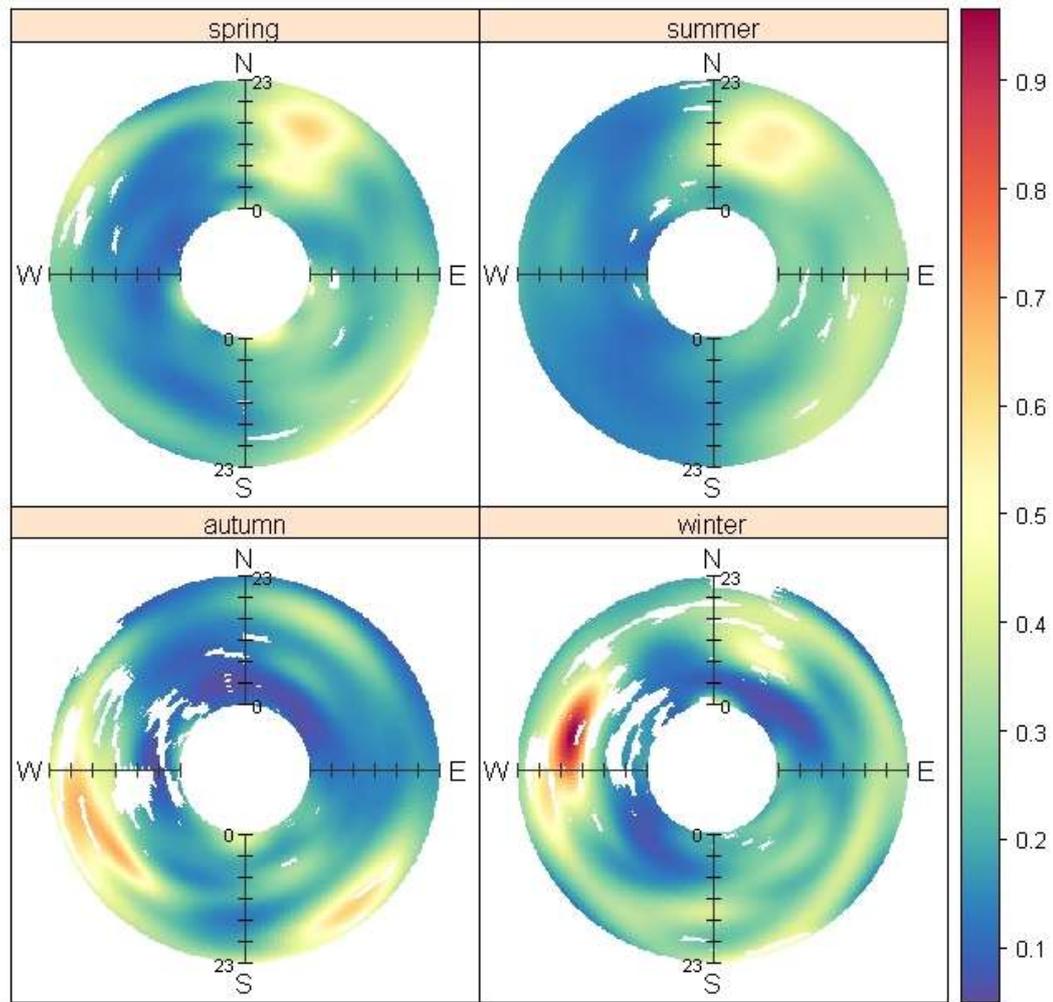


Figure 3e. Directional and diurnal dependence of black carbon (soot) concentrations ($\mu\text{g m}^{-3}$, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

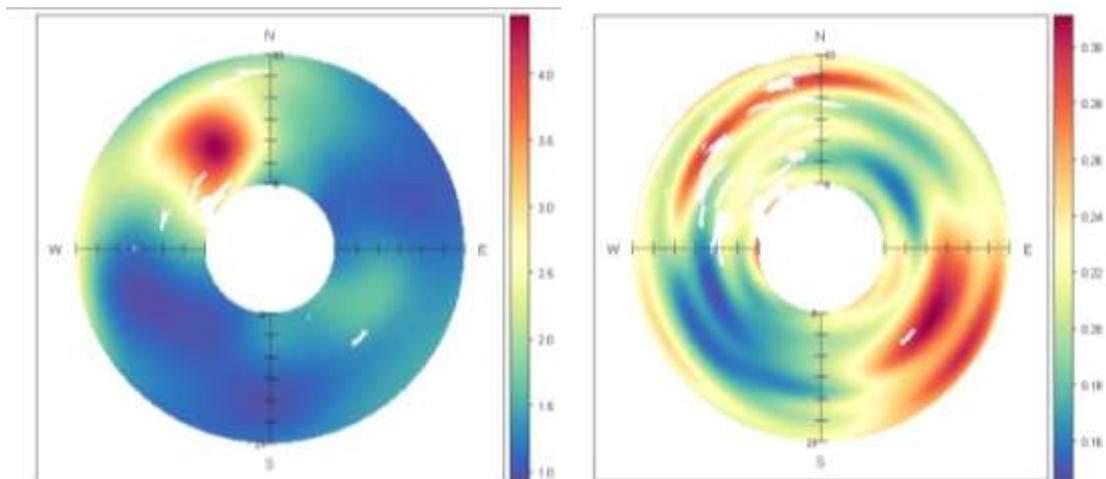


Figure 3f (left) and 3g (right). Annual directional and diurnal dependence of propane (left) and benzene (right) ($\mu\text{g m}^{-3}$, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

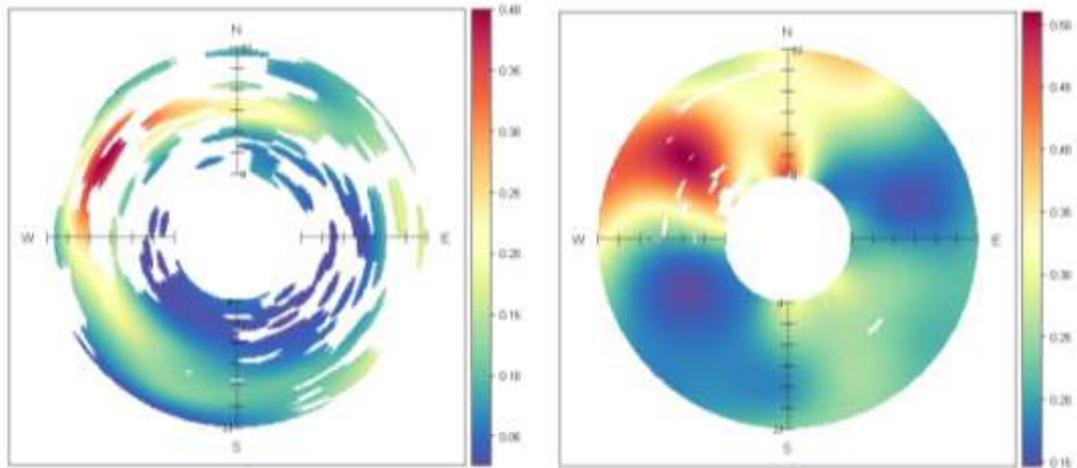


Figure 3h (left) and 3i (right). Annual directional and diurnal dependence of isoprene (left) and ethene (right) ($\mu\text{g m}^{-3}$, colour scale). Average concentrations at midnight are on the inside of the ring, and midday concentrations in the centre.

The directional dependence of elemental mercury concentrations is dominated by an episode with high concentrations in mid-February, associated with wind coming from Edinburgh and/or Cockenzie power station (coal burning) to the NNE (Figure 3j). Concentrations of reactive gaseous mercury (RGM) and particulate mercury (HgP) showed no clear directional dependence (data not shown).

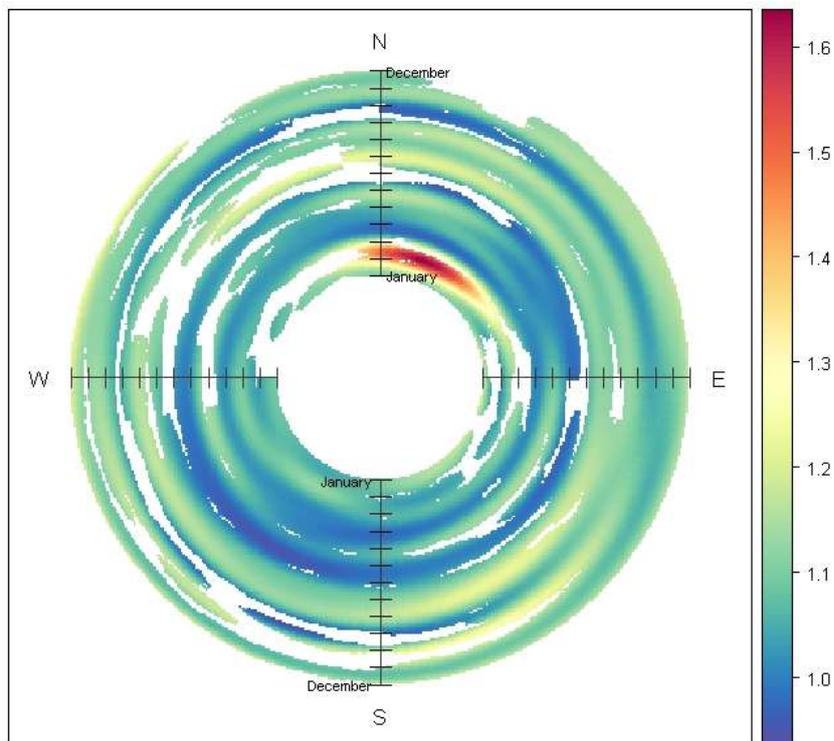


Figure 3j. Annual directional and seasonal dependence of elemental mercury concentrations (ng m^{-3} , colour scale). Average concentrations in January are on the inside of the ring, and summer concentrations in the centre.

7.2 Diurnal and seasonal cycles

Throughout the day, gas and particle concentrations vary systematically, as shown in Figure 4a-e, where averages across the whole year throughout the day are presented (left-hand frame). The middle frame shows the seasonal variation, as the average value each month, and the right-hand frame shows the variation averaged for each day of the week. All these plots were produced using the “openair” package.

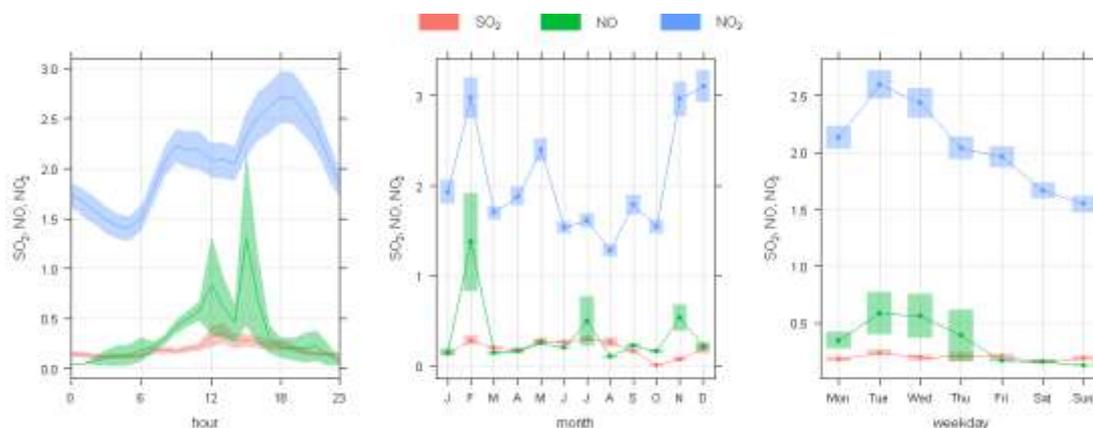


Figure 4a: Average temporal variation of sulphur dioxide and nitrogen oxides concentrations at Auchencorth in 2008. LH: diurnal, centre: monthly, RH: day of week. Plotted using *openair* (www.openair-project.org). the shading shows the 95% confidence interval of the mean.

Sulphur dioxide concentrations (Figure 4a) were very low overnight, with a broad peak in the middle of the day, then decreasing slowly through the evening. Average concentrations were much smaller than for the nitrogen oxides. For NO the peak is in daytime, as expected from photochemical production, but the pattern is noisy (the absolute detection limit for the instrument is 0.5 ppb). For NO₂ concentrations are lowest at night and show two peaks during the day which presumably relate to morning and evening increases in traffic flow. For all gases the peak concentrations are at the start of the week and are lowest at the weekend.

By contrast, ozone concentrations (Figure 4b) show a typical diurnal trend for rural air, with a clear diurnal cycle peaking in mid afternoon, and a minimum in the early morning. The small diurnal amplitude is consistent with a windy site, with a well-mixed boundary layer and little night-time depletion through dry deposition or reaction with nitric oxide. The seasonal pattern is also typical of a rural site in northern Europe, with a marked spring maximum in May, as noted above (Figure 3b). It is less easy to explain the weekday variation, other than to recognise the opposite pattern to that from NO₂, showing the effect of increased NO emissions at the start of the week and lower emissions at weekends.

The patterns for black carbon (Figure 4c) are similar to those for NO₂ (Figure 4a) as noted also in 2007, with a particularly clear minimum at the weekend (RH plot), but no obvious annual cycle. The hydrocarbons ethane and propane show similar behaviour (Figure 4d) with maxima in winter; for propane there is evidence of a diurnal and day-of-week pattern suggesting a contribution from vehicle emissions. Elemental mercury shows a strong diurnal cycle (Figure 4e) possibly reflecting night-

time deposition, and a minimum in spring. The day-of-week plot is dominated by the single large peak (Figure 3j) which occurred on a Wednesday.

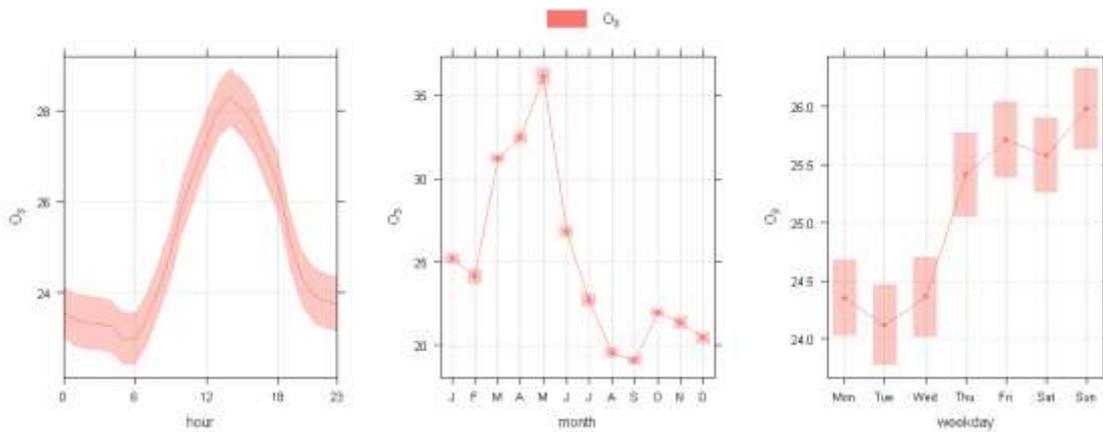


Figure 4b: Average temporal variation of ozone concentrations at Auchencorth in 2008. LH: diurnal, centre: monthly, RH: day of week. Plotted using *openair* (www.openair-project.org). the shading shows the 95% confidence interval of the mean.

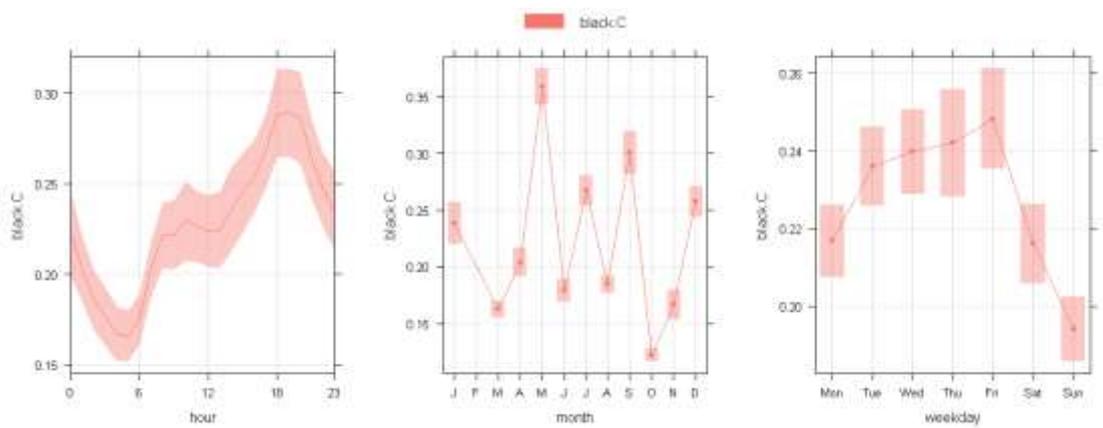


Figure 4c: Average temporal variation of black carbon concentrations at Auchencorth in 2008. LH: diurnal, centre: monthly, RH: day of week. Plotted using *openair* (www.openair-project.org). the shading shows the 95% confidence interval of the mean.

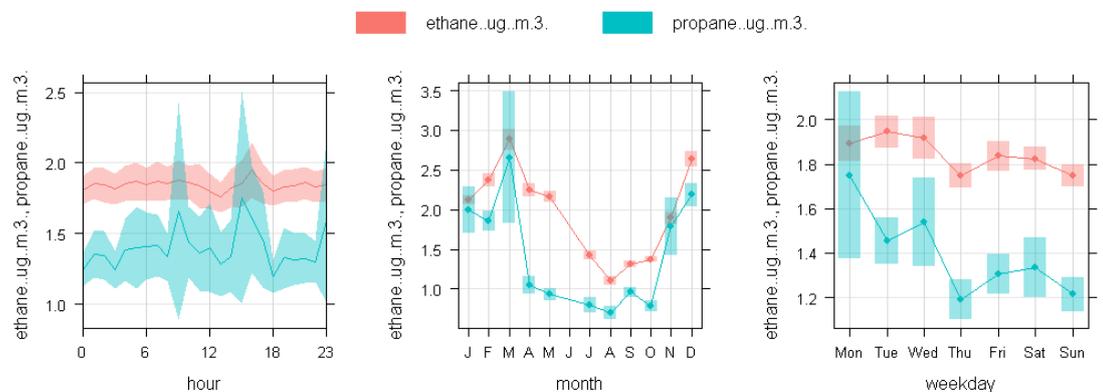


Figure 4d: Average temporal variation of hydrocarbon (ethane and propane) concentrations at Auchencorth in 2008. LH: diurnal, centre: monthly, RH: day

of week. Plotted using *openair* (www.openair-project.org). the shading shows the 95% confidence interval of the mean.

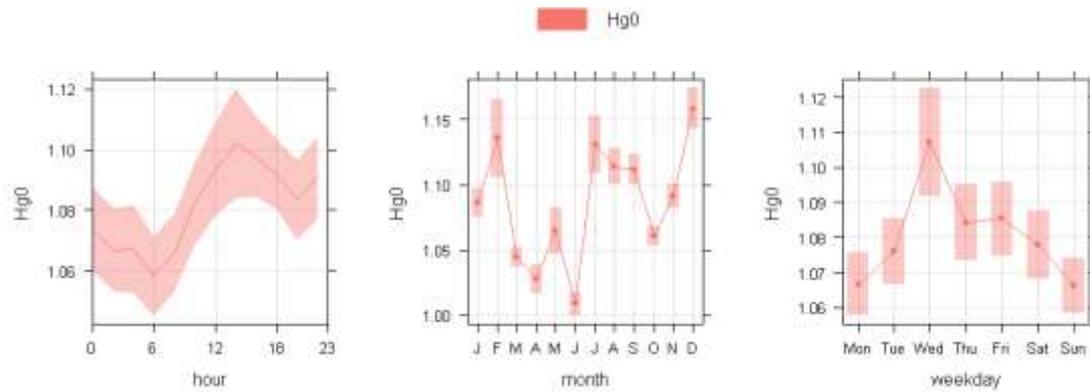


Figure 4e: Average temporal variation of elemental mercury concentrations at Auchencorth in 2008. LH: diurnal, centre: monthly, RH: day of week. Plotted using *openair* (www.openair-project.org). the shading shows the 95% confidence interval of the mean.

7.3 Frequency distributions

As discussed above, the air concentrations of gases and particles measured in 2007 at Auchencorth show rather different frequency distributions. Many gases and particles show a ‘classical’ log-normal distribution (Figure 5a), which can be used to predict the probability of particular concentrations being observed.

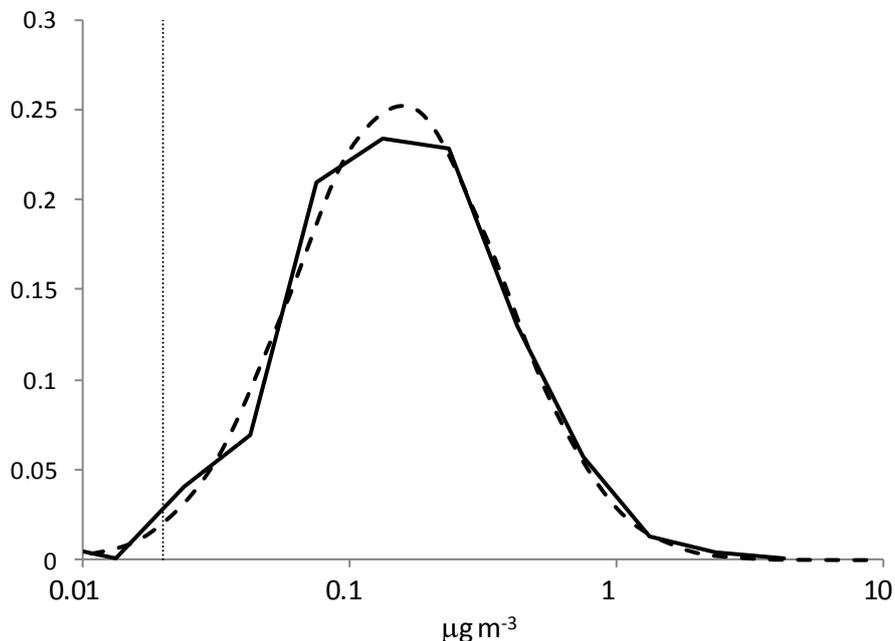


Figure 5a: Frequency distribution of 30-minute black carbon concentrations during 2008. Note the log scale on the x-axis. The dotted line shows the lower limit of detection, and the dashed line shows the best-fit to a log-normal distribution (see Figure 5b).

In order to investigate the form of the distribution in more detail it can be helpful to plot the data as a (log)-normal probability plot, where the x-axis is the standard normal variate (z); the (geometric) mean of the distribution is given when $z=0$, and unit differences from zero ($z = 1$ or -1) give the concentrations at 1 standard deviation either side of the mean. The slope of the plot therefore gives an estimate of the standard deviation, and the intercept with the vertical axis gives the mean. For black carbon the normal probability plot for the data above is shown in Figure 5b. A perfect log-normal distribution would give a straight-line plot. For black carbon, the points deviate from a straight line at higher concentrations, implying that high concentrations occur less frequently than would be expected for a perfect log-normal distribution. The effective limit of detection is at or below $0.02 \mu\text{g m}^{-3}$ ($\log_{10} = -1.7$). The best-fit line from Figure 5b is shown on Figure 5a as the equivalent frequency distribution, as a dashed line. Note that the slope of this plot for 2008 is identical to that for 2006 and 2007, and the geometric mean ($0.15 \mu\text{g m}^{-3}$) is also the same as in 2007.

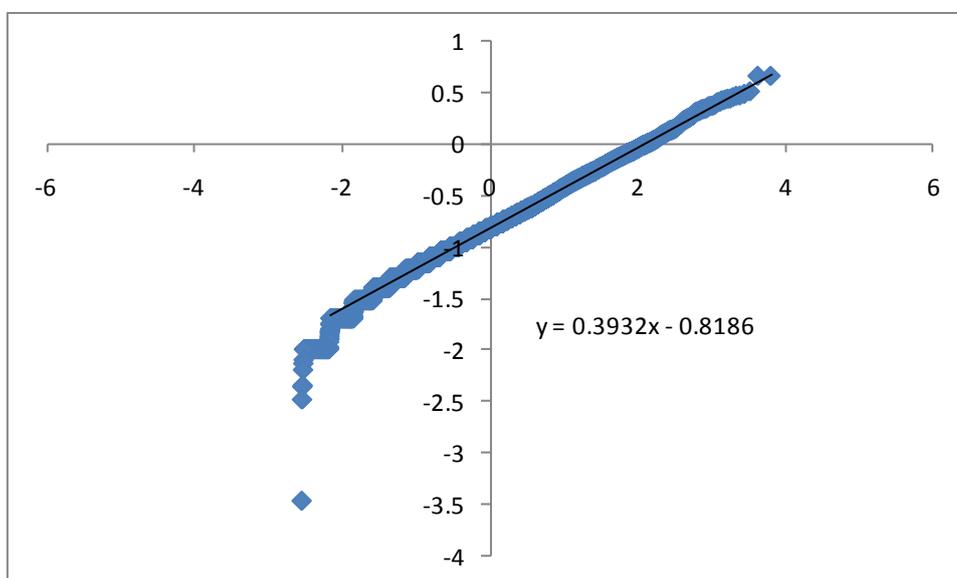


Figure 5b: Log-normal probability plot of black carbon concentrations during 2008. The horizontal axis shows the standard normal variate (z), and the vertical axis the \log_{10} concentration of black carbon in $\mu\text{g m}^{-3}$. The geometric mean is given by the intercept after conversion from logarithms ($-0.8186 \Rightarrow 0.152 \mu\text{g m}^{-3}$), and the geometric standard deviation is given from the slope ($0.3932 \Rightarrow 2.47 \mu\text{g m}^{-3}$)

The frequency distributions and normal probability plots are shown below for some of the other gases and particles measured in 2008. Figure 6 shows the daily data for particulate matter (PM).. The smaller number of valid data points than for hourly sampling give a less smooth pattern than for the black carbon data, but the PM data can be seen to follow an approximately log-normal distribution.

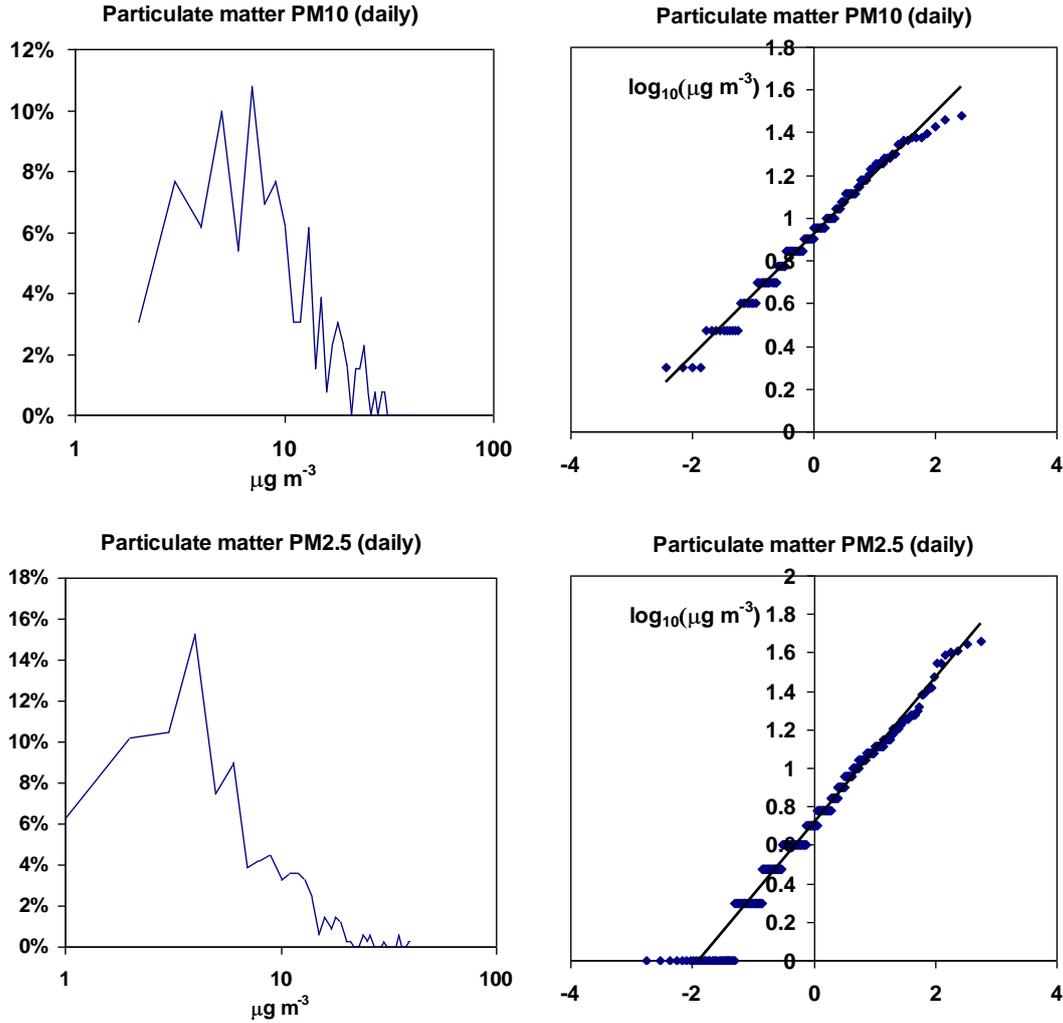


Figure 6: Frequency distribution and log-normal probability plot for daily PM₁₀ and PM_{2.5} mass concentrations during 2007.

Figure 7 shows data for sulphur dioxide. The data are fairly well described by a log-normal distribution (RH plot), but almost half the data set occurs below the instrumental detection limit (no data points for $z < 0$ in RH plot). The peak in the frequency distribution at 0.35 ppb may represent an artefact of making measurements close to the instrumental detection limit.

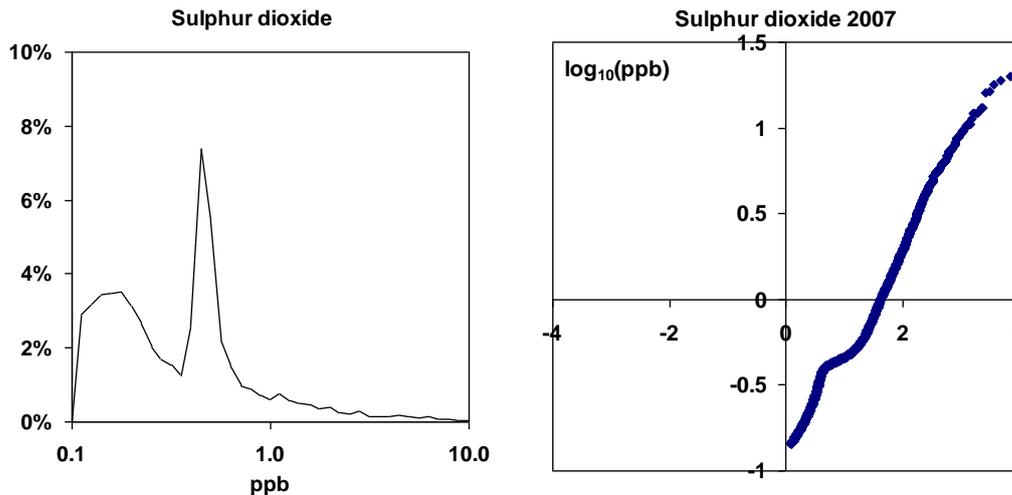


Figure 7: Frequency distribution and log-normal probability plot for sulphur dioxide during 2007

The frequency distribution of nitrogen dioxide also follows a log-normal pattern (Figure 8). There are too few data for nitric oxide above the instrumental detection limit (0.5 ppb) to determine the form of the distribution from the frequency plot alone, but the few measurable data suggest a log-normal distribution (dashed line in the normal-probability plot).

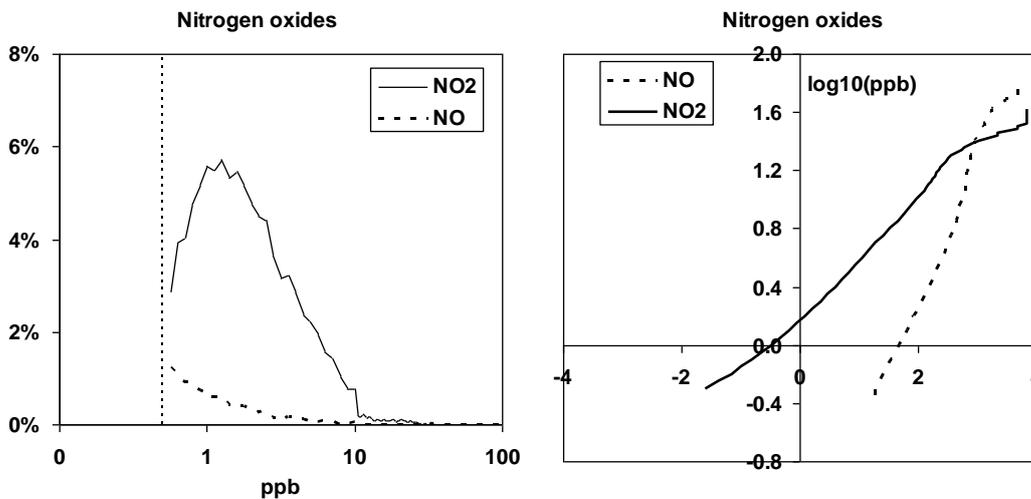


Figure 8: Frequency distribution and log-normal probability plot for nitrogen oxides during 2007

The frequency distribution for ozone is different from those above, in that most of the time at this site, the ozone frequency distribution is approximately normal (rather than log-normal), with only a small proportion of time when the ozone concentrations were greater than 50 ppb and higher than predicted from the overall distribution – these might be regarded as ozone ‘episodes’ (Figure 9.)

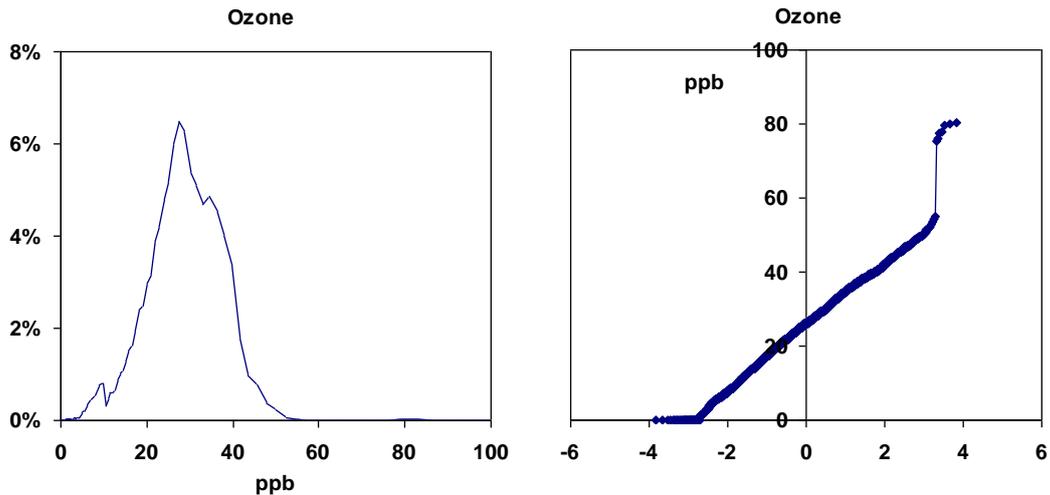


Figure 9: Frequency distribution and normal probability plot for ozone, 2007

Similar patterns of behaviour are also seen for elemental mercury (Hg^0), where the distribution of concentrations is approximately normal about the mean, and with a small proportion of the time where larger concentrations are observed (Figure 10). However particulate mercury shows a distribution more similar to log-normal, even though only 30% of the data are above the limit of detection (Figure 11). The picture for reactive gaseous mercury is even less clear, with only 10% of data above the limit of detection (Figure 12).

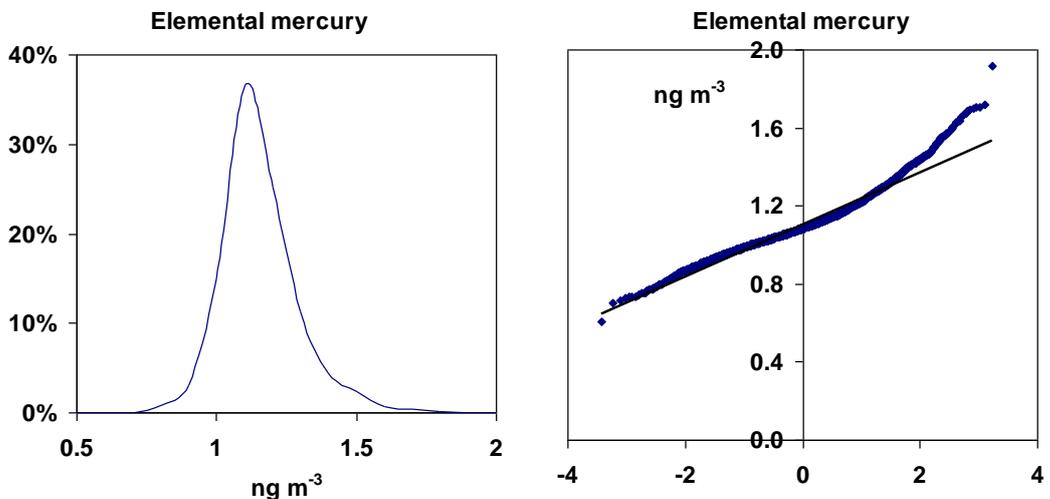


Figure 10: Frequency distribution and normal probability plot for elemental mercury, 2007

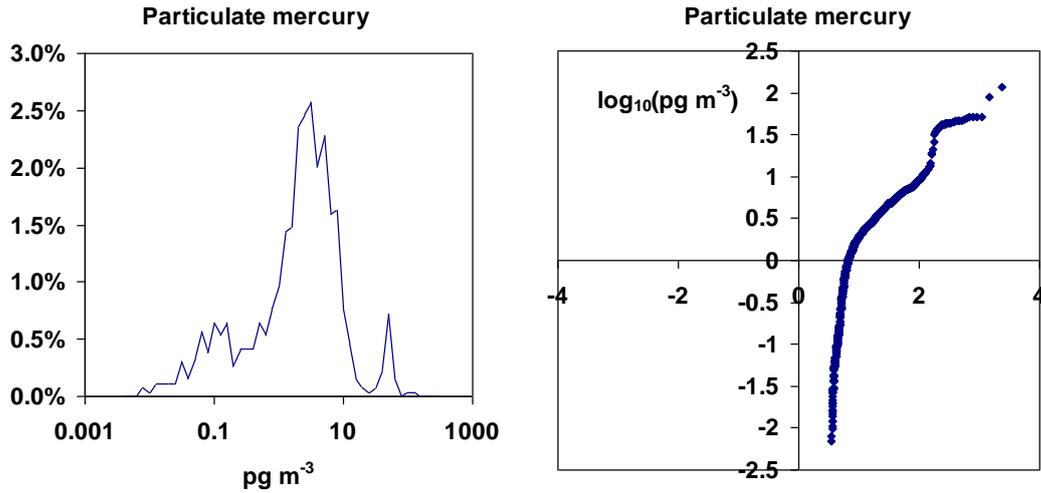


Figure 11: Frequency distribution and log-normal probability plot for particulate mercury, 2007

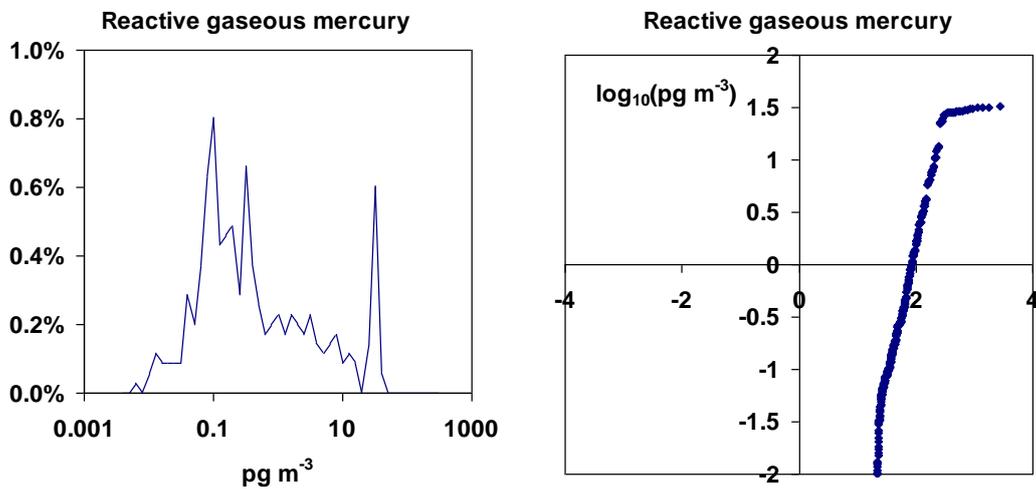


Figure 12: Frequency distribution and log-normal probability plot for reactive gaseous mercury, 2007

In 2007 for the first time there are data for a full year for VOCs. The frequency distributions for these compounds provide an insight into their sources and transport in the atmosphere. For example, the log-normal probability plots for 1,3-butadiene and isoprene show similar slopes (standard deviation), even though the absolute concentrations are very different (Figure 13). Both are compounds that are relatively rapidly oxidised in the atmosphere – typical lifetimes with respect to oxidation by the OH radical (assuming a concentration of $10^6 \text{ molec cm}^{-3}$) are 4.1h and 2.8h respectively. Lifetimes with respect to ozone reaction (at 25 ppb) are much longer, 60h and 47h respectively.

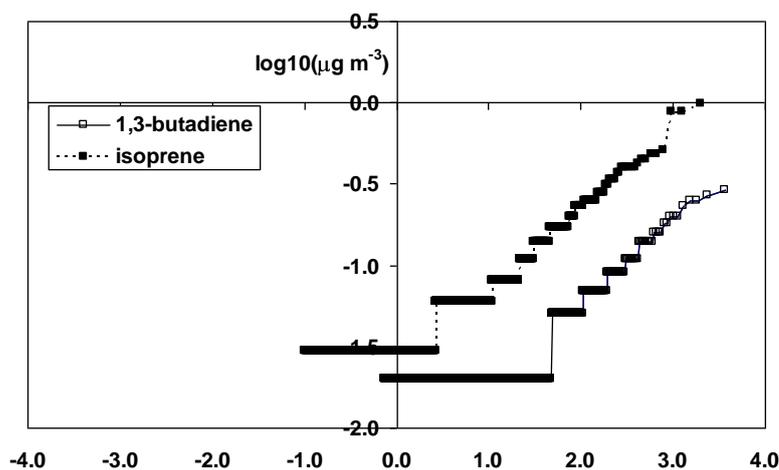


Figure 13. Log-probability distribution of 1,3-butadiene and isoprene, 2007.

8. Case studies

10-13 June 2007

During this period there was a consistent northerly air flow bringing surface air across the site. Wind speeds were initially low, increasing through the 12th and 13th June; local surface winds were north-easterly, with occasional movement to the north. The 48h air mass back-trajectories are shown in Figure 14, showing air flowing from the north-east, with periods of very low wind speed, particularly on 11th June (Figure 15) when local wind directions were variable.

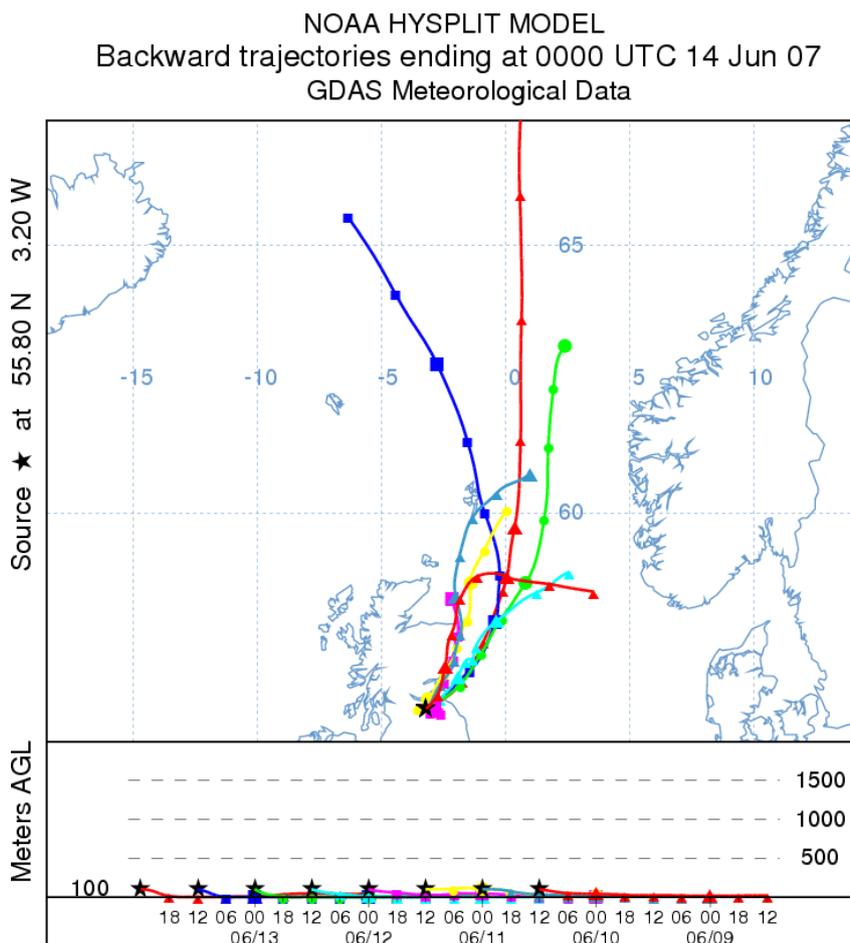


Figure 14a: HYSPLIT (www.ready.noaa.gov) trajectory maps for arrival times between 1200 on 10 June 2007 and 0000 on 14 June, showing the paths air parcels took during the 48h before arriving at the site. Symbols are shown every 6h. The lower figure shows the vertical path of the air parcels. Trajectories shown above start (red – origin N.Sea) with those arriving at 1200 on 10 June, and finish (red – origin Arctic) with those arriving at midnight on 13 June, and are shown at 6h intervals. The later air parcels (green, blue, red) are shown as more rapidly moving (longer).

NOAA HYSPLIT MODEL
 Backward trajectories ending at 0000 UTC 14 Jun 07
 GDAS Meteorological Data

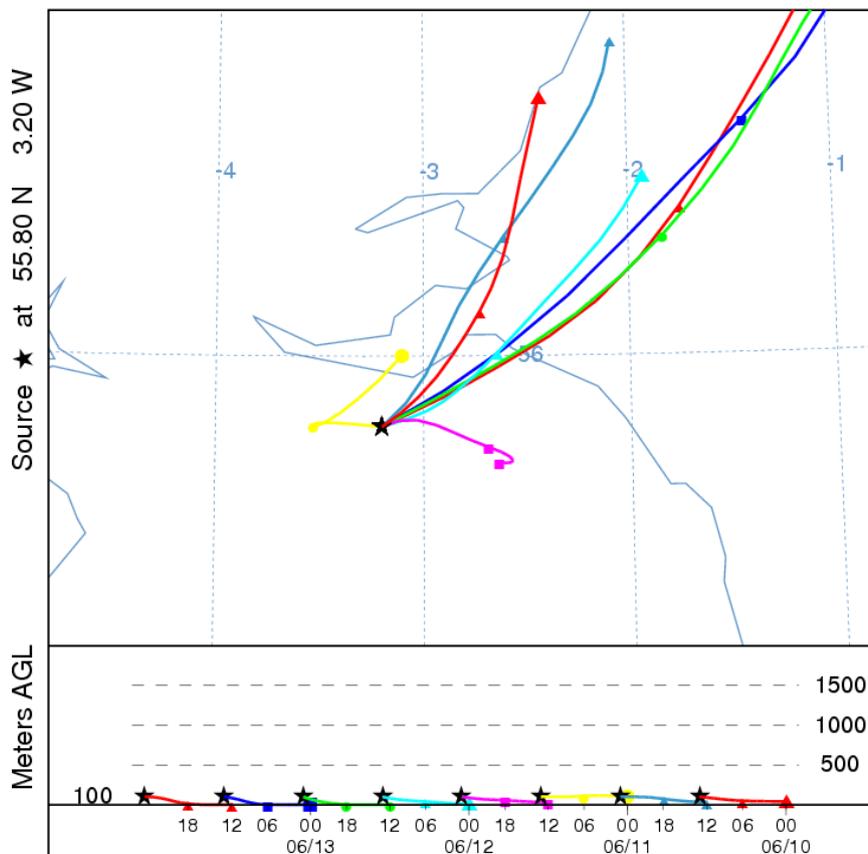


Figure 14b: As Figure 14a but showing only the last 12 h of each trajectory before arriving at the site, showing the air passing over Edinburgh, and the very low wind speeds (stagnation) during 11th June (yellow and pink lines).

The transport of slow-moving air over the city and upwind areas of Midlothian is reflected in the locally high SO₂ concentrations (Figure 16), and also HCl, associated with coal combustion. Sulphate particle and nitrate particle concentrations were also high, while sea-salt concentrations were very low, as would be expected in low-wind conditions, despite the trajectories from across the North Sea (Figure 17). Gas concentrations decreased rapidly as wind speed increased on 12th, even though the overall wind direction was unchanged, but particle concentrations, particularly for sulphate, remained high throughout 12th June, possibly reflecting long-distance transport of continental air (see Figure 14a, turquoise trajectory). Throughout the period, the nitrate and sulphate content of particulate matter was approximately balanced by ammonium ions.

This episode illustrates that even ‘remote rural’ sites occasionally experience the influence of regional air pollution, albeit at air concentrations that are still well below the short-term air quality standards for human exposure (1-hour mean concentration of SO₂ of 350 µg m⁻³). The maximum hourly SO₂ concentration recorded in the city of Edinburgh (St. Leonards) was 67 µg m⁻³, at 10:00 on 11th June (AURN data).

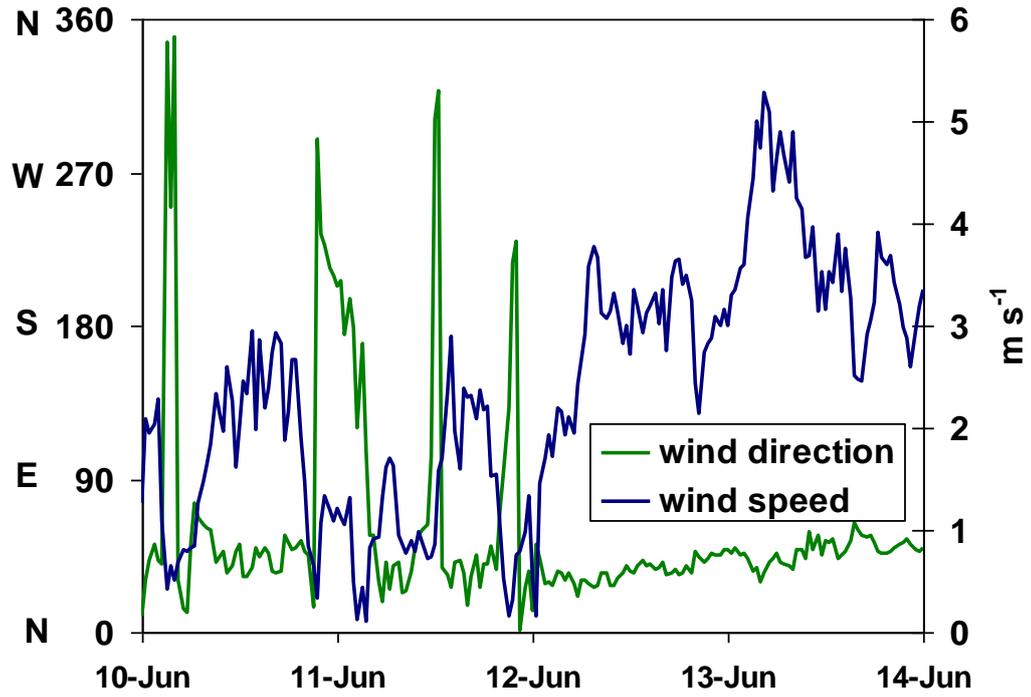


Figure 15: Wind direction and wind speed measured at the site between 10 and 14 June 2007.

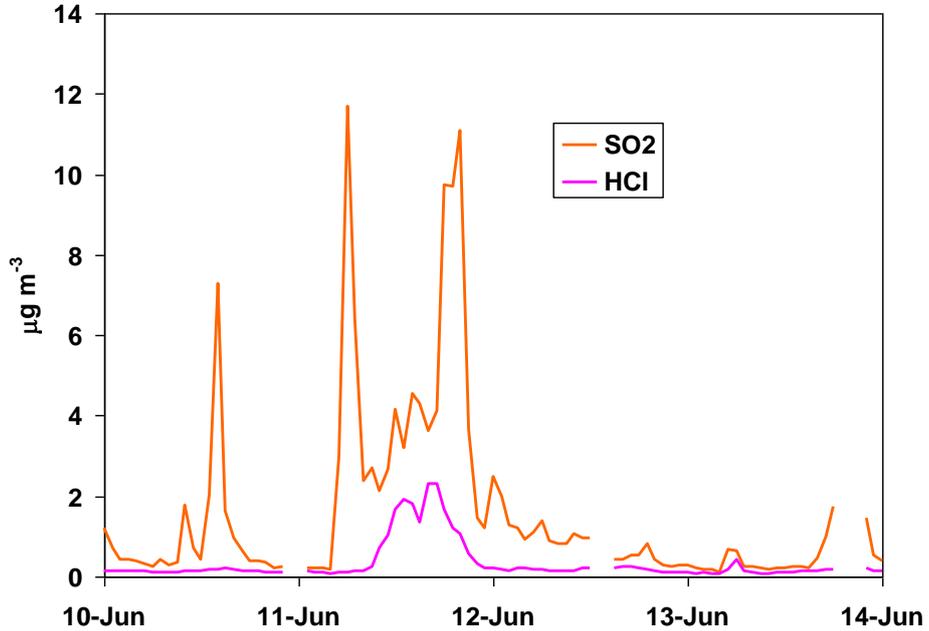


Figure 16: Concentrations of sulphur dioxide and hydrogen chloride sampled by the MARGA between 10 and 14 June 2007.

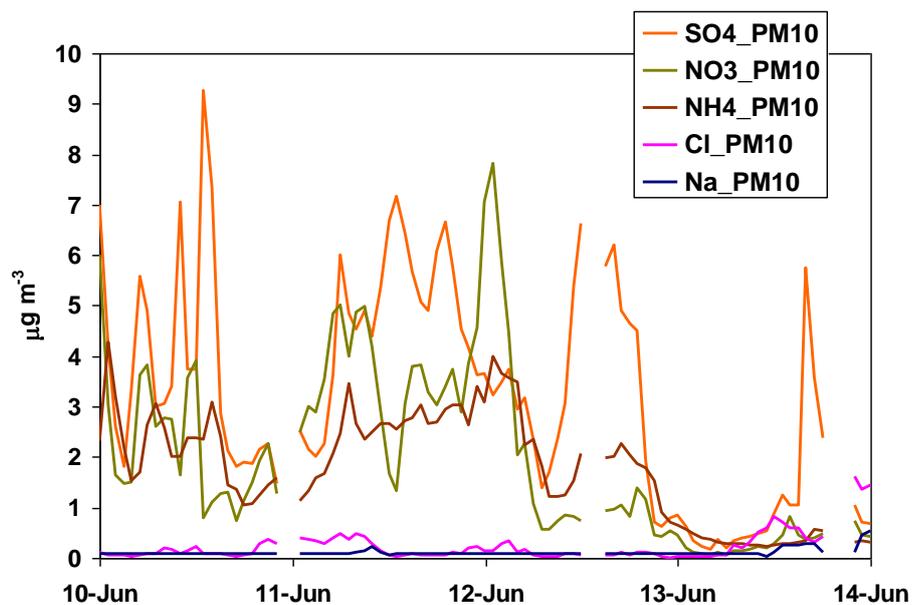


Figure 17: Concentrations of particulate matter (PM₁₀) sampled by the MARGA between 10 and 14 June 2007

APPENDIX – Data recorded during 2008

Meteorological data

Air temperature
Solar radiation and Photosynthetically Active radiation
Relative humidity
Atmospheric pressure
Surface wetness
30 minute rainfall
Wind speed
Wind direction

MARGA data (PM₁₀, PM_{2.5} and gases)

Ammonium
Ammonia
Sodium
Calcium
Magnesium
Chloride
Sulphur dioxide and Hydrogen chloride
Nitrate
Nitric and nitrous acid
Sulphate

Hydrocarbon data

1,3-butadiene and 1-butene
Ethene and ethyne
Ethane and propane
Isoprene and propene
iso-butane and iso-pentane
n-butane and n-pentane
Benzene and toluene

AURN data

Ozone
PM₁₀ and PM_{2.5} (hourly)
PM₁₀ (volatile and non-volatile)
PM_{2.5} (volatile and non-volatile)
PM₁₀ and PM_{2.5} (daily)

Mercury data

Elemental mercury (Hg⁰)
Reactive gaseous and particulate mercury

CEH flux gases

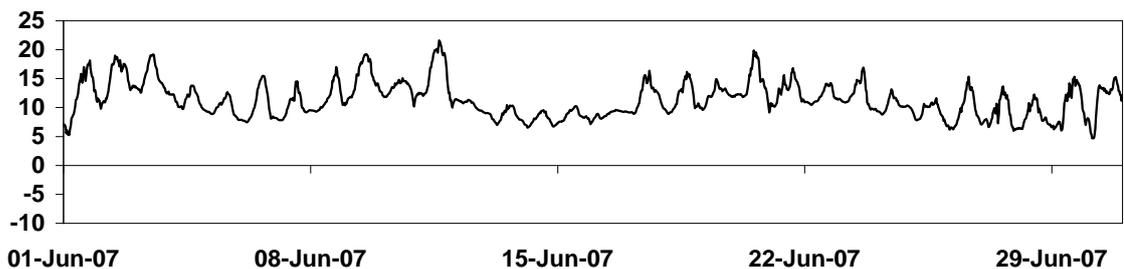
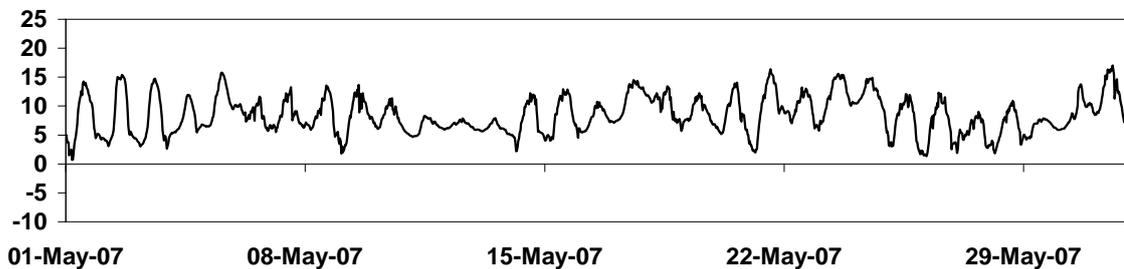
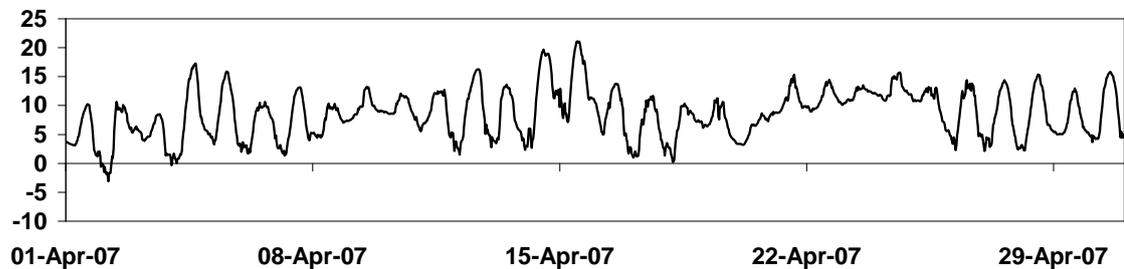
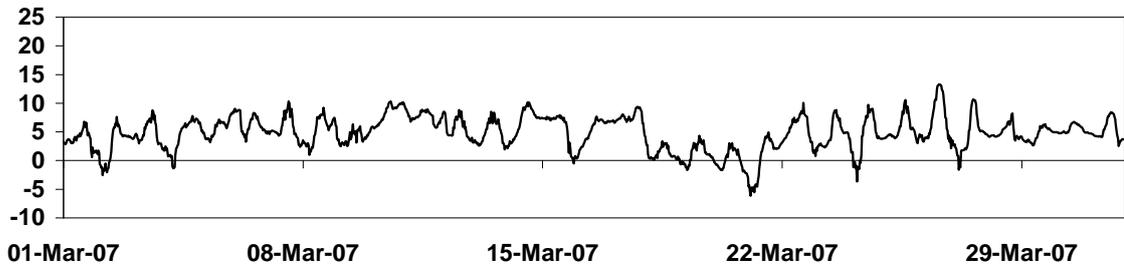
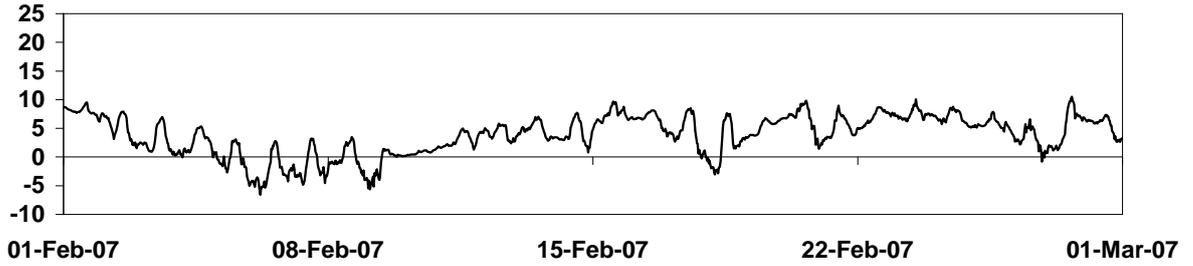
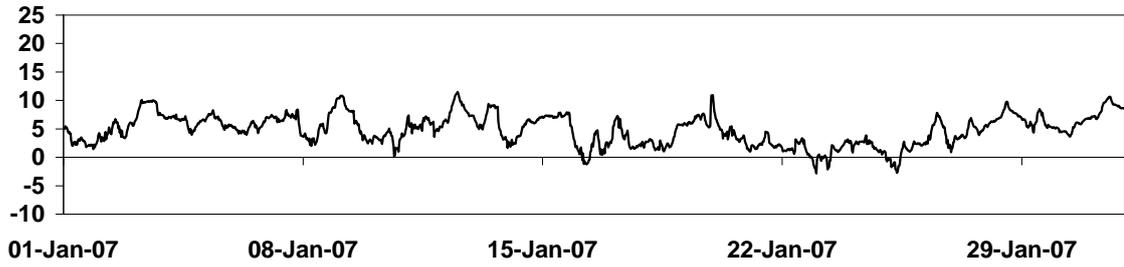
Nitrogen oxides
Ozone
Sulphur dioxide

Black carbon

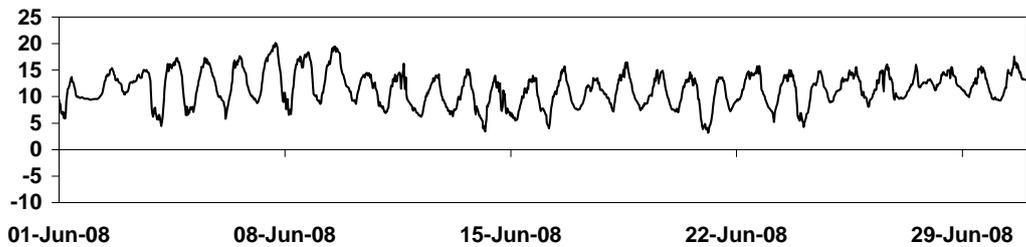
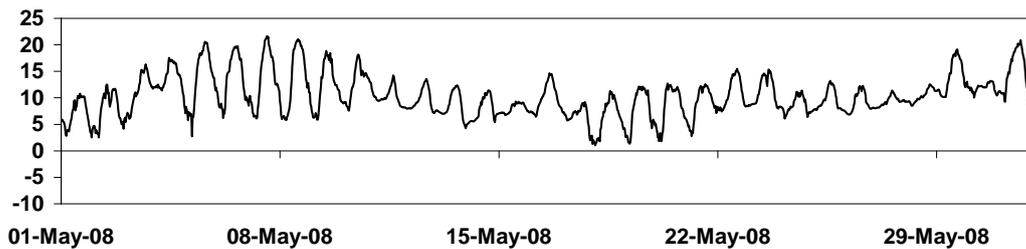
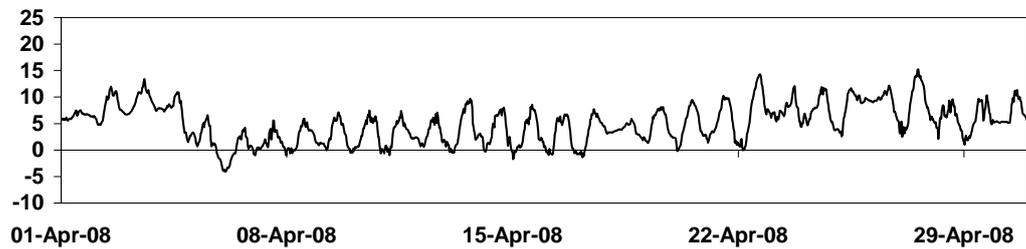
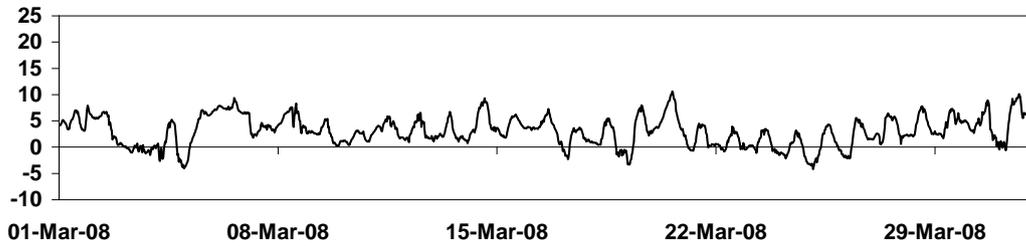
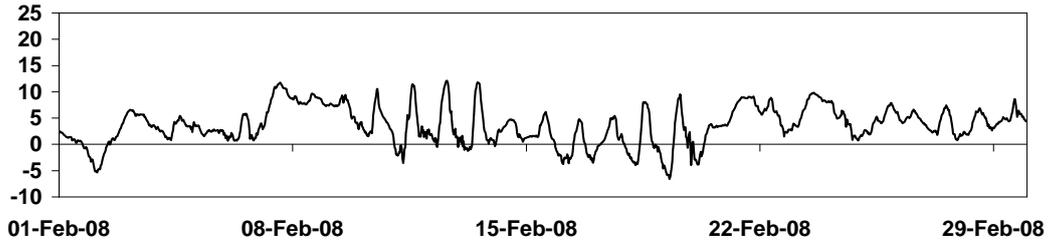
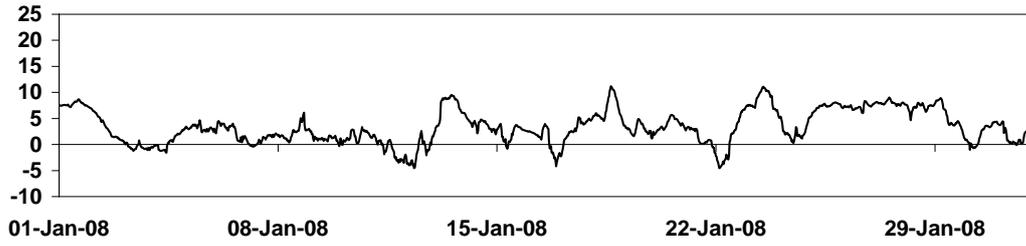
Wet-only precipitation

Daily rainfall
Concentrations of NH₄⁺, H⁺, Na⁺, Cl⁻; Mg²⁺, Ca²⁺; SO₄²⁻, NO₃⁻

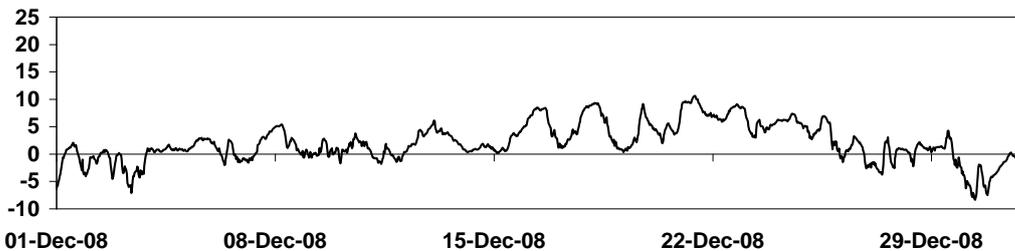
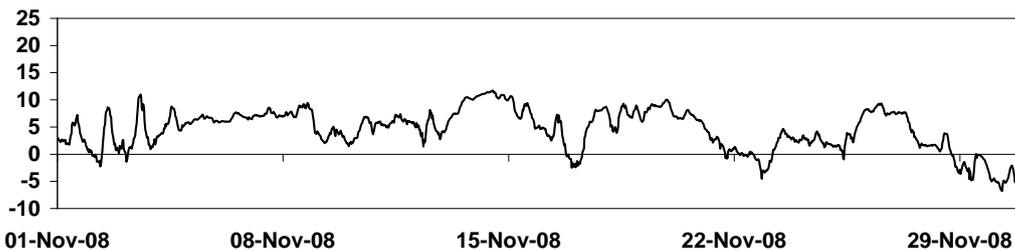
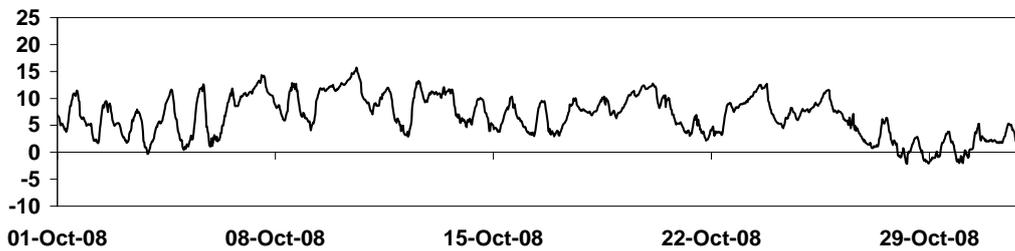
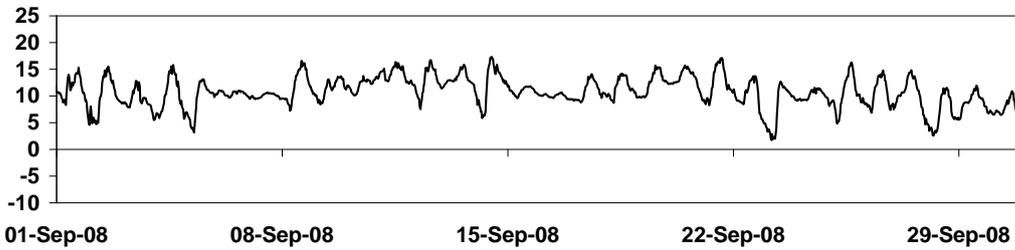
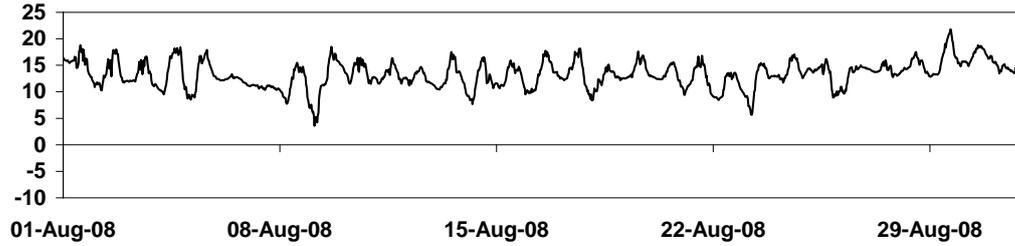
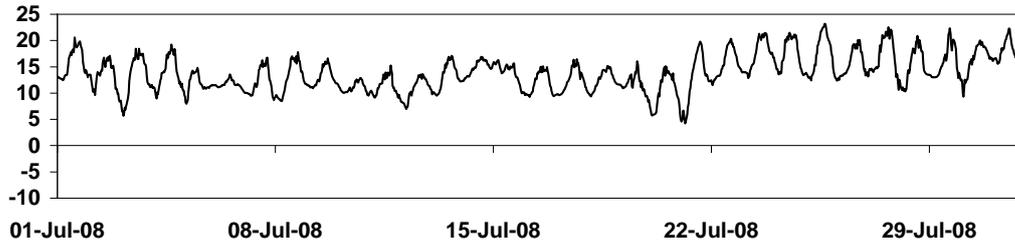
Air temperature °C



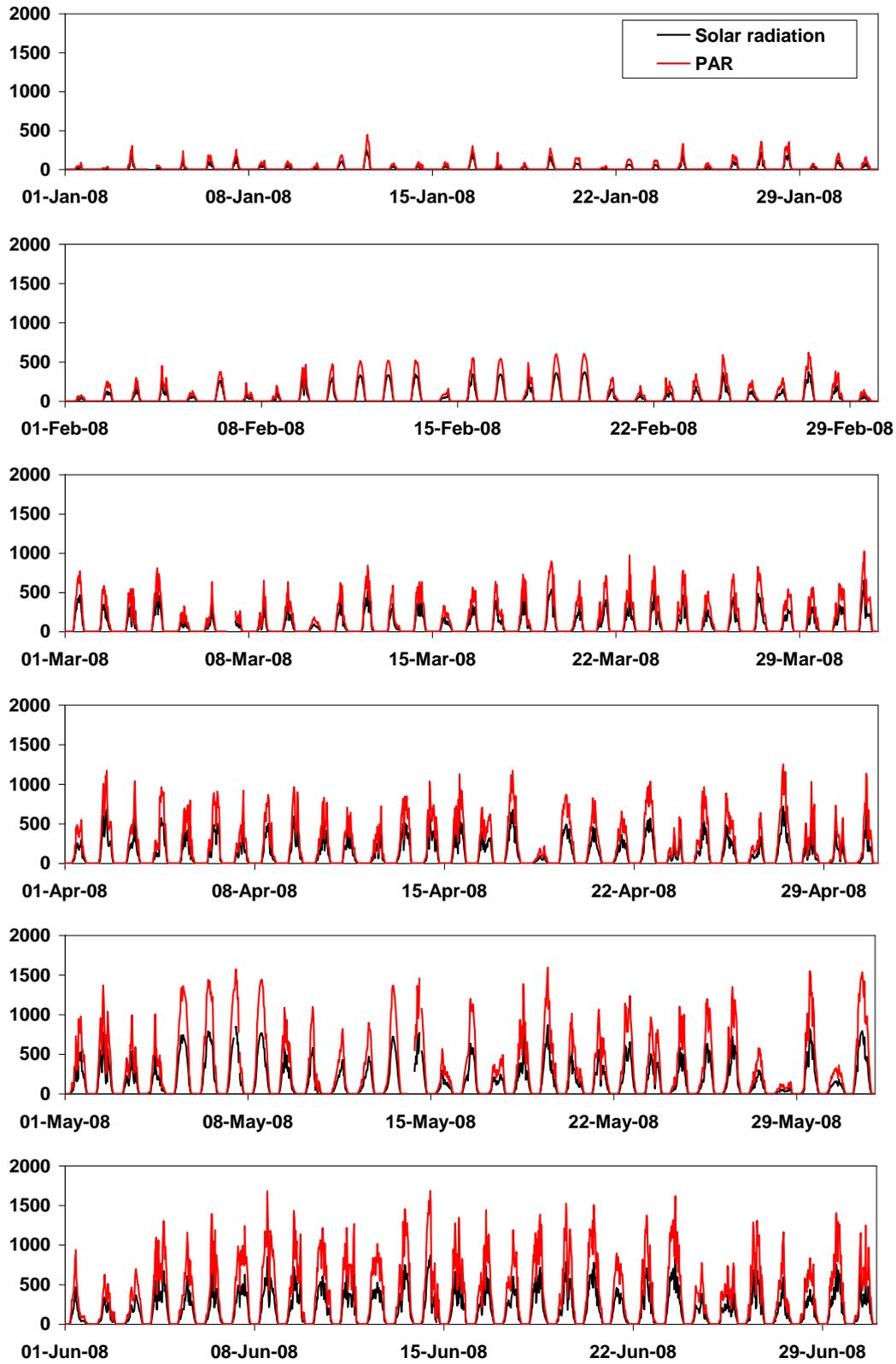
Air temperature °C



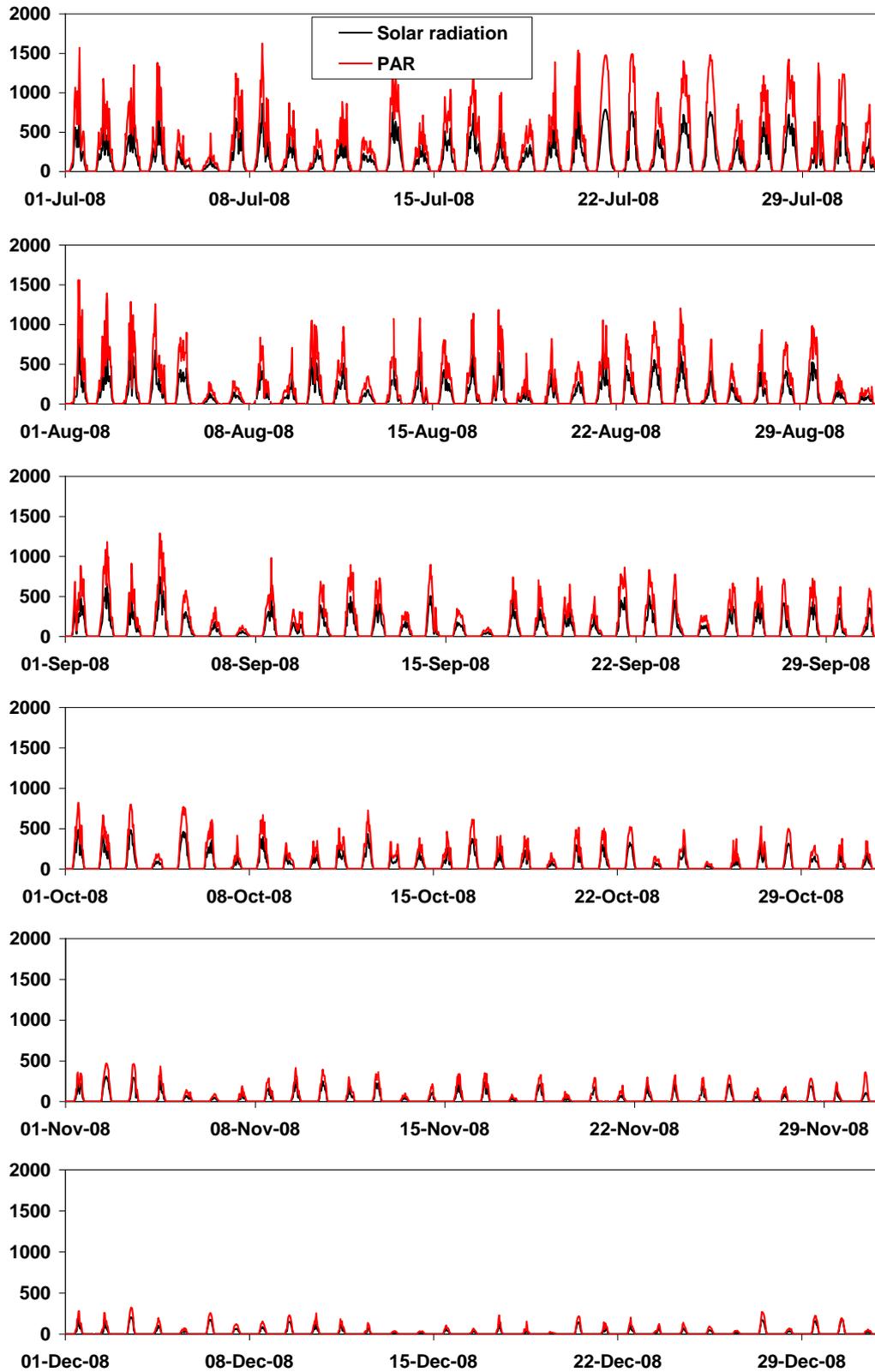
Air temperature °C



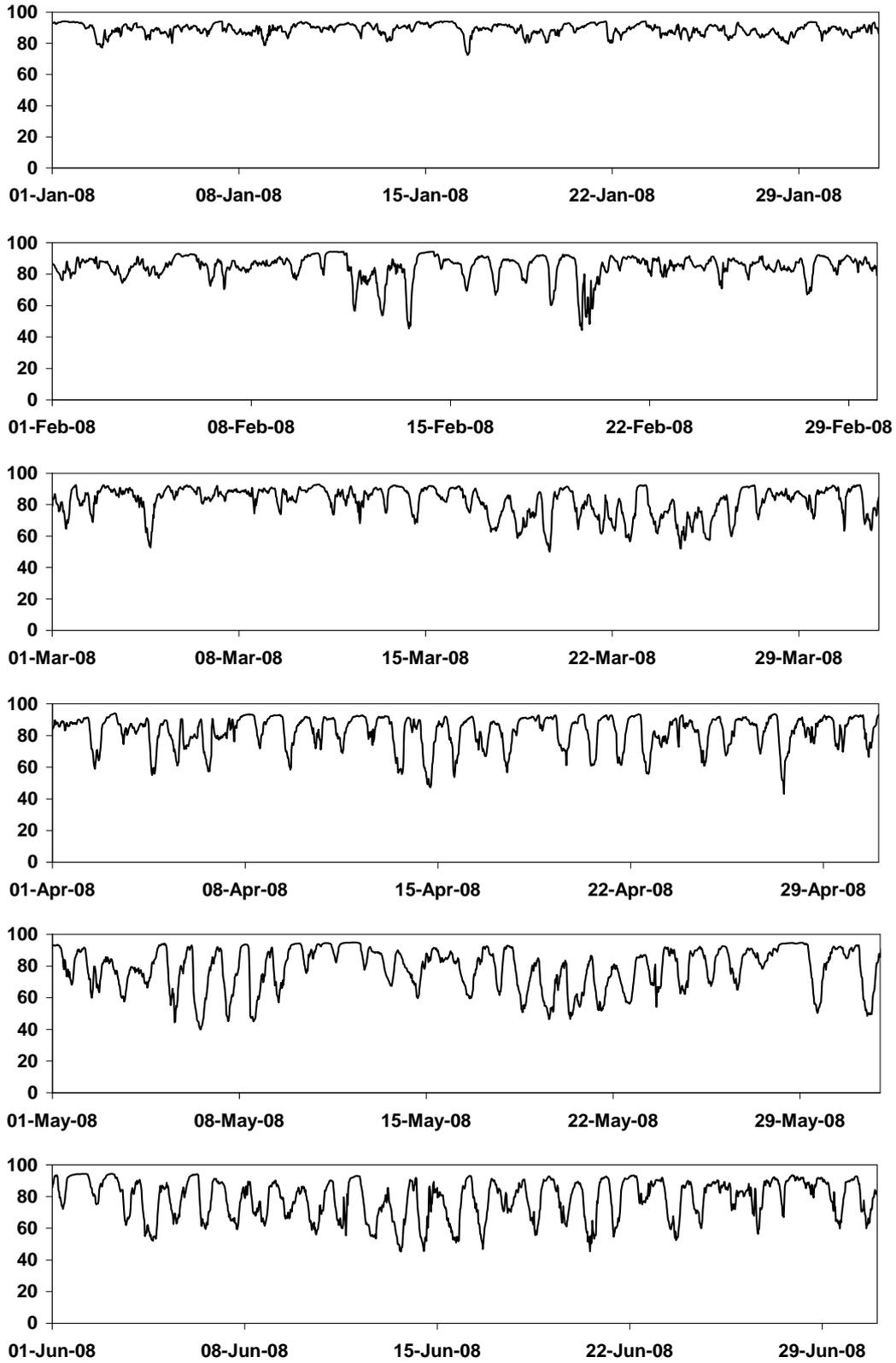
Solar radiation ($W m^{-2}$) and PAR ($\mu mol m^{-2} s^{-1}$)



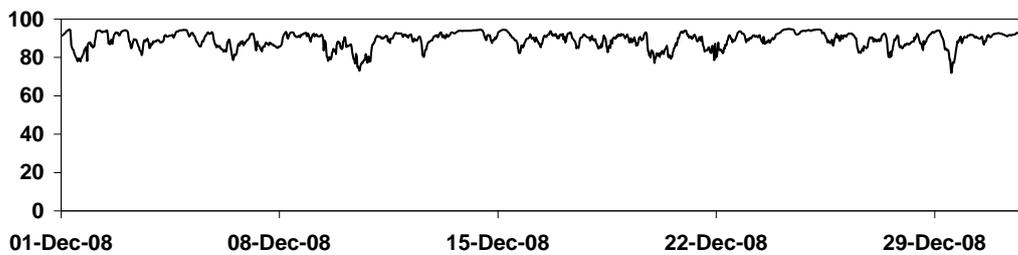
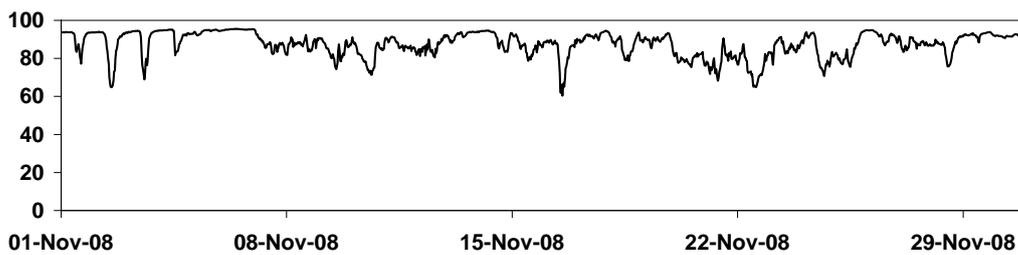
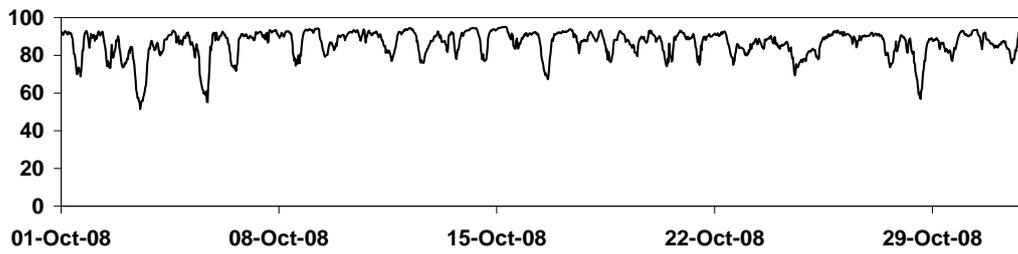
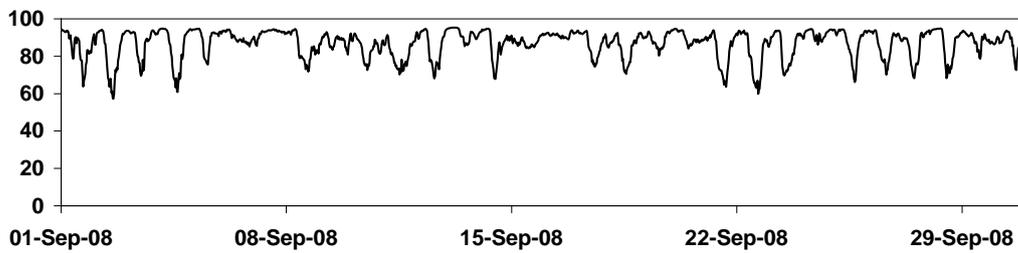
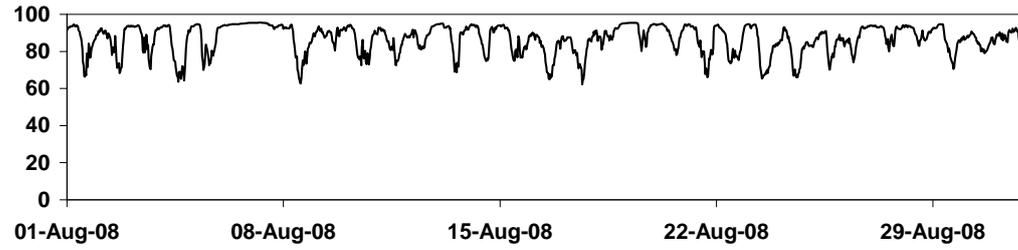
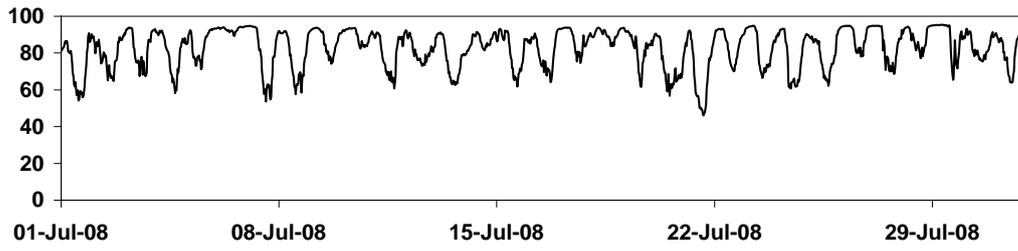
Solar radiation (W m^{-2}) and PAR ($\mu\text{mol m}^{-2}\text{s}^{-1}$)



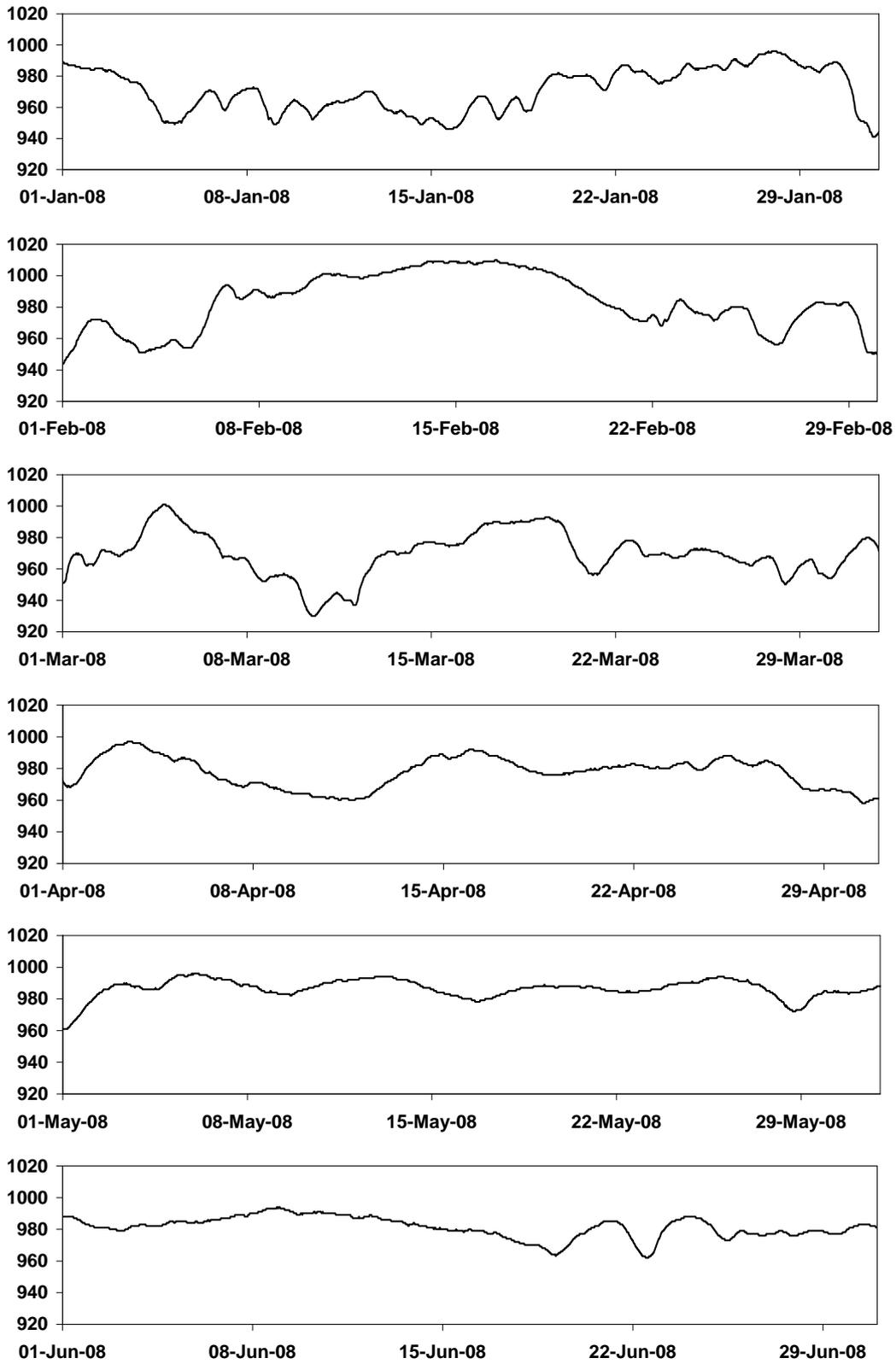
Relative humidity (%)



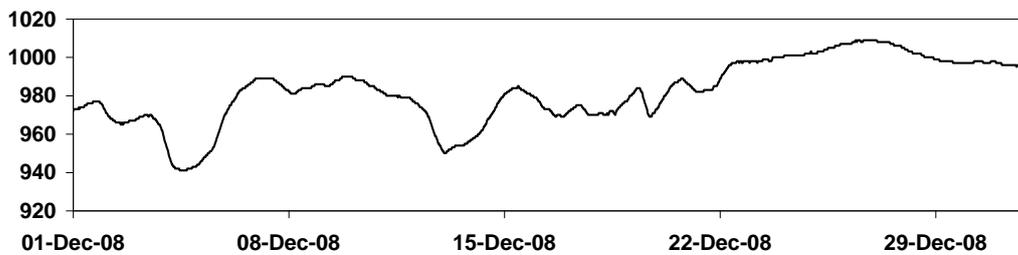
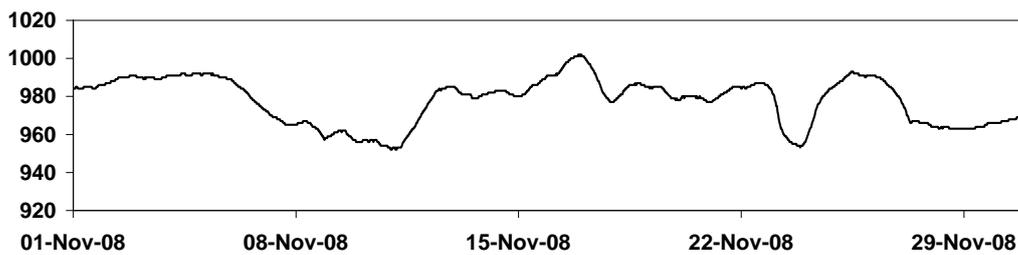
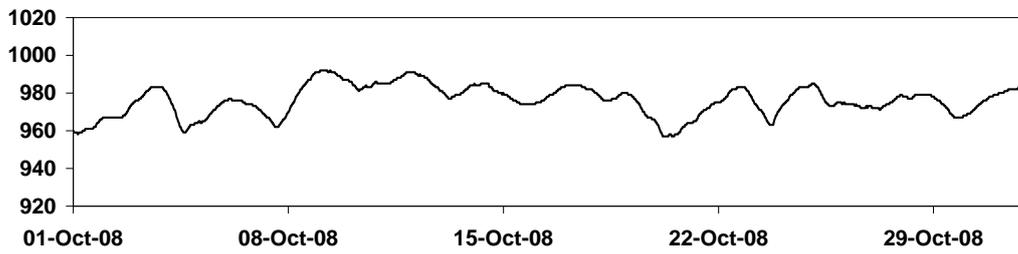
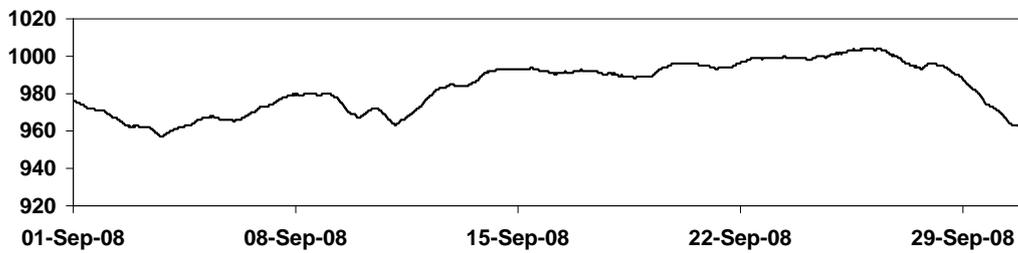
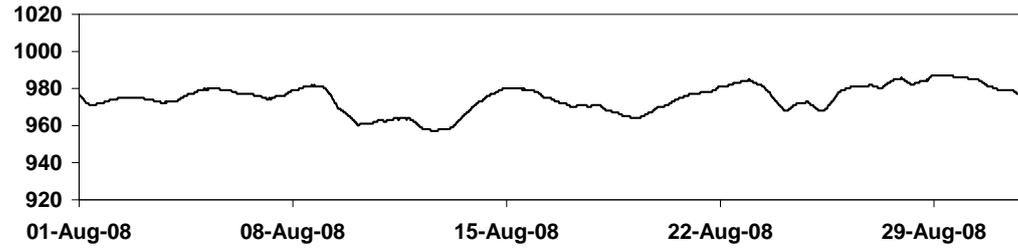
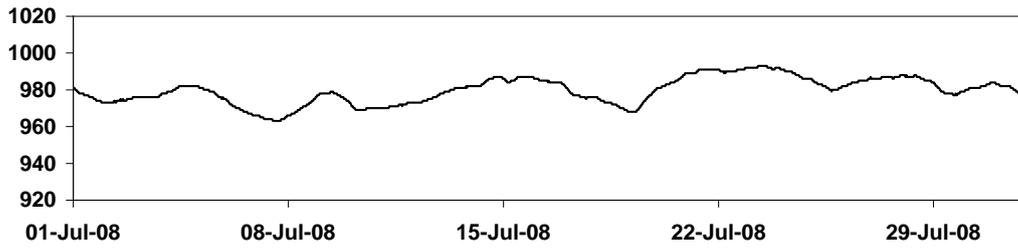
Relative humidity (%)



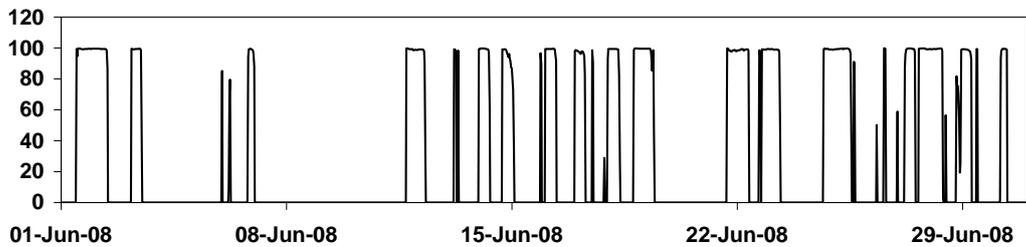
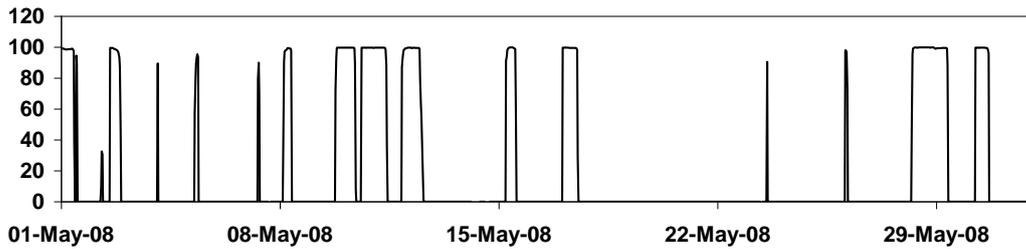
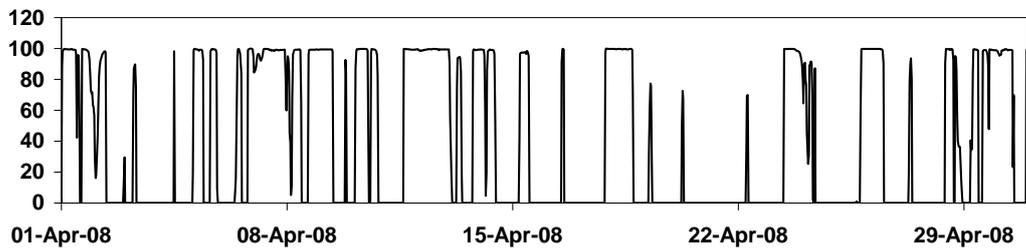
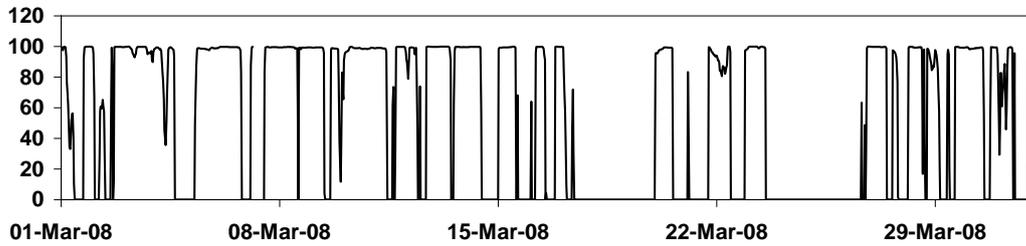
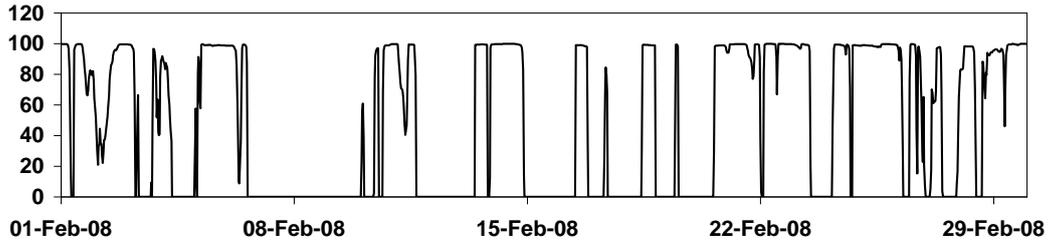
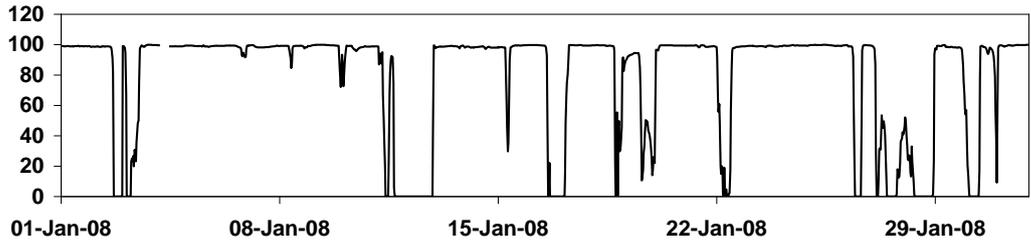
Atmospheric pressure mbar



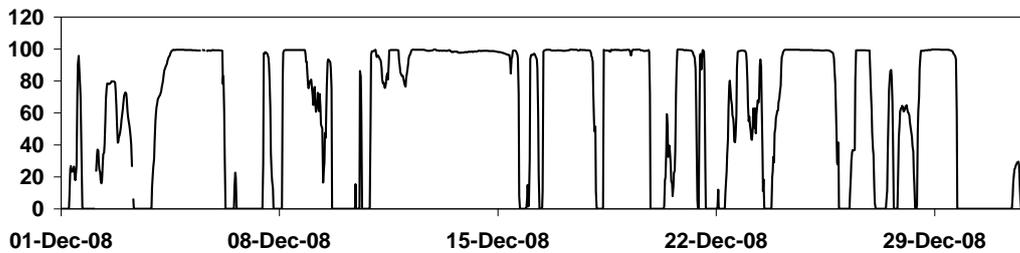
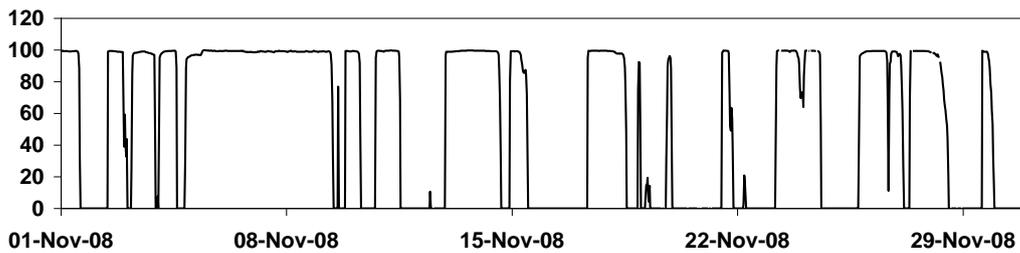
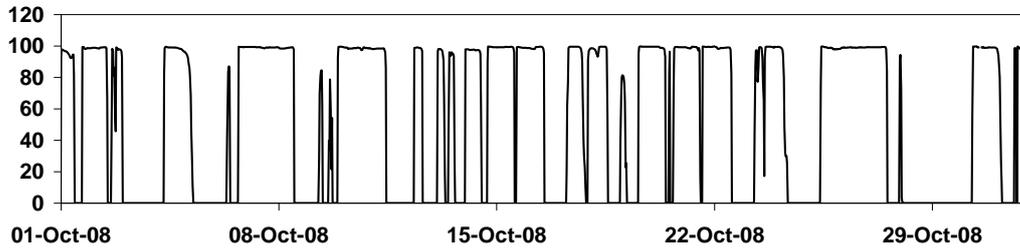
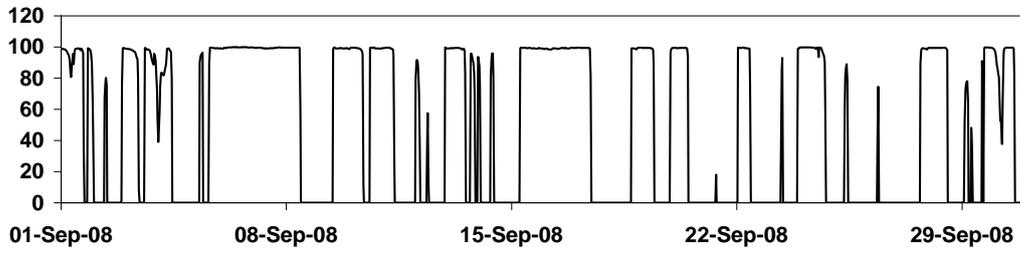
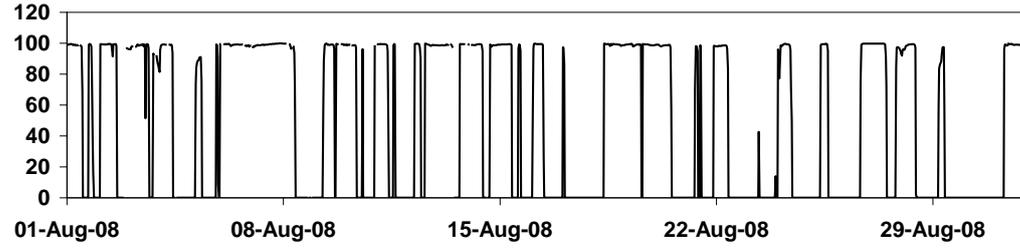
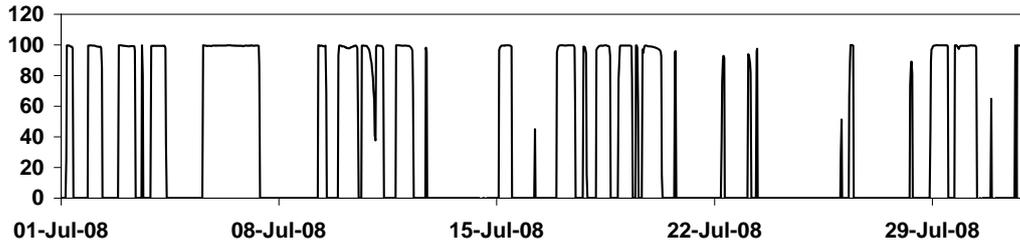
Atmospheric pressure mbar



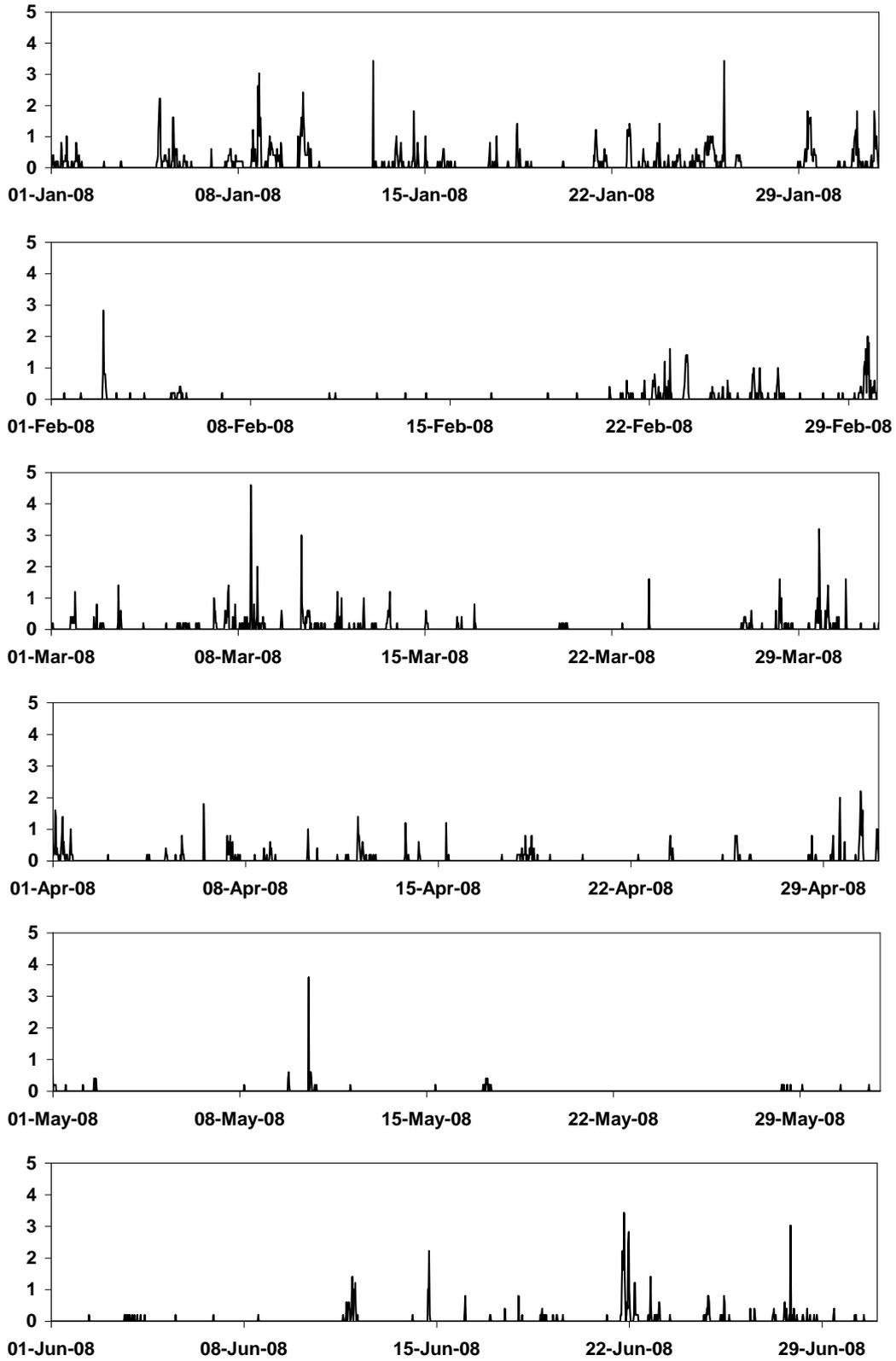
Wetness %



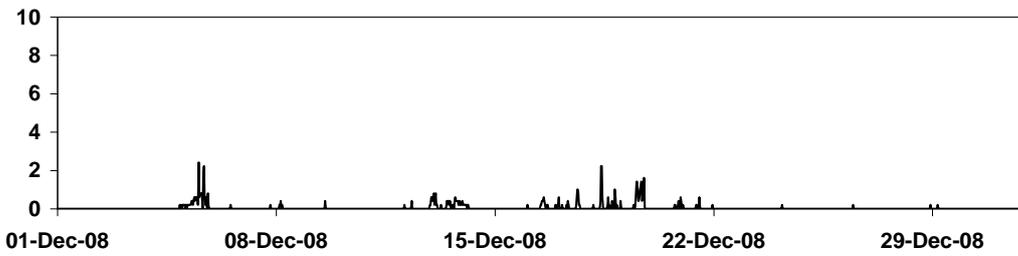
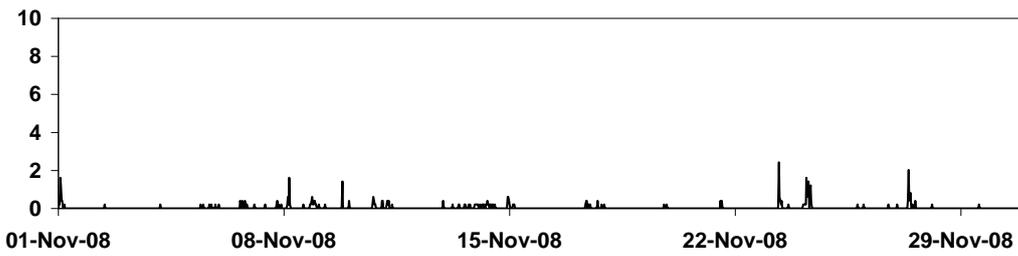
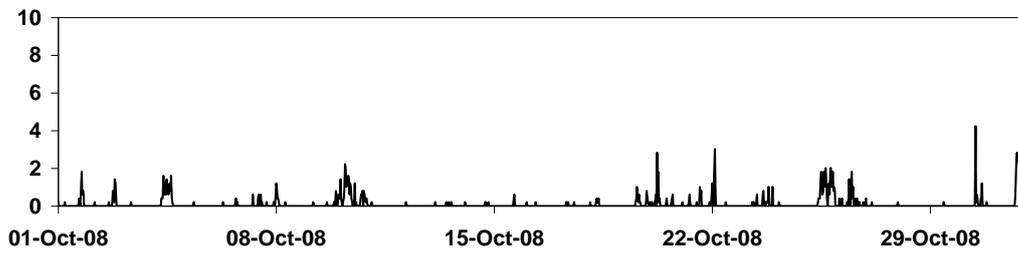
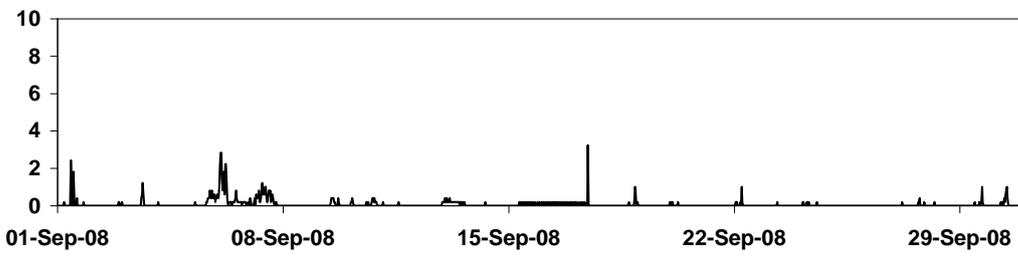
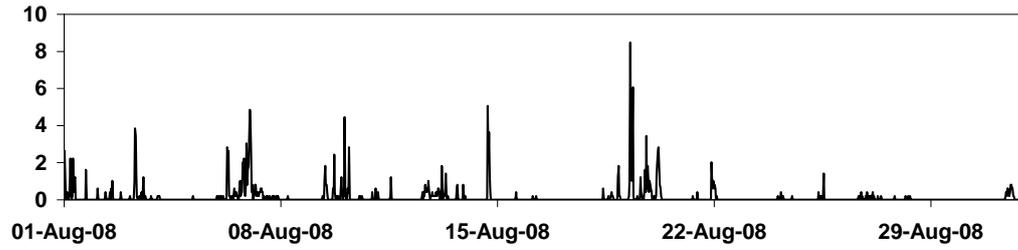
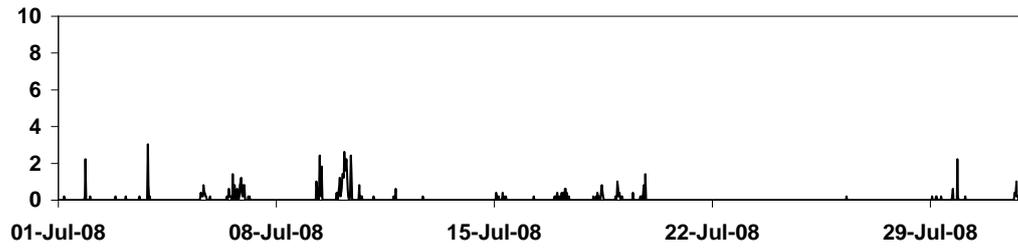
Wetness %



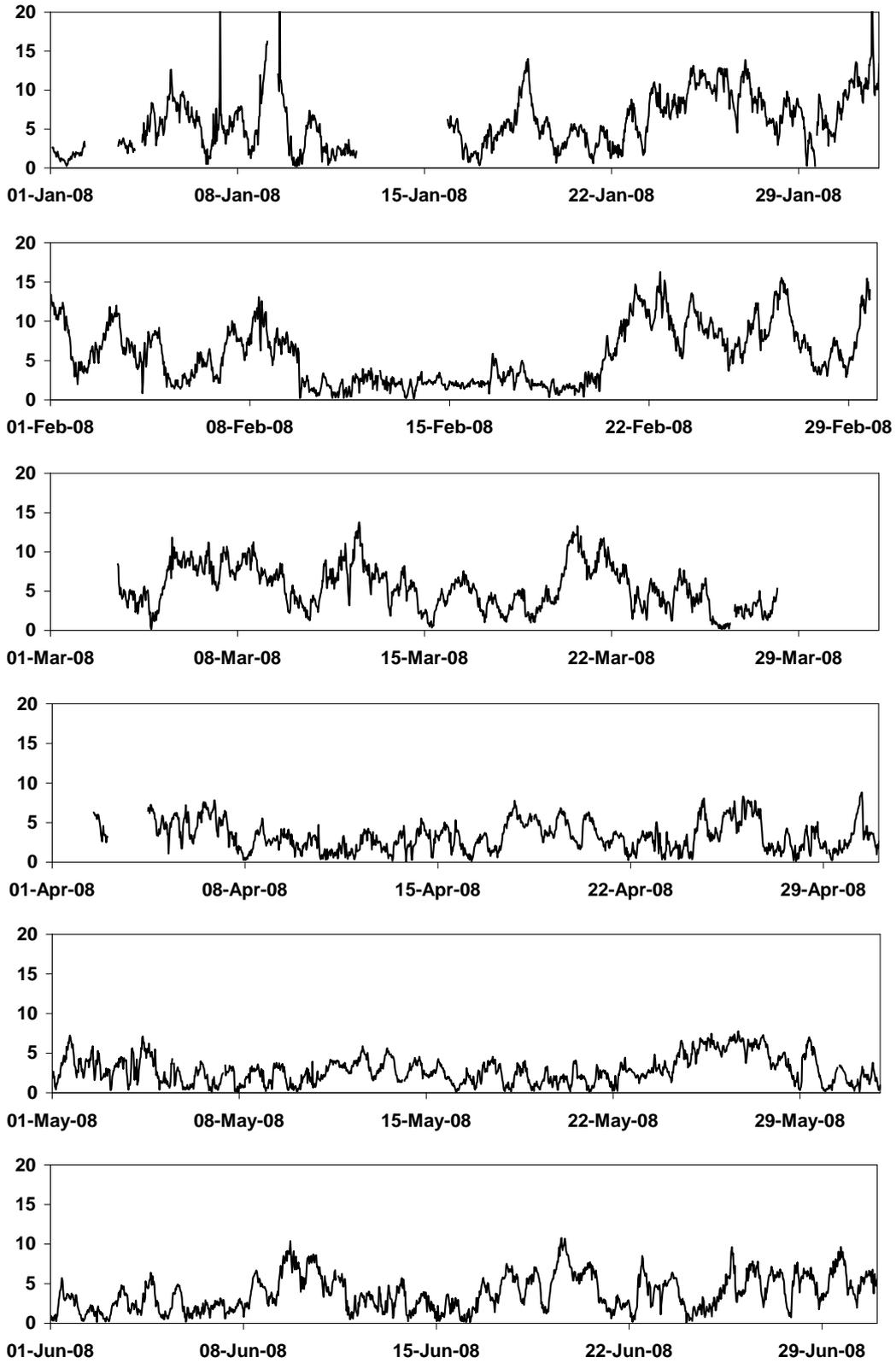
30-minute rainfall mm



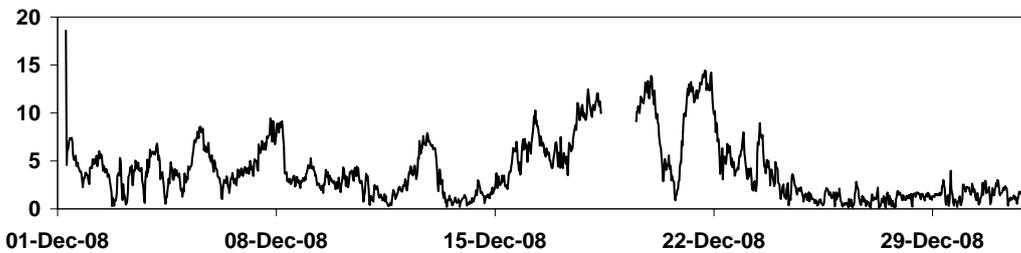
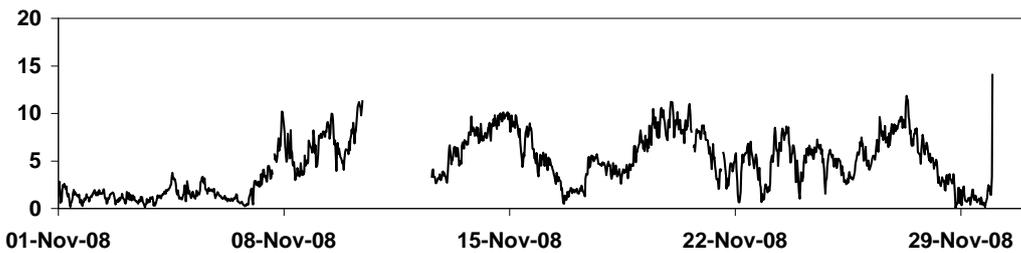
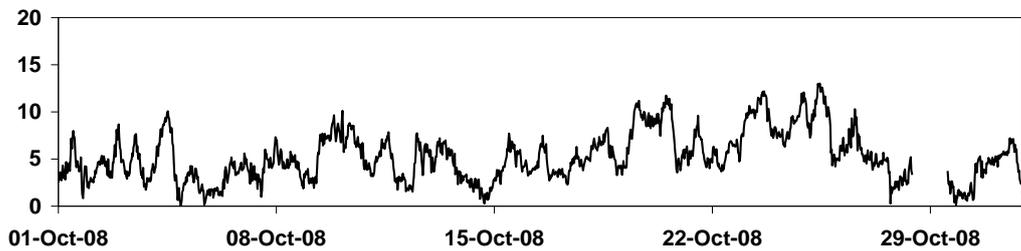
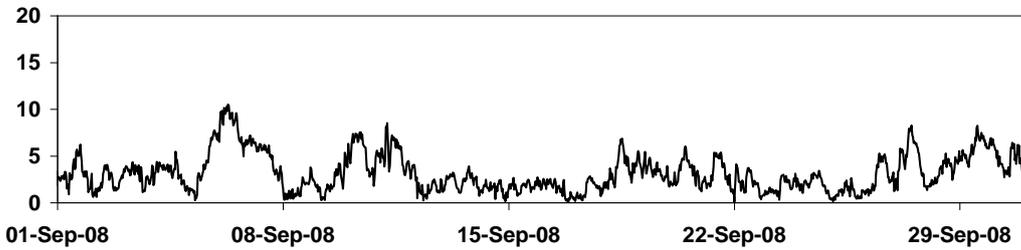
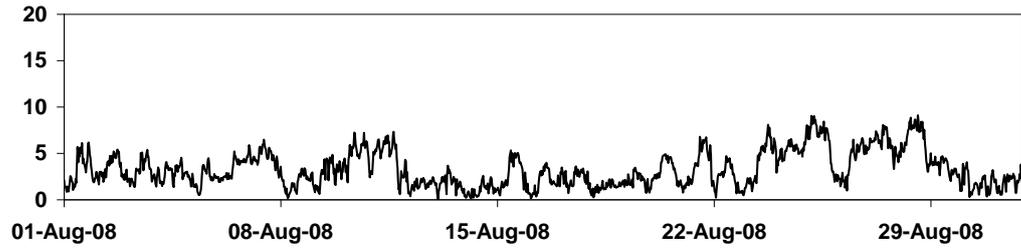
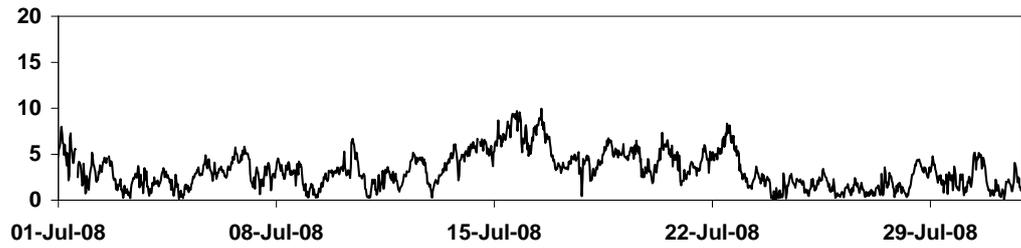
30-minute rainfall mm



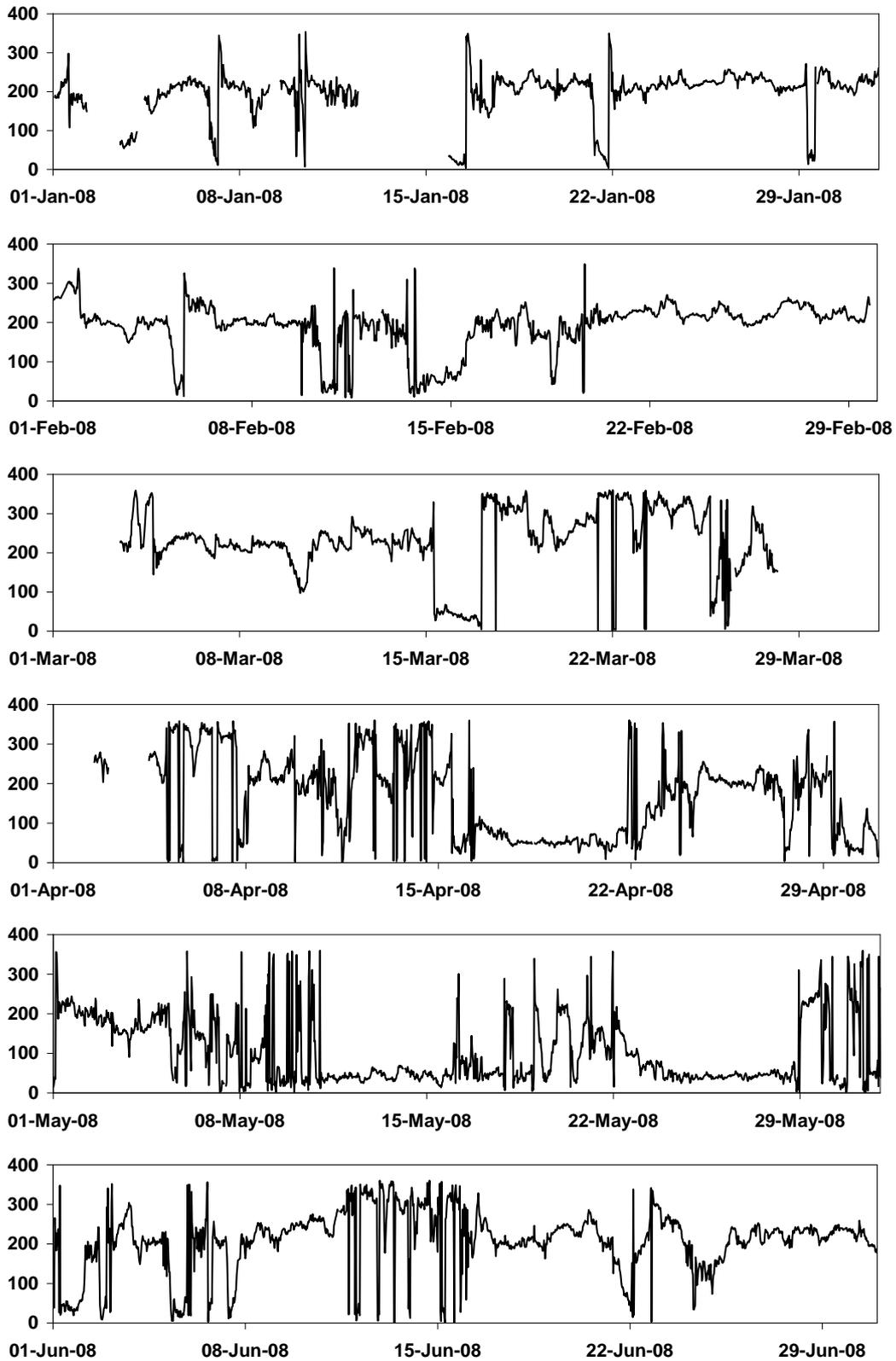
wind speed m s⁻¹



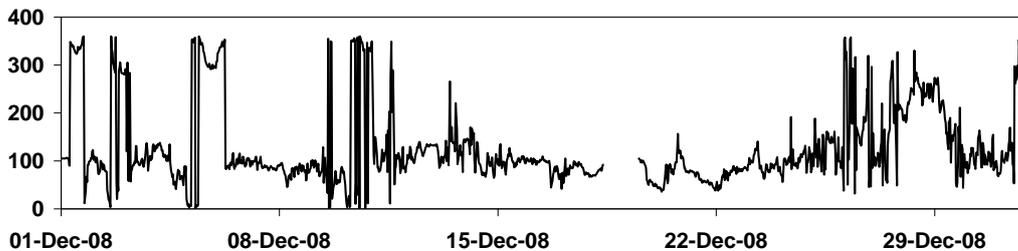
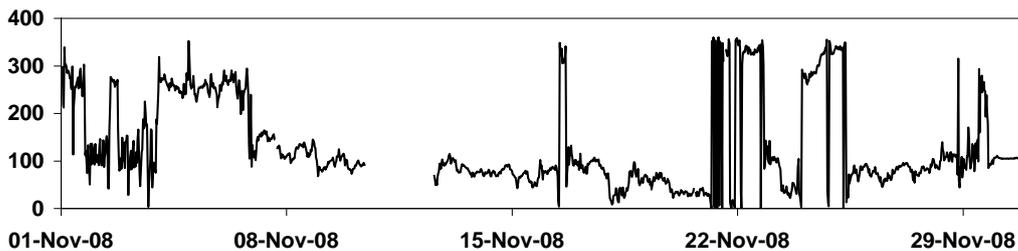
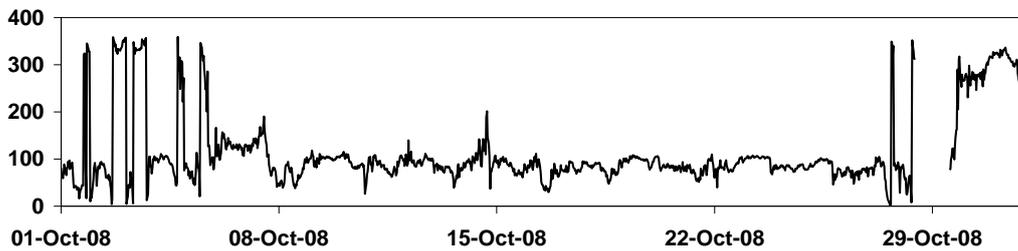
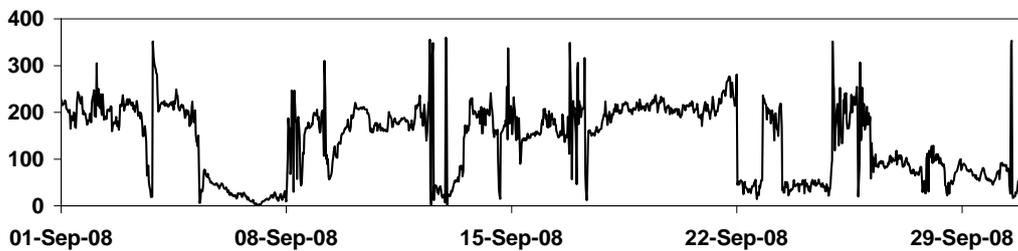
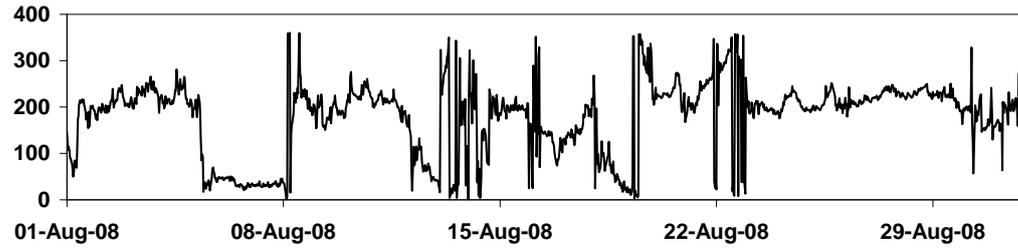
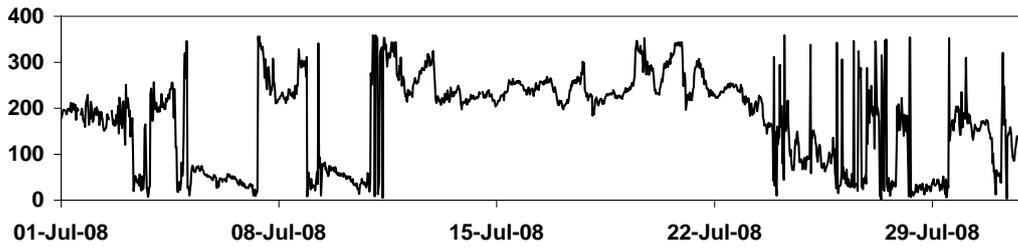
wind speed m s^{-1}



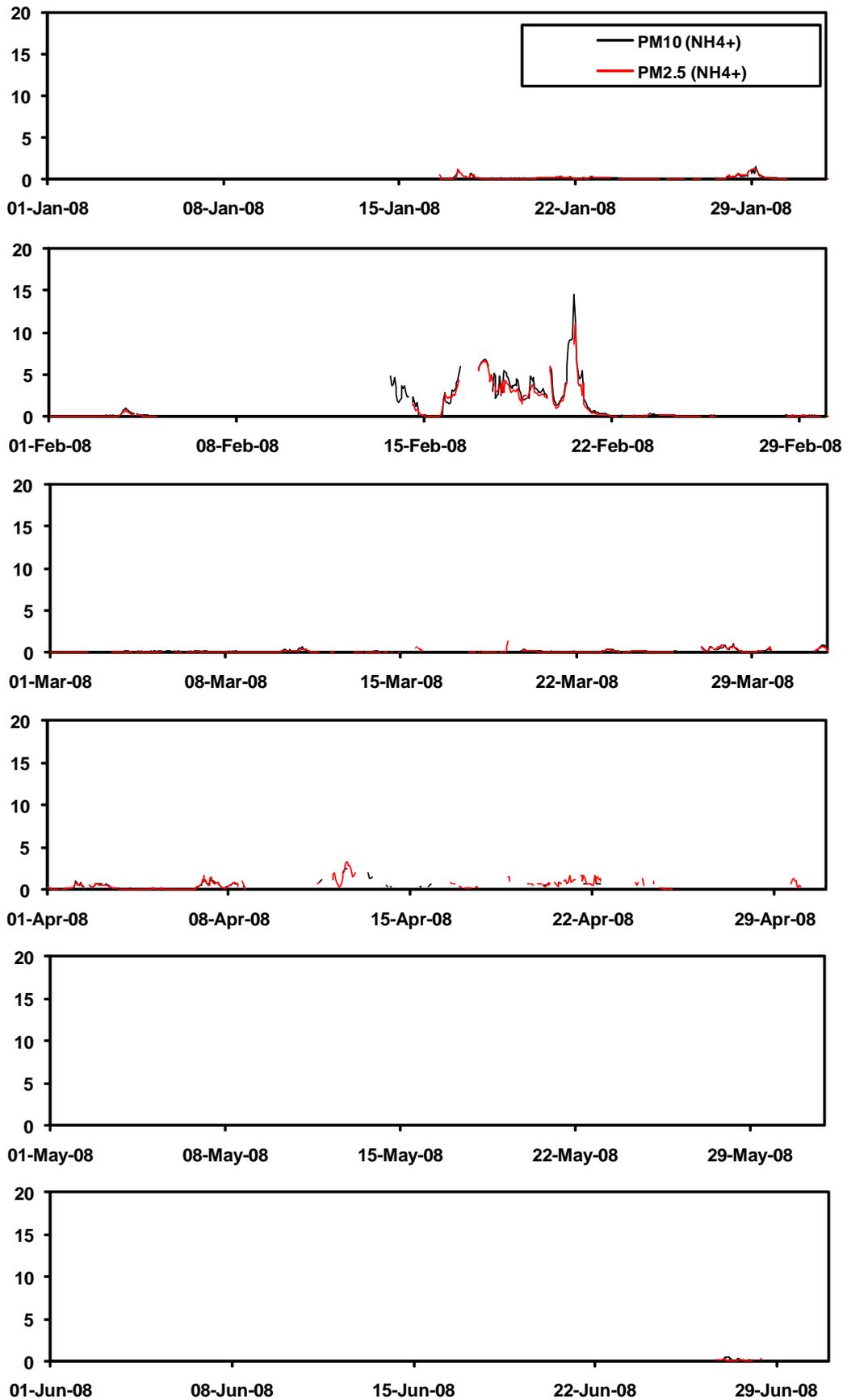
wind direction degrees



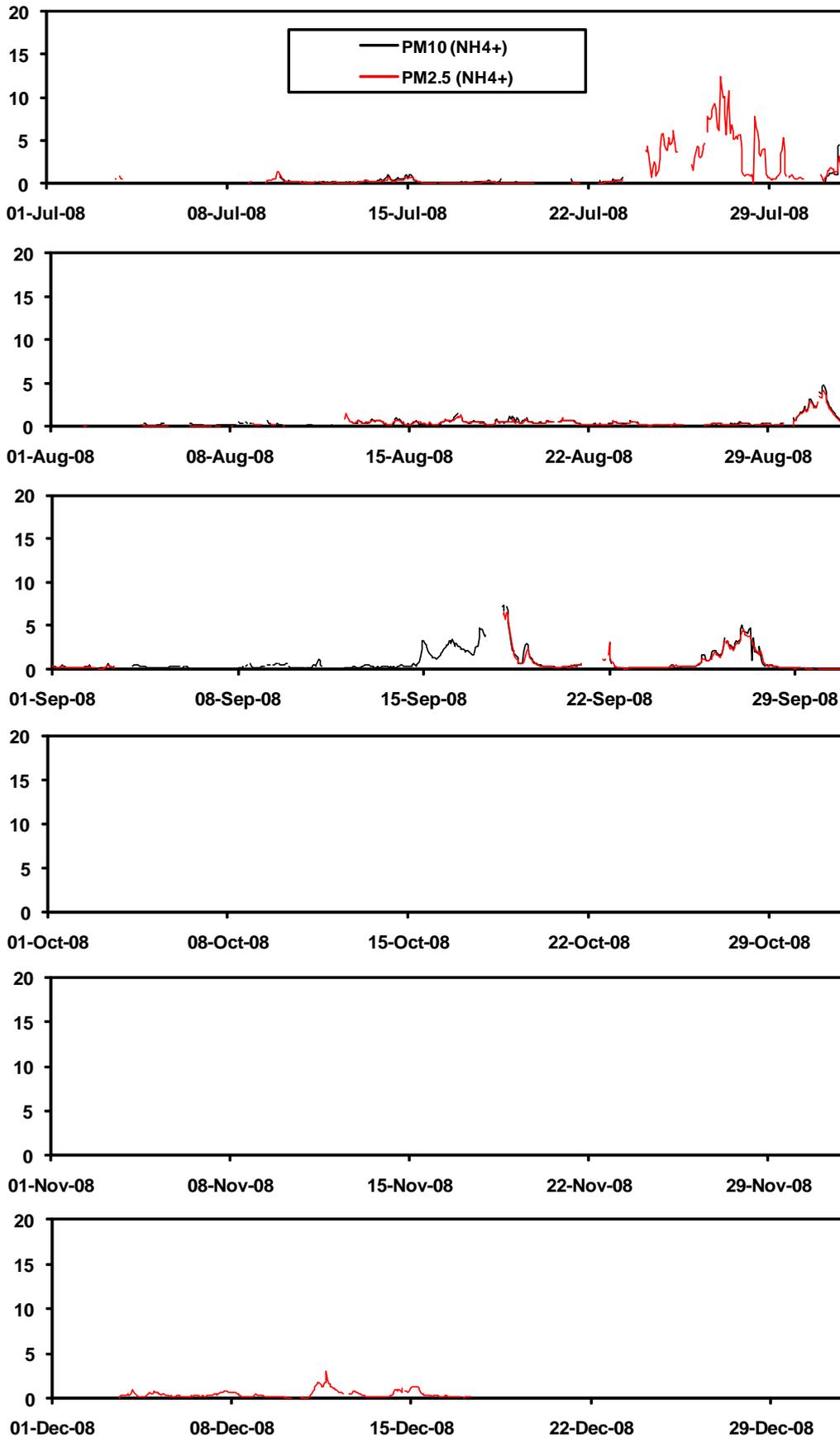
wind direction degrees



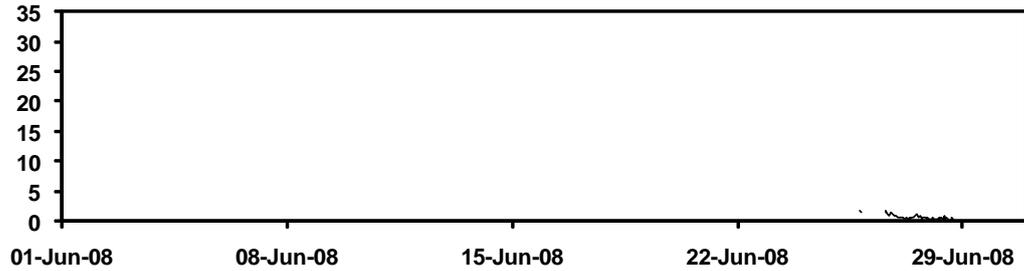
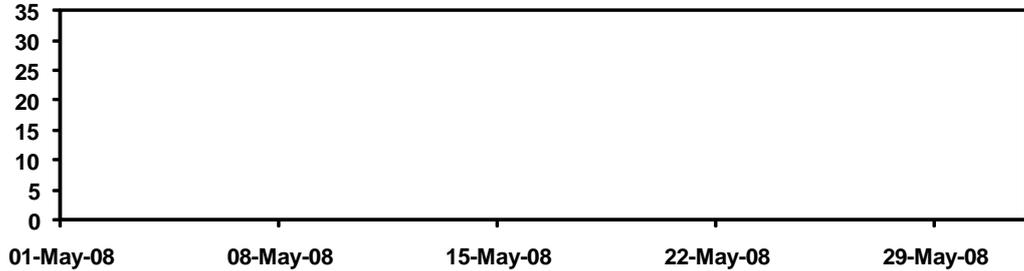
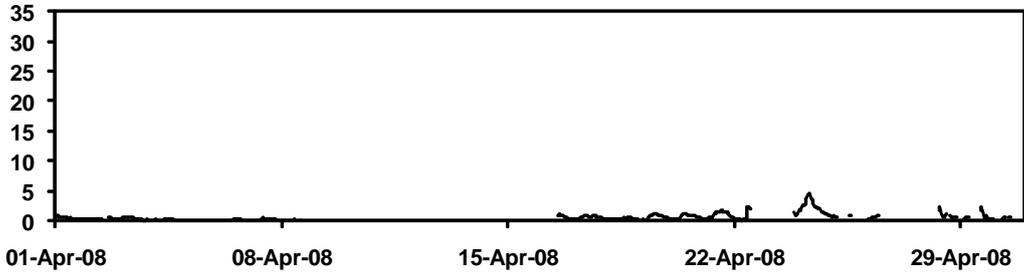
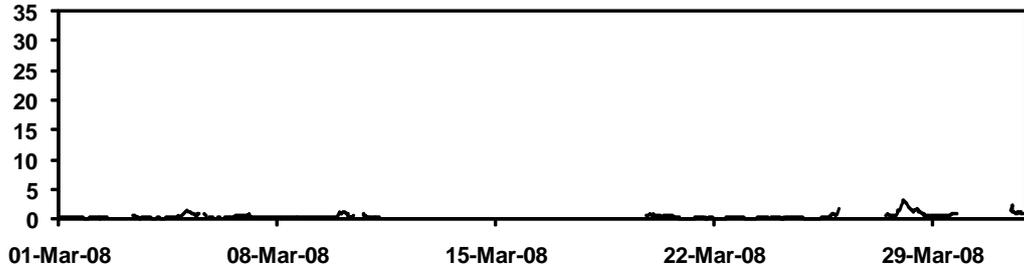
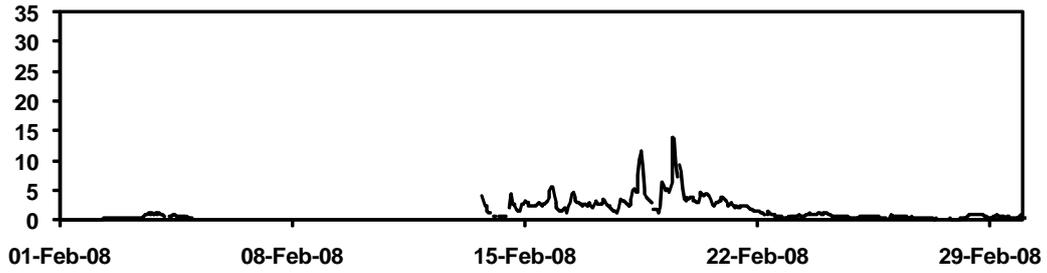
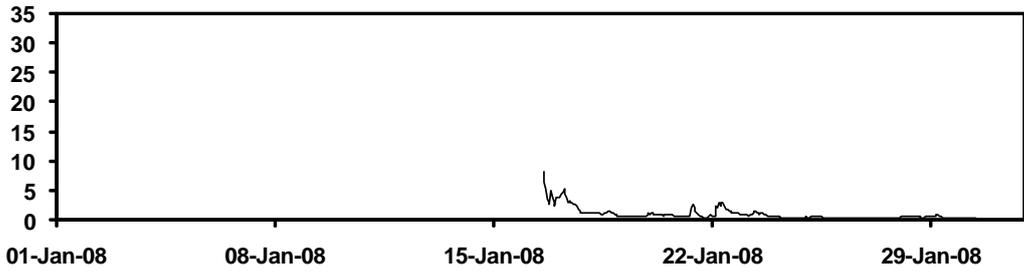
Ammonium aerosol $\mu\text{g m}^{-3}$



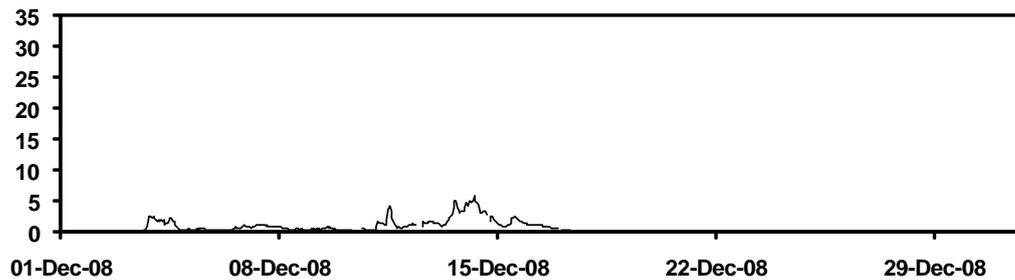
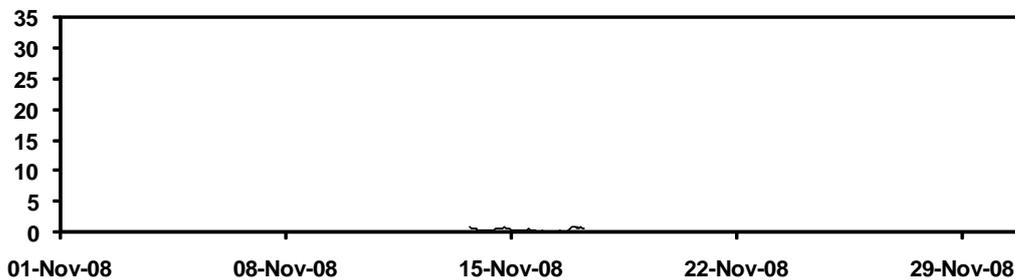
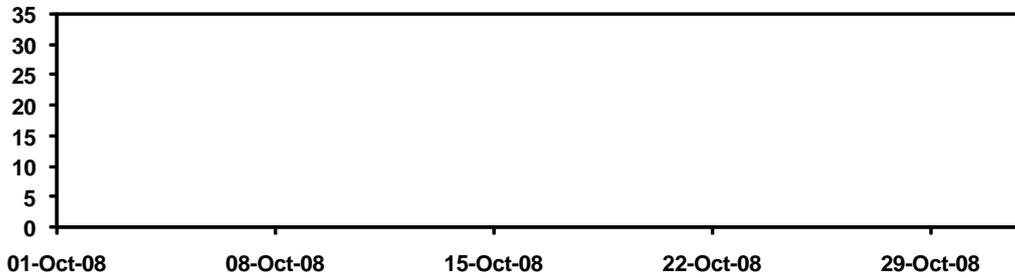
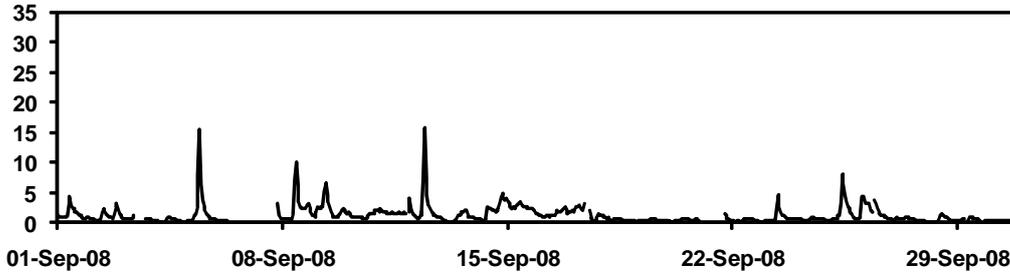
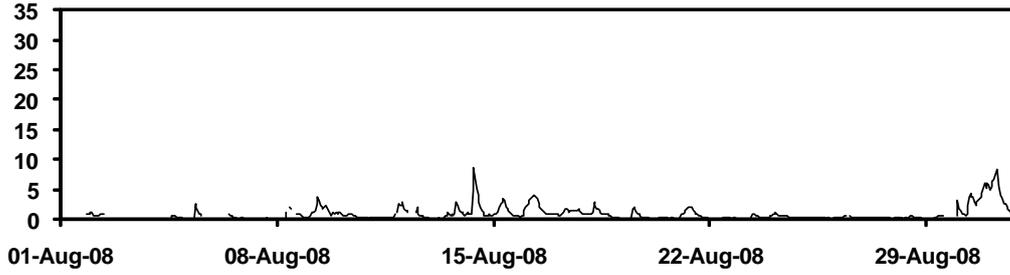
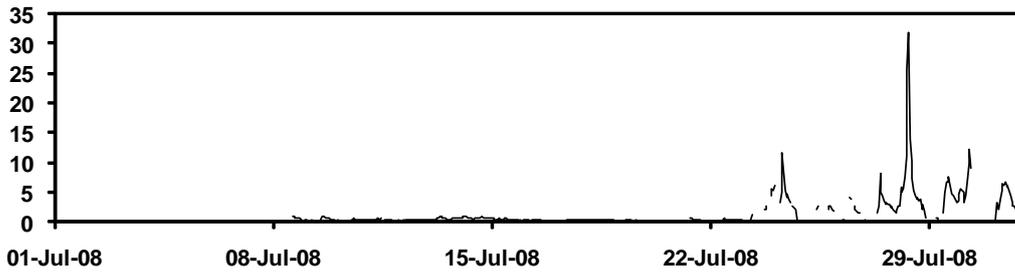
Ammonium aerosol $\mu\text{g m}^{-3}$



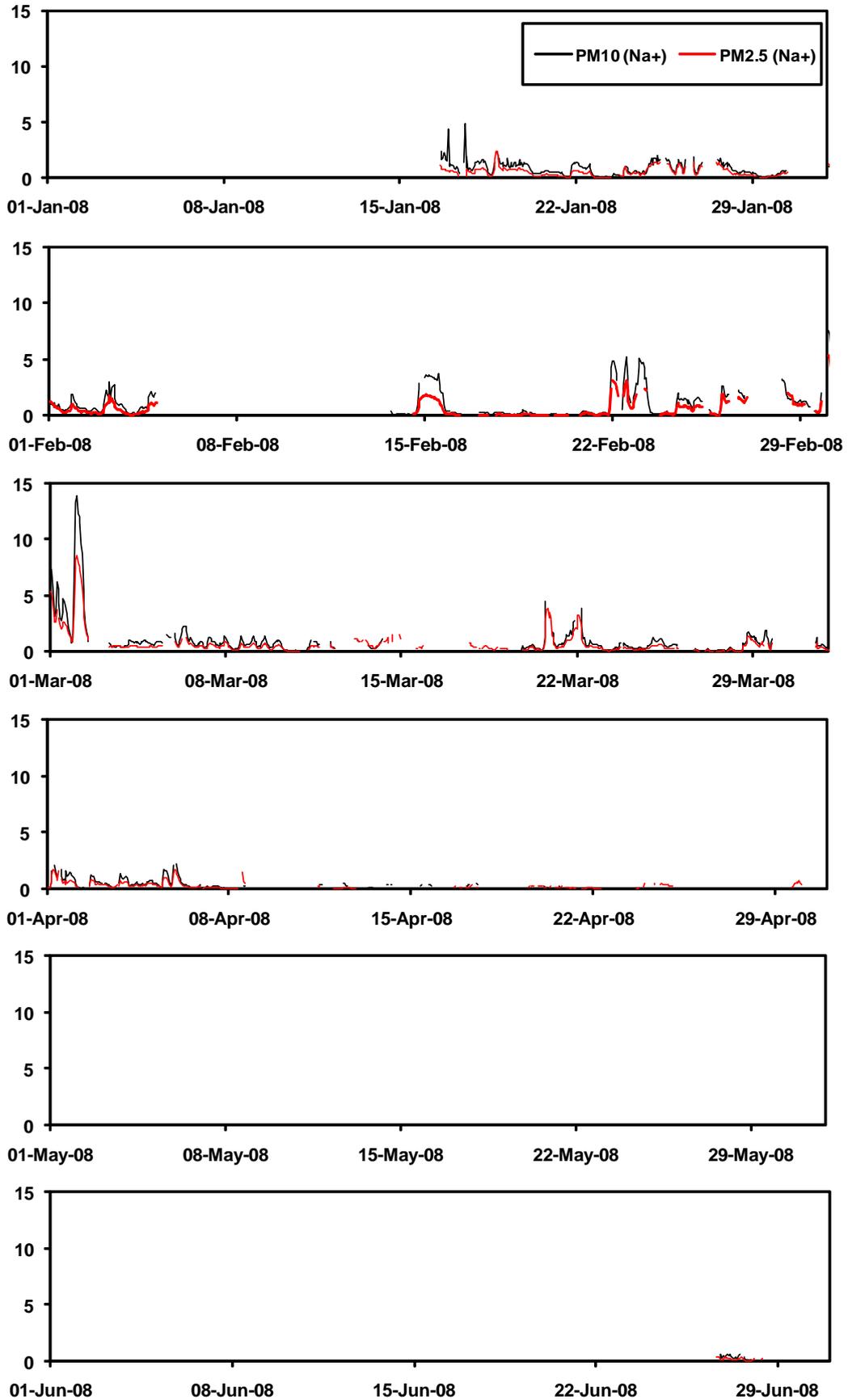
Ammonia $\mu\text{g m}^{-3}$



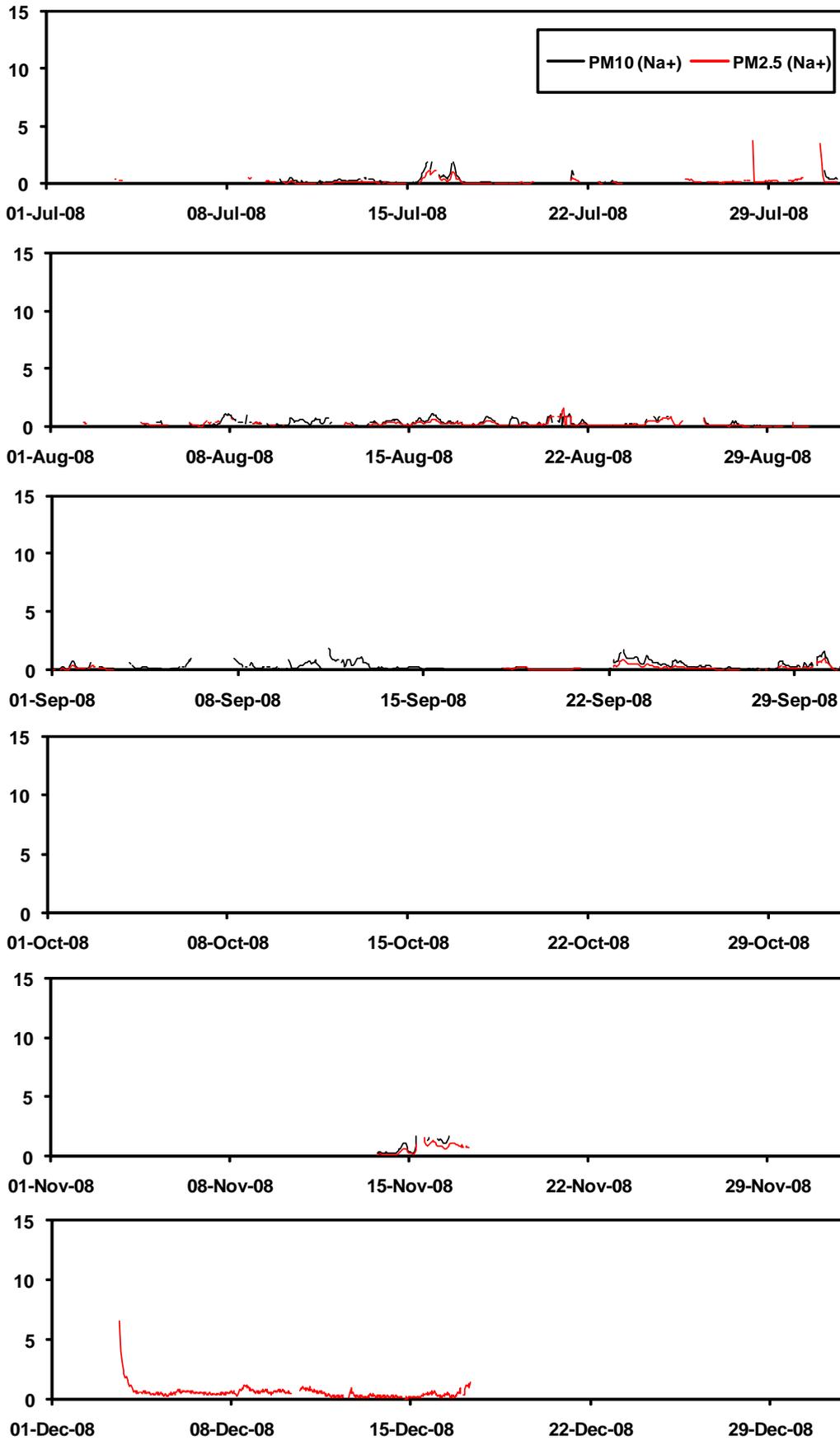
Ammonia $\mu\text{g m}^{-3}$



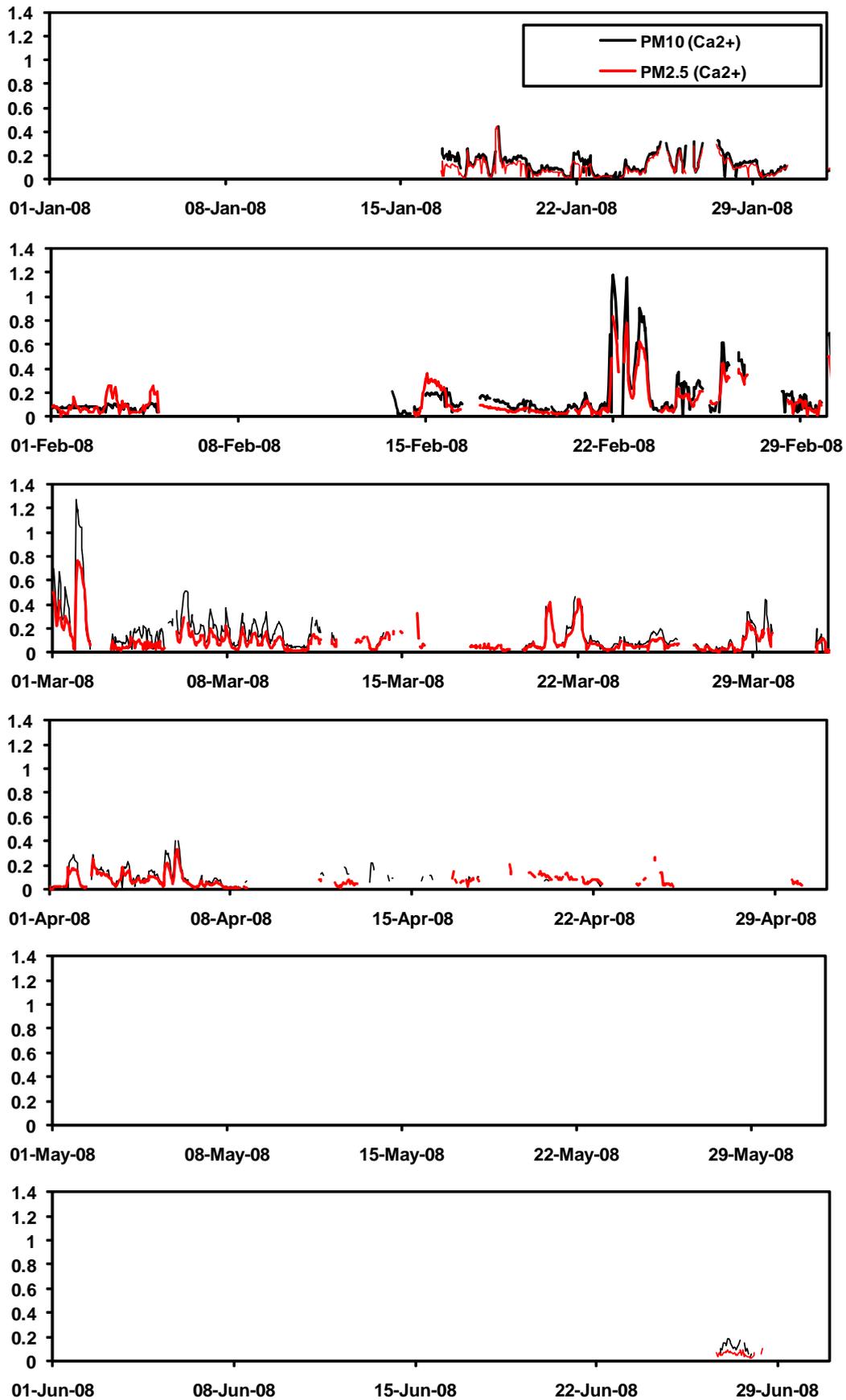
Sodium aerosol $\mu\text{g m}^{-3}$



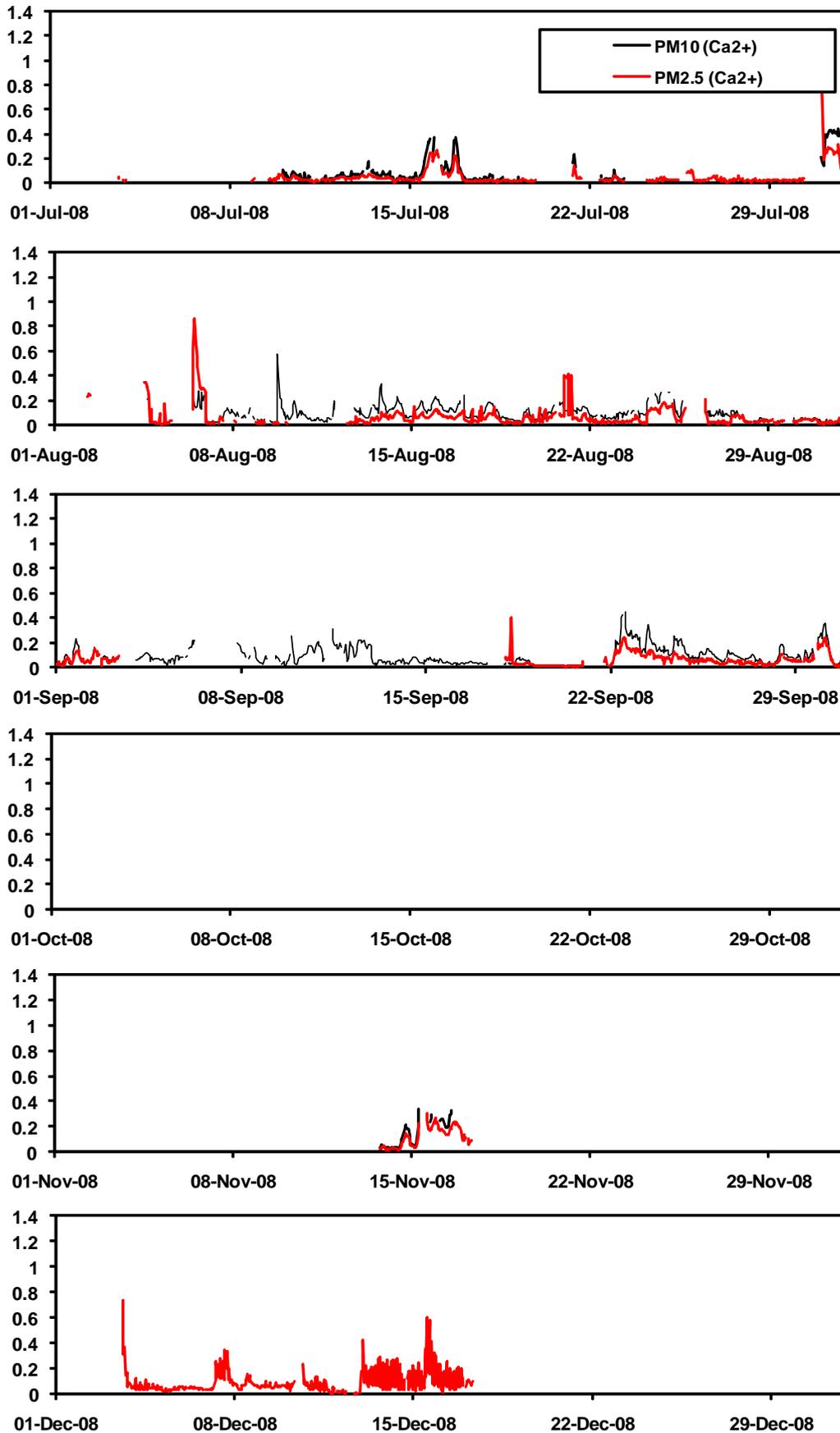
Sodium aerosol $\mu\text{g m}^{-3}$



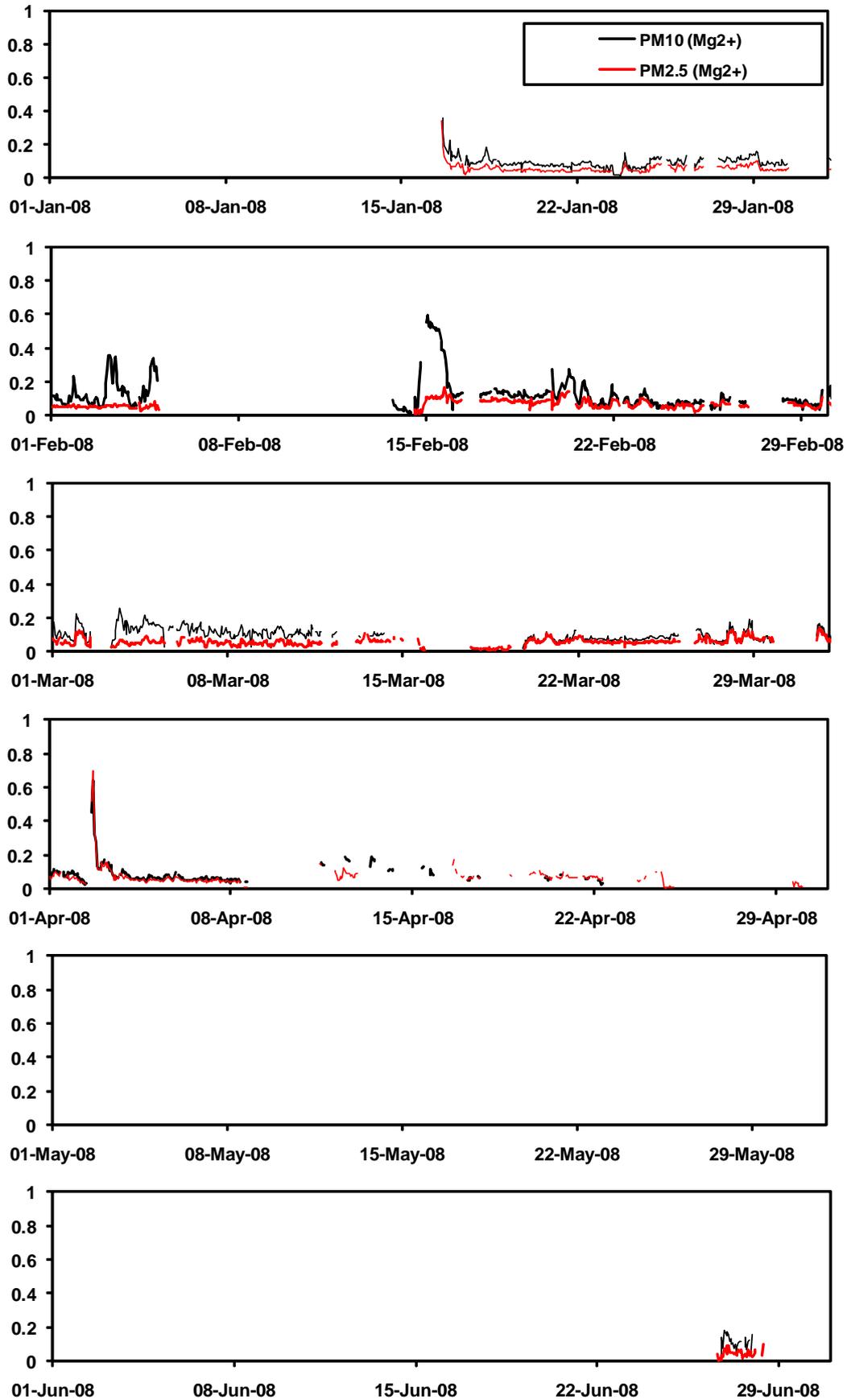
Calcium aerosol $\mu\text{g m}^{-3}$



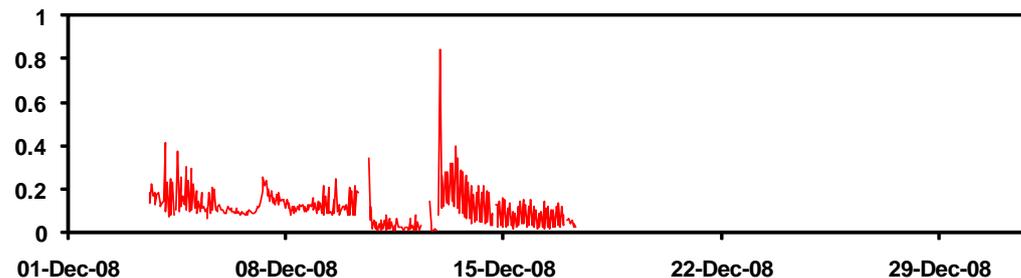
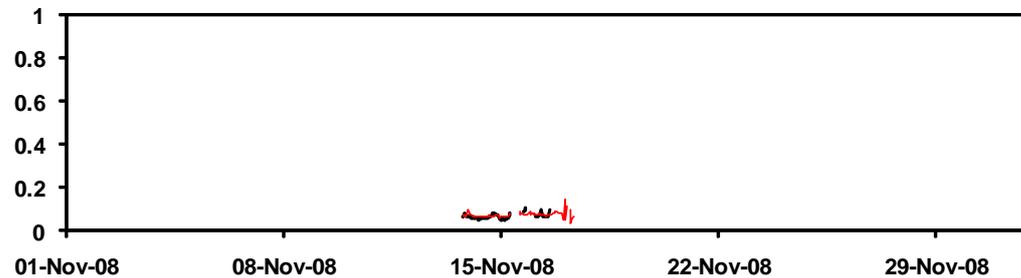
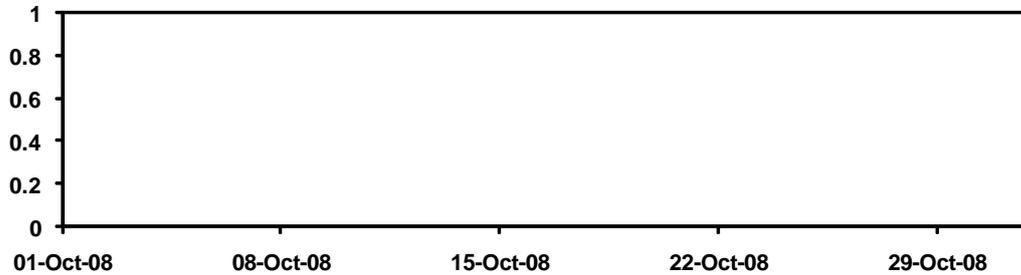
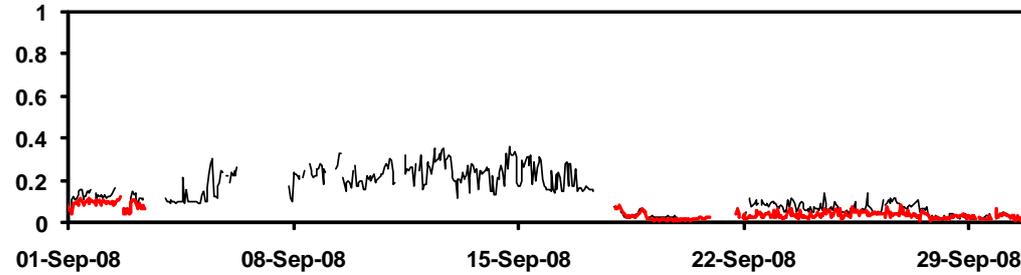
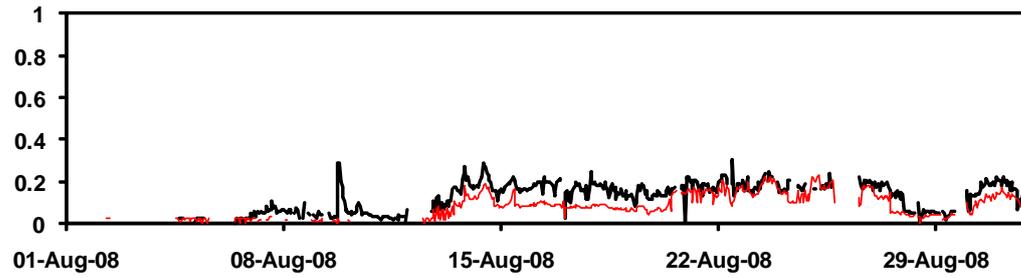
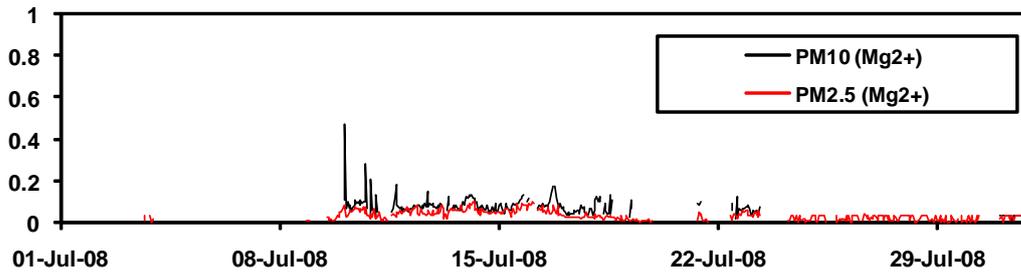
Calcium aerosol $\mu\text{g m}^{-3}$



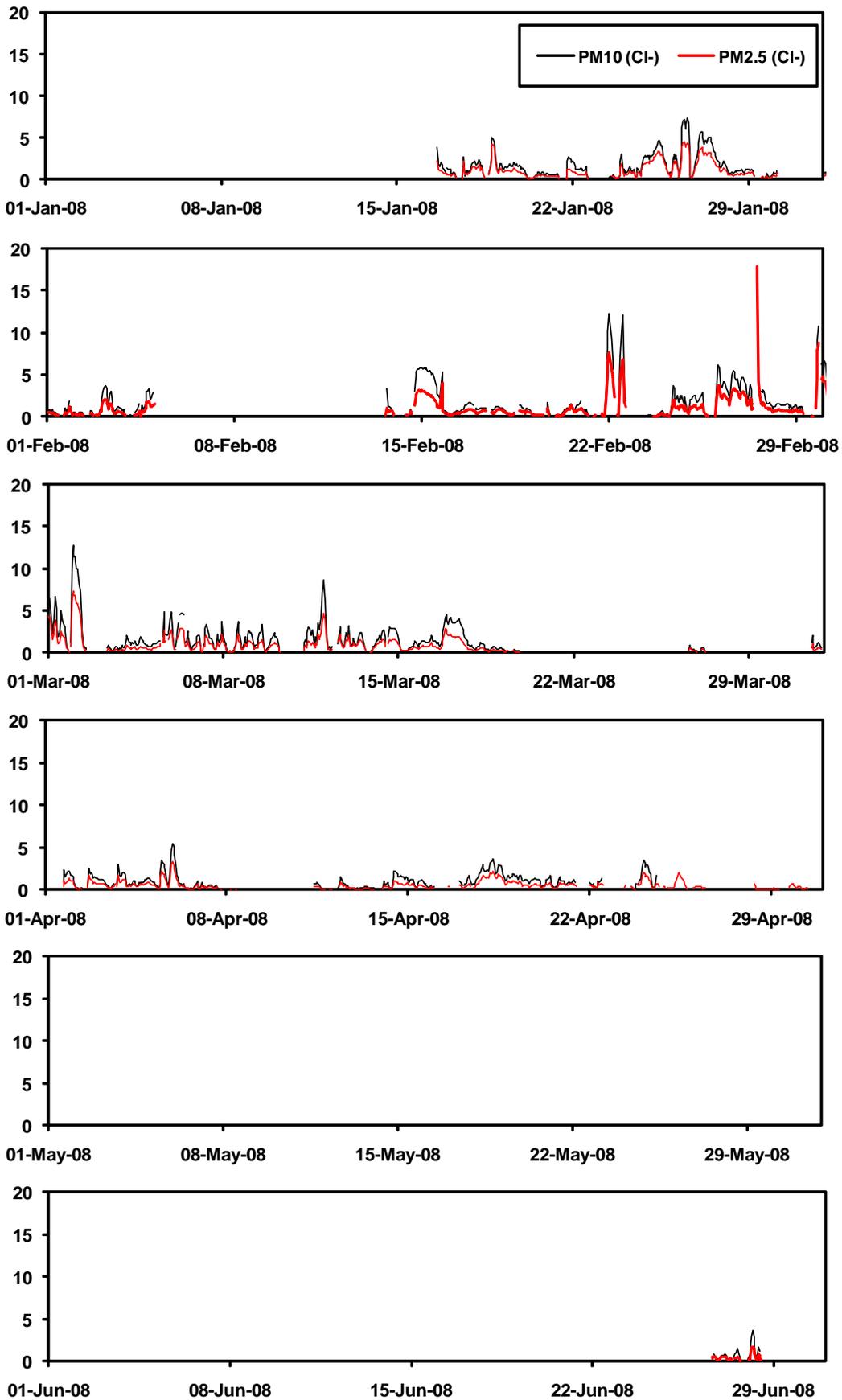
Magnesium aerosol $\mu\text{g m}^{-3}$



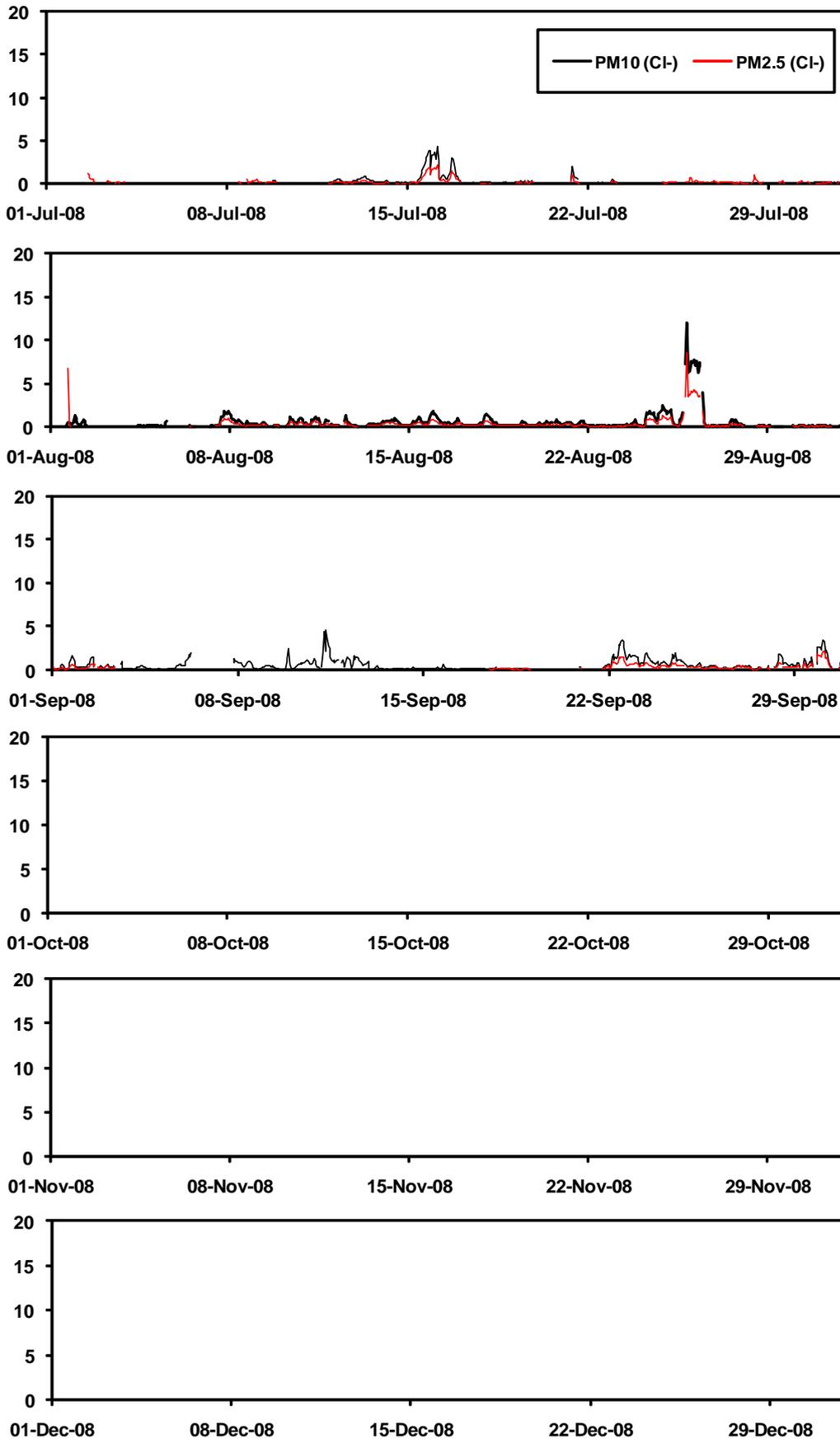
Magnesium aerosol $\mu\text{g m}^{-3}$



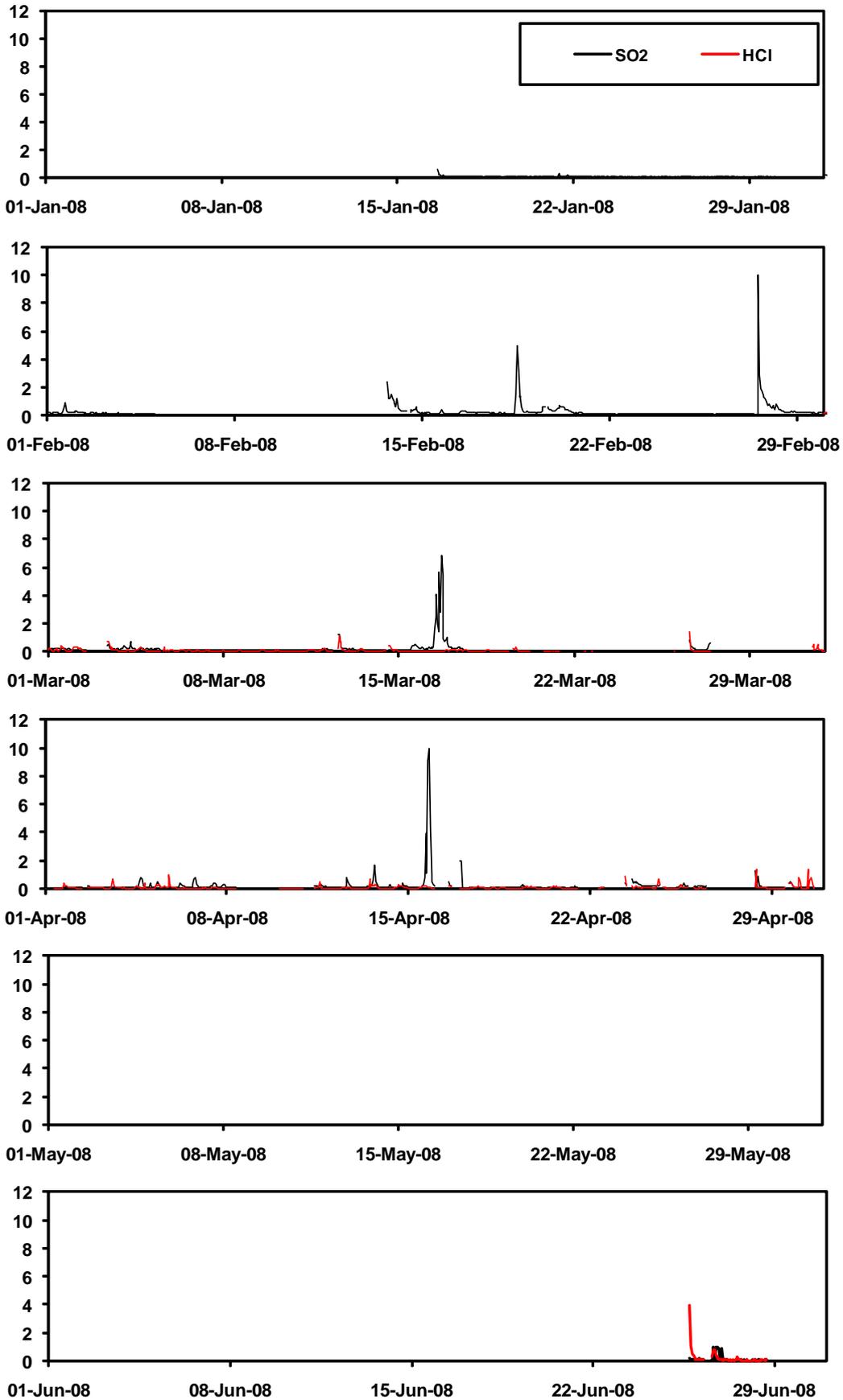
Chloride aerosol $\mu\text{g m}^{-3}$



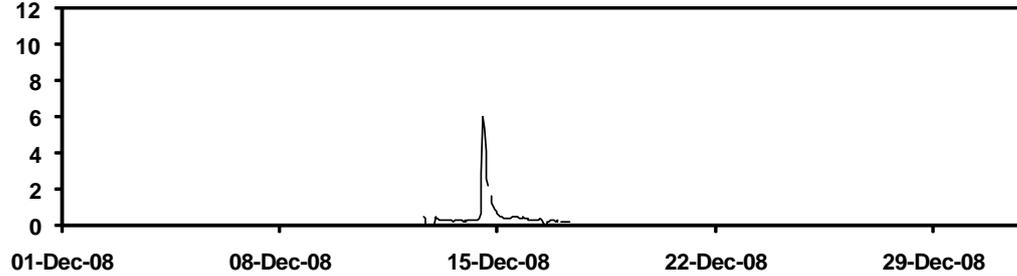
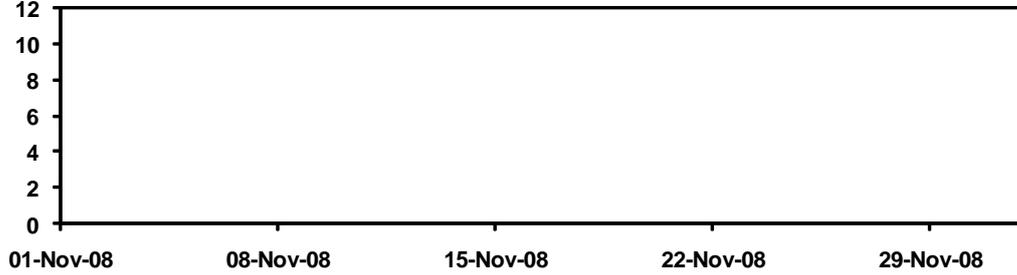
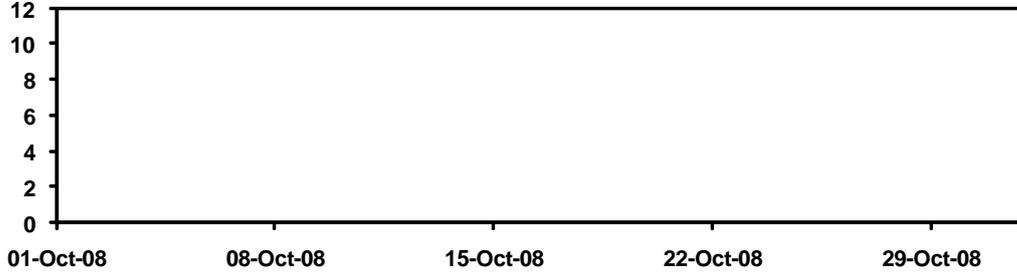
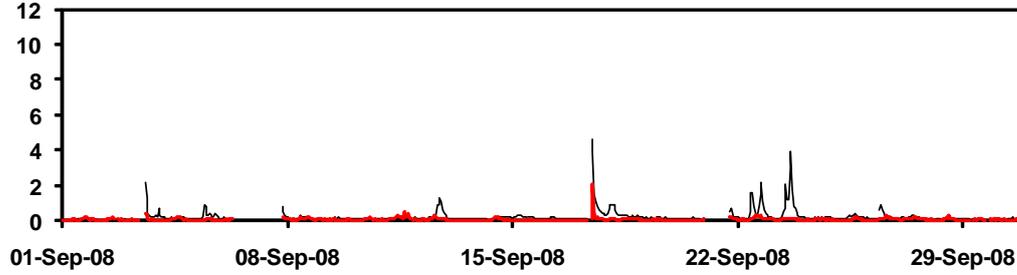
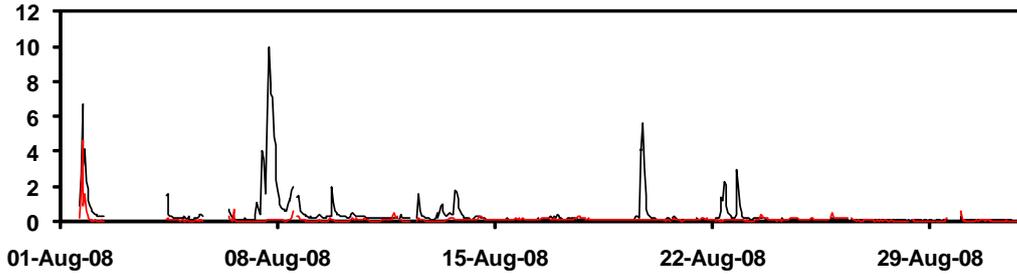
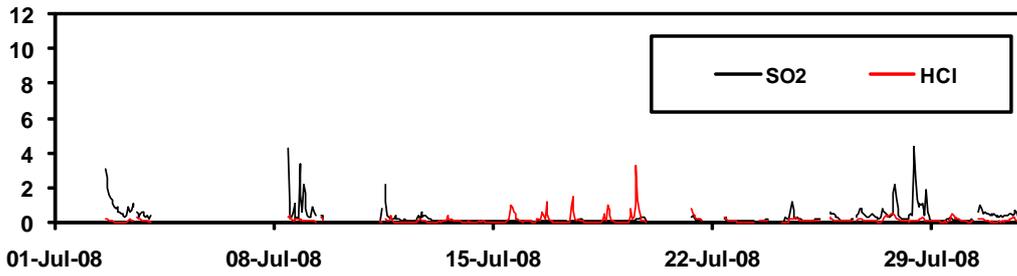
Chloride aerosol $\mu\text{g m}^{-3}$



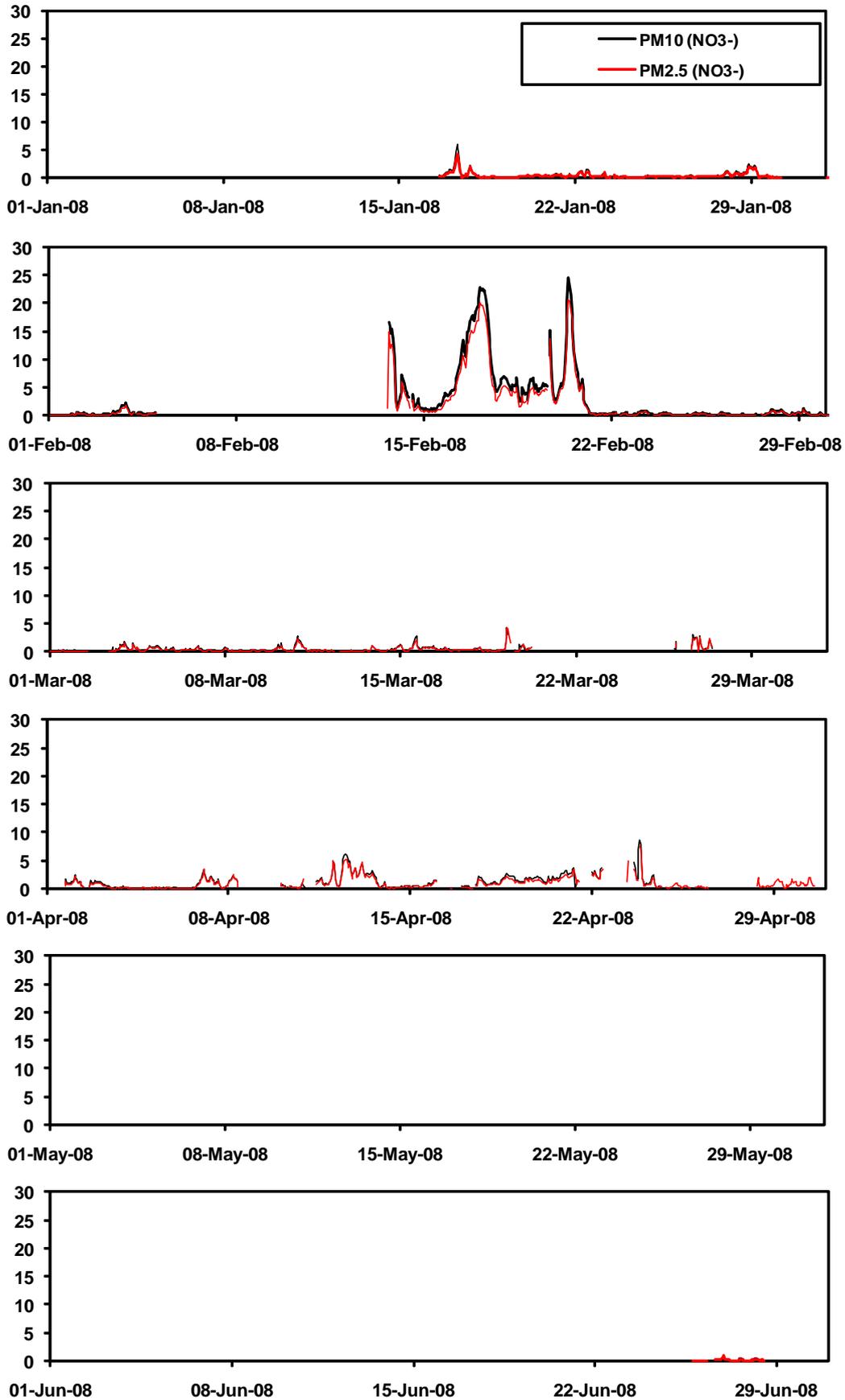
Sulphur dioxide/hydrogen chloride $\mu\text{g m}^{-3}$



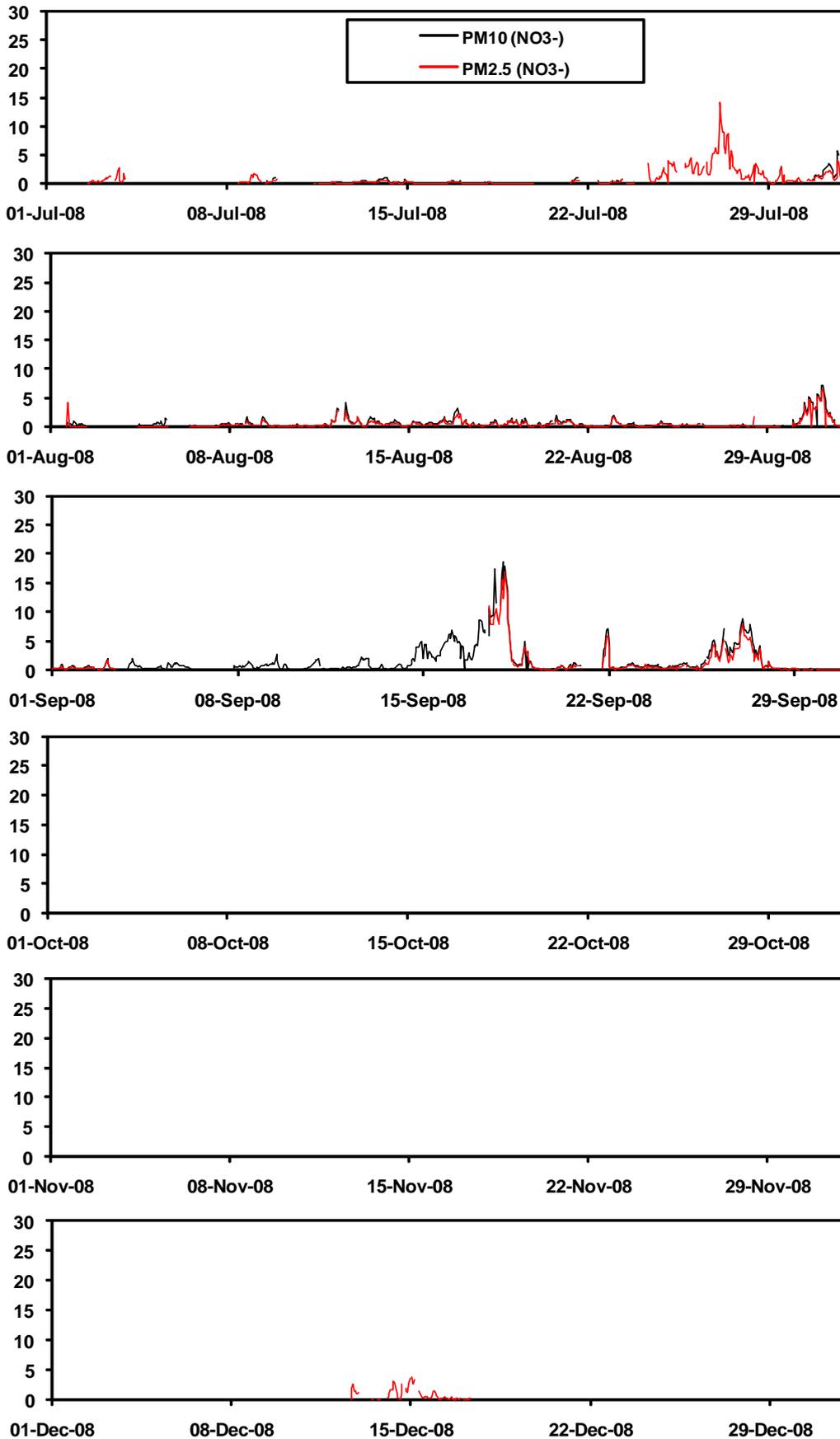
Sulphur dioxide/hydrogen chloride $\mu\text{g m}^{-3}$



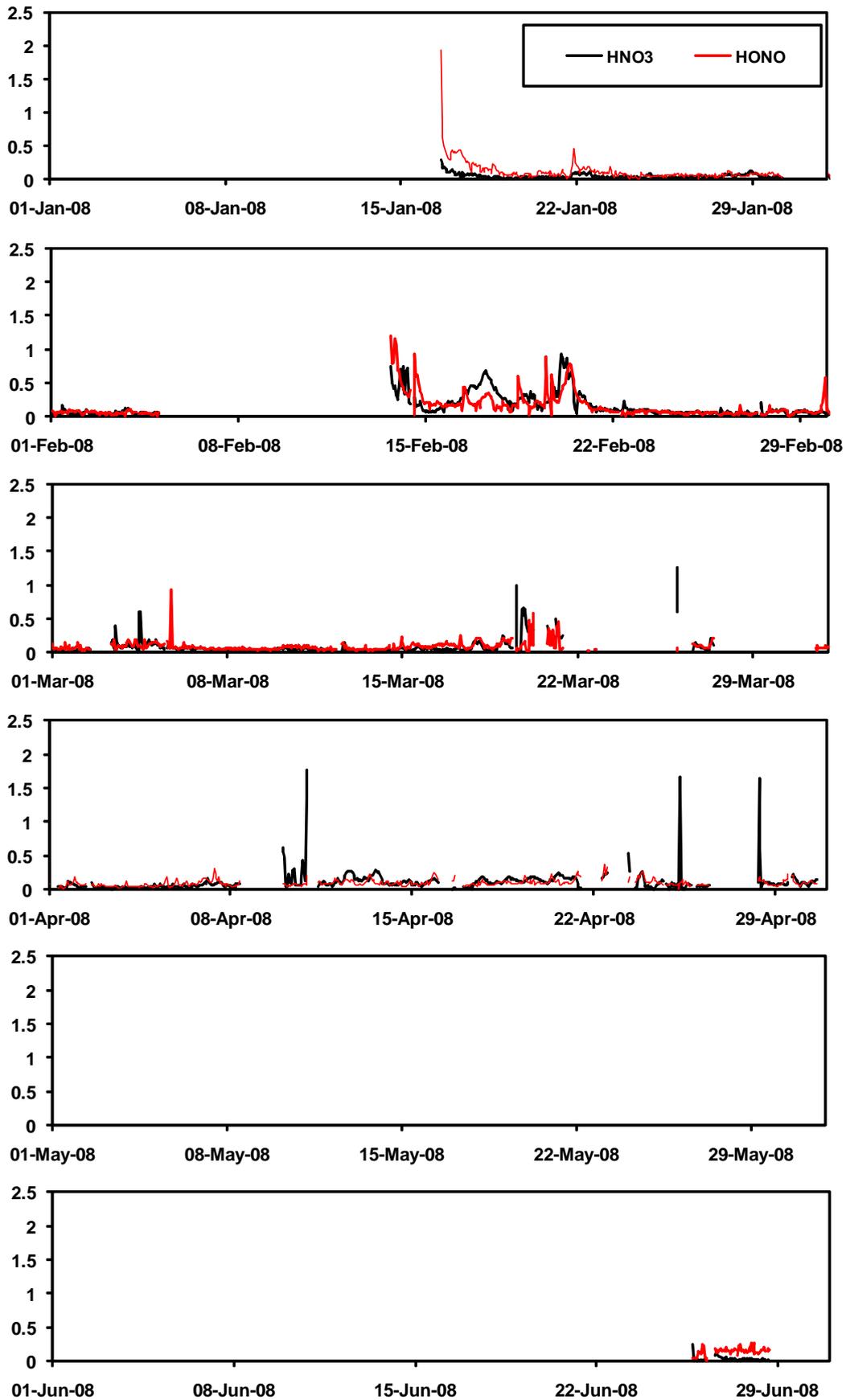
Nitrate aerosol $\mu\text{g m}^{-3}$



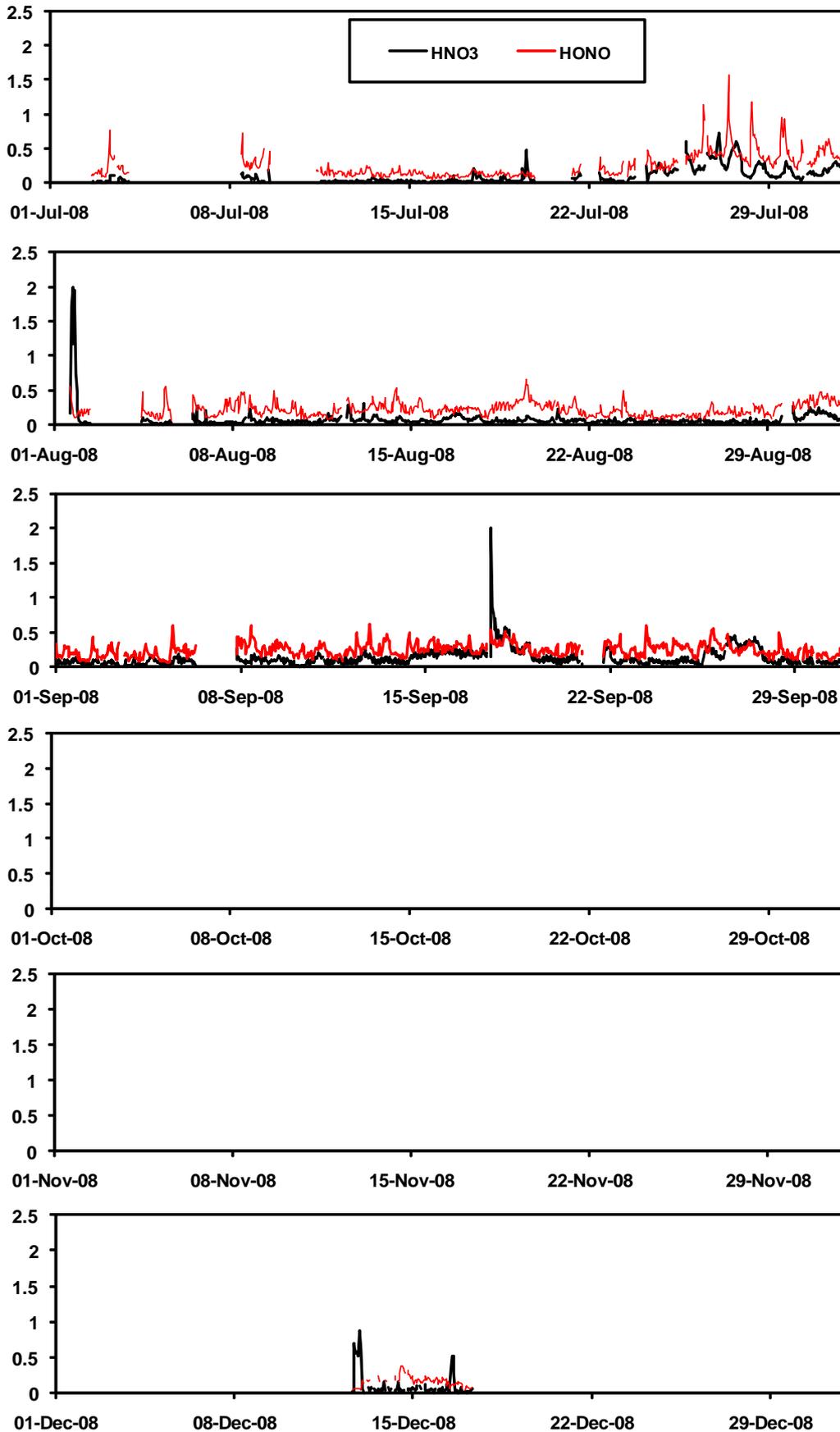
Nitrate aerosol $\mu\text{g m}^{-3}$



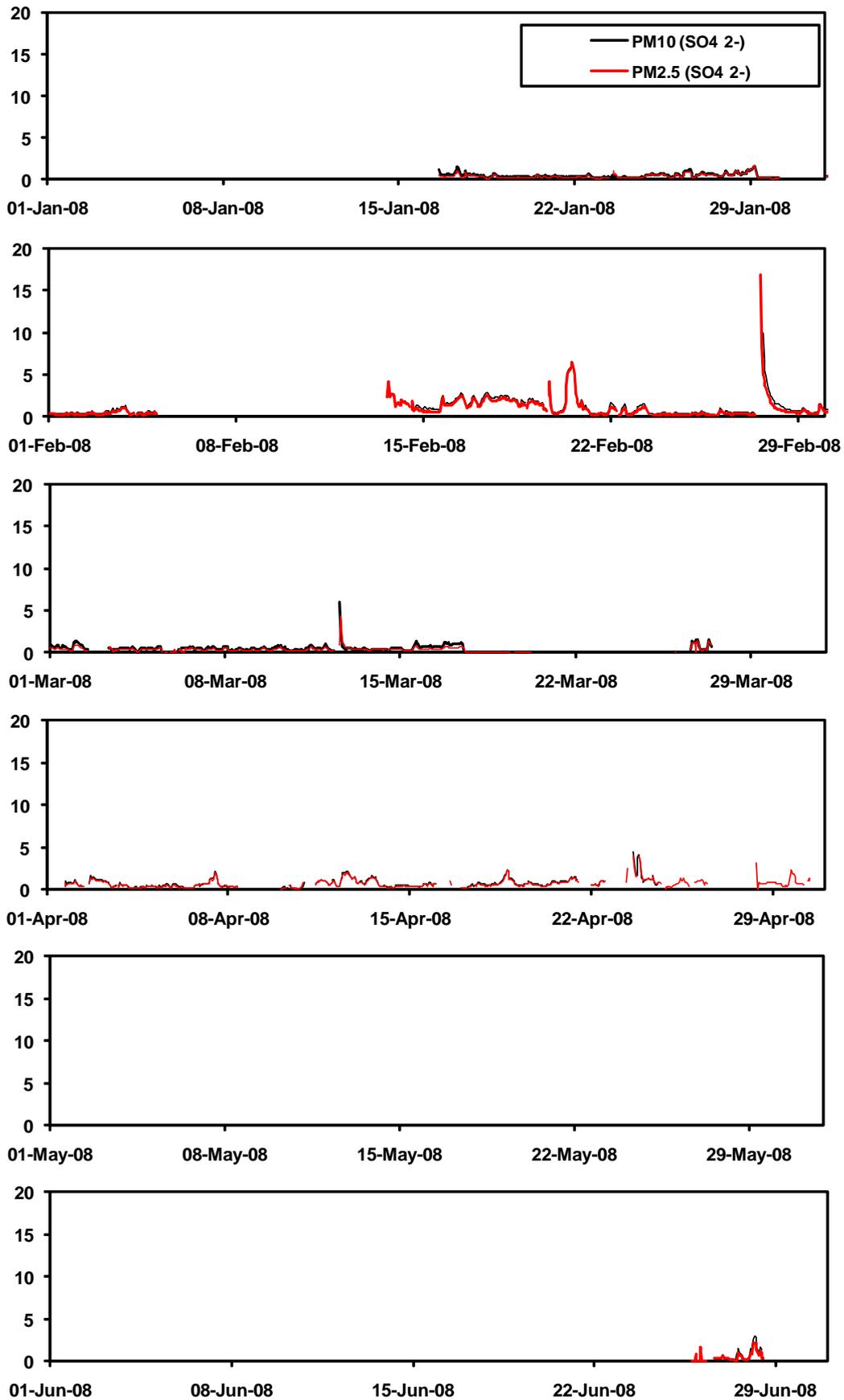
Nitric/nitrous acid $\mu\text{g m}^{-3}$



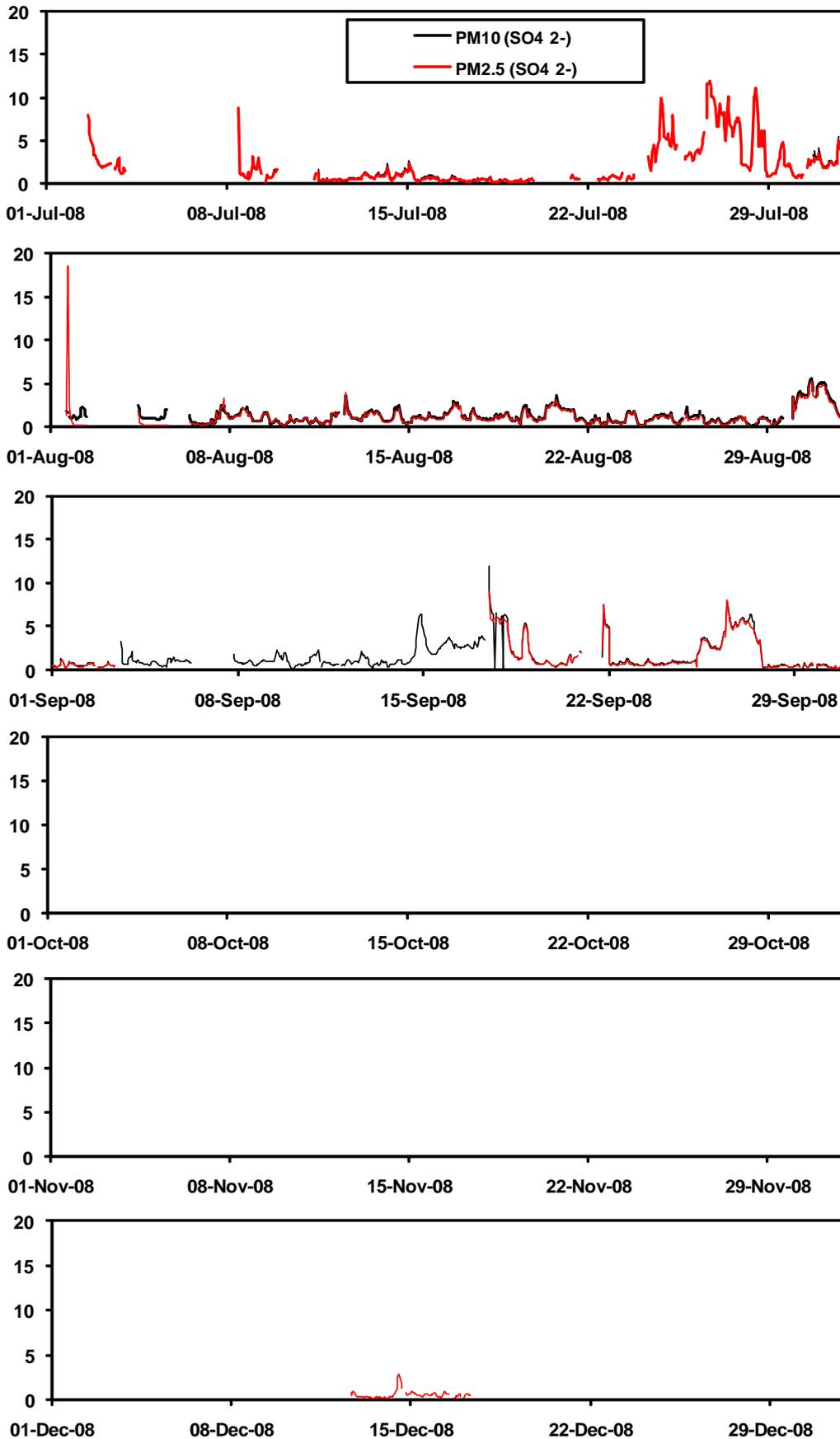
Nitric/nitrous acid $\mu\text{g m}^{-3}$



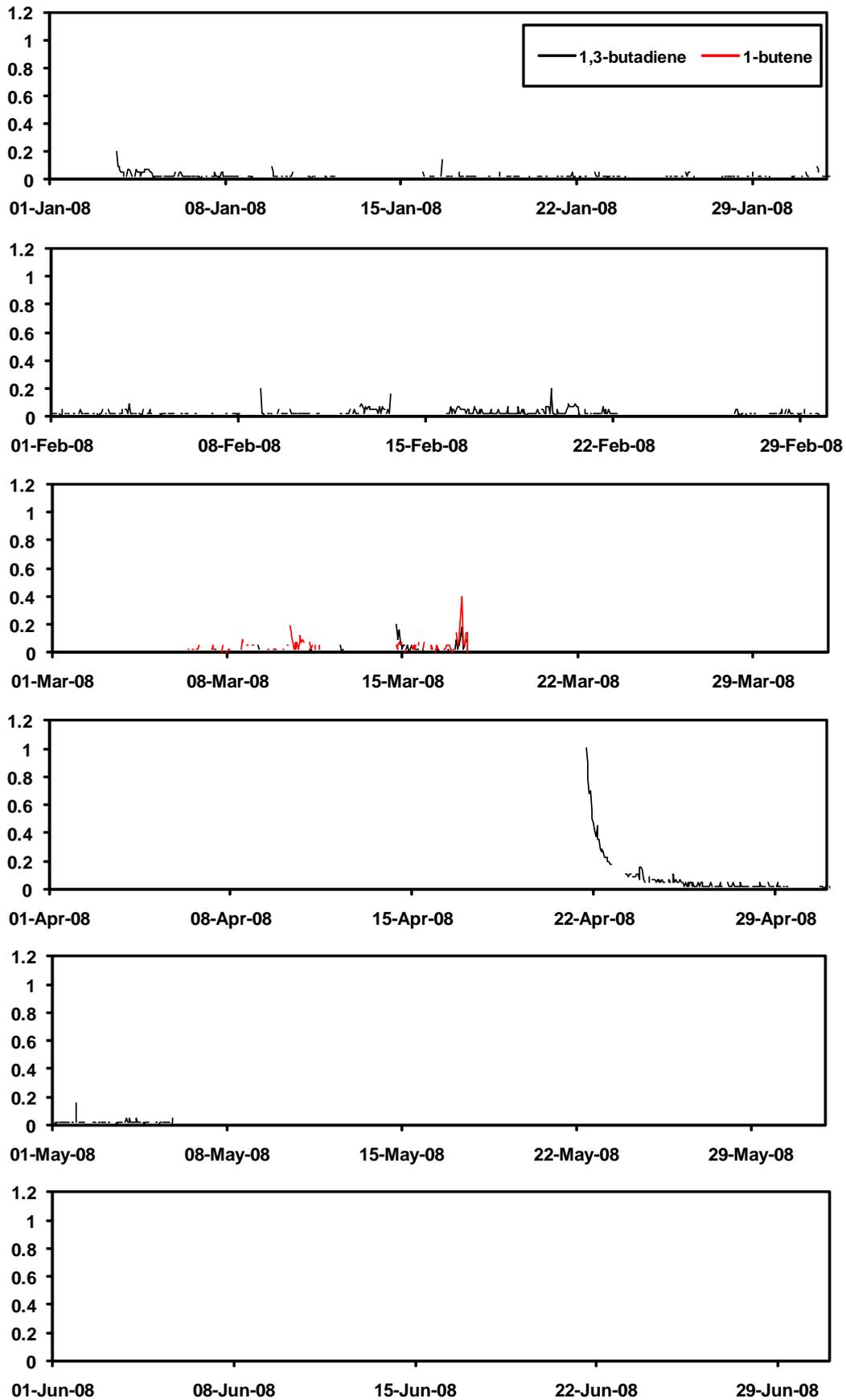
Sulphate aerosol $\mu\text{g m}^{-3}$



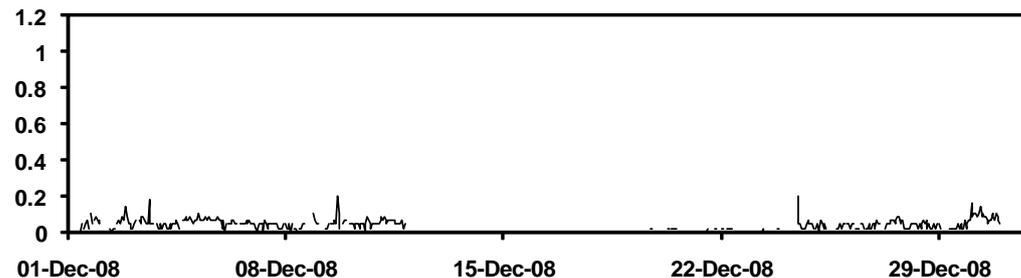
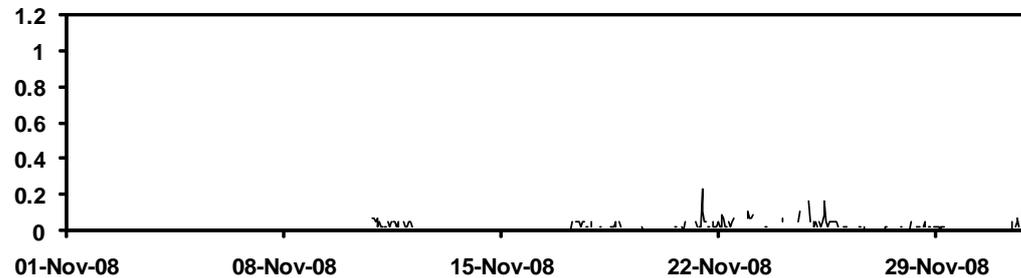
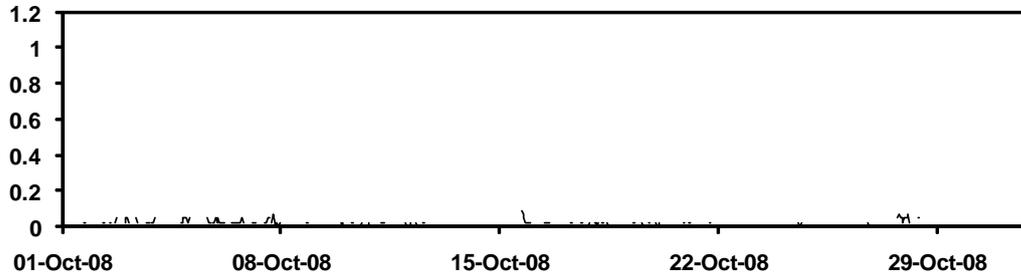
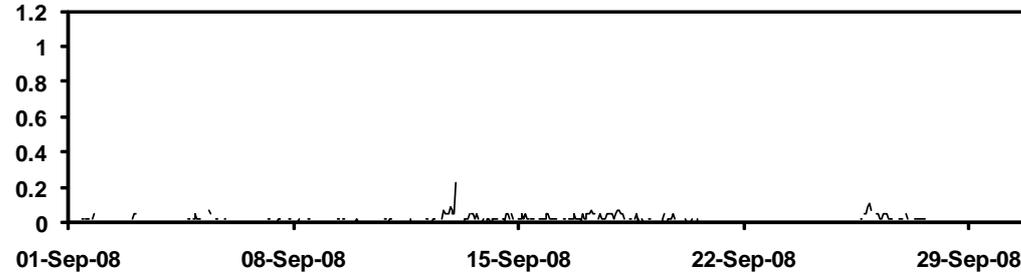
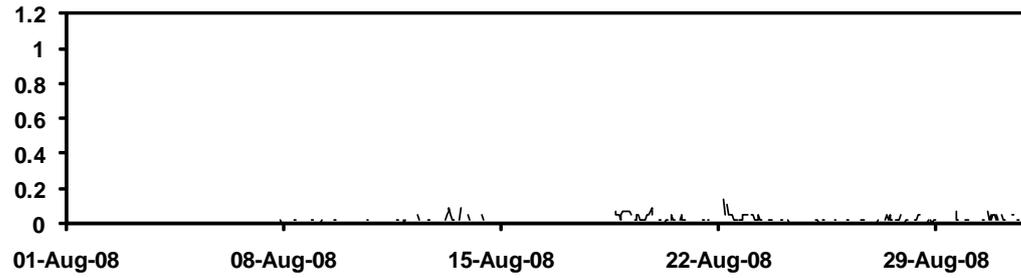
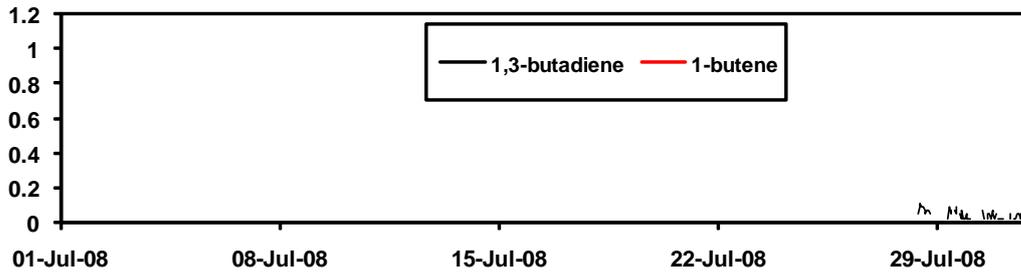
Sulphate aerosol $\mu\text{g m}^{-3}$



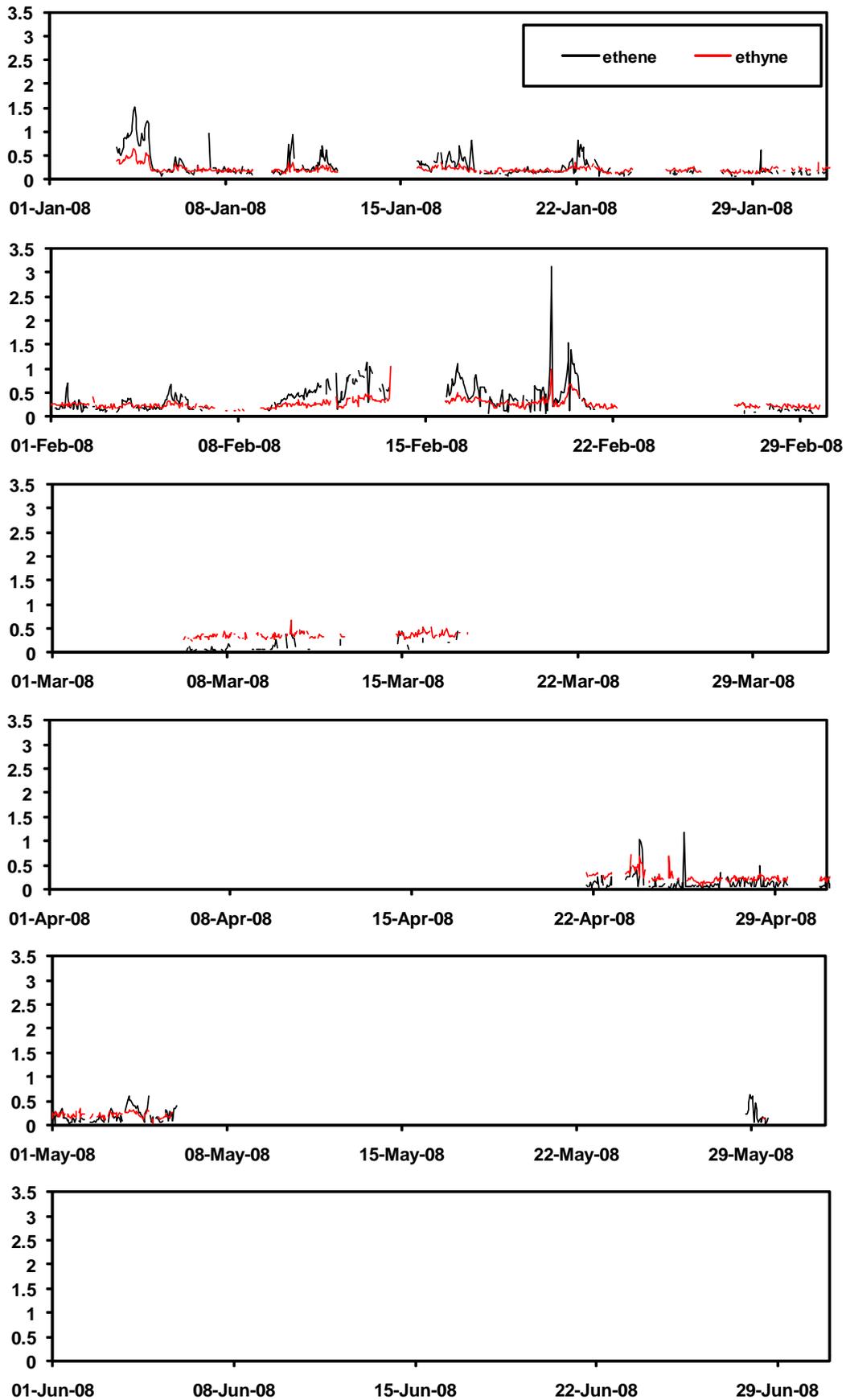
1,3-butadiene/1-butene $\mu\text{g m}^{-3}$



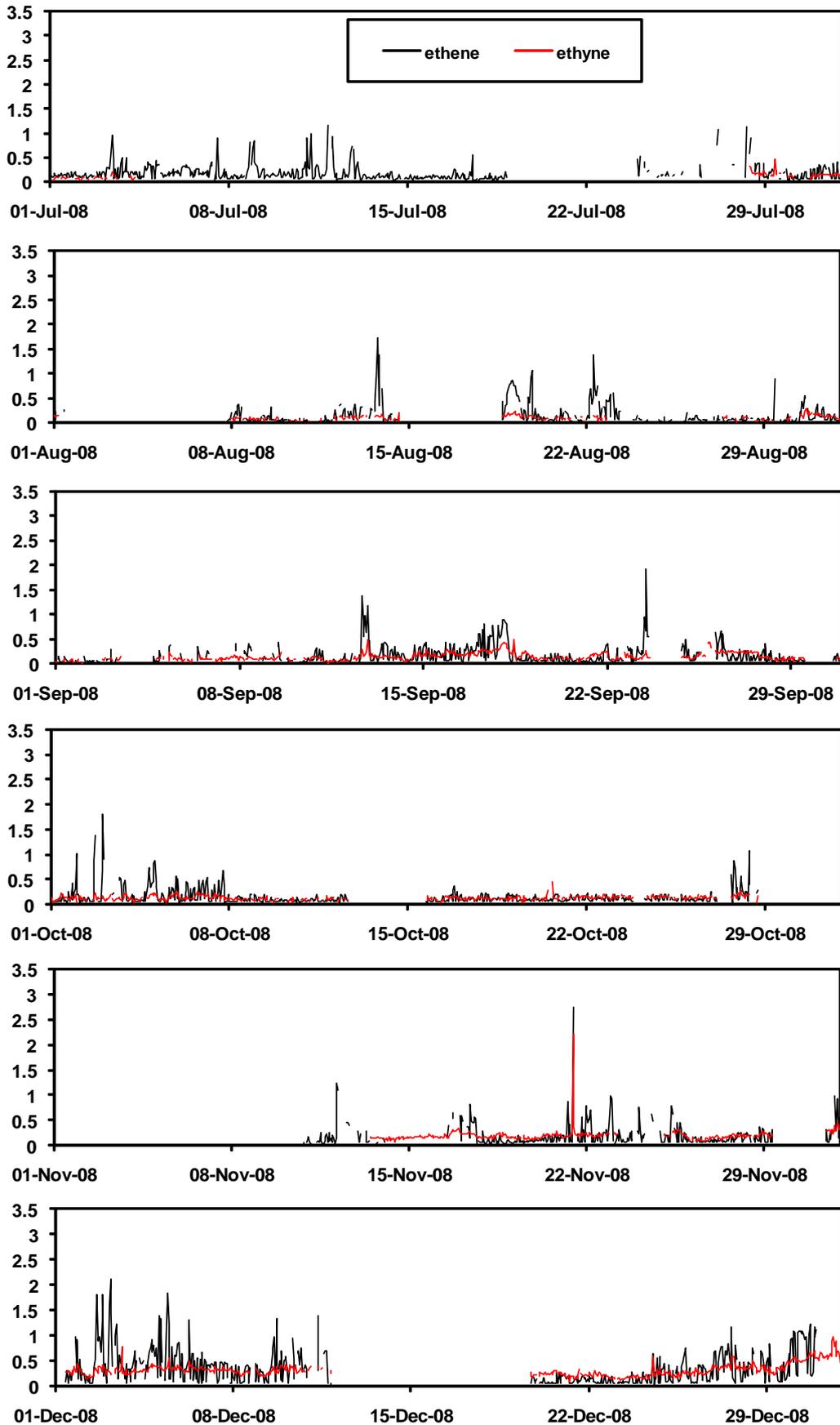
1,3-butadiene/1-butene $\mu\text{g m}^{-3}$



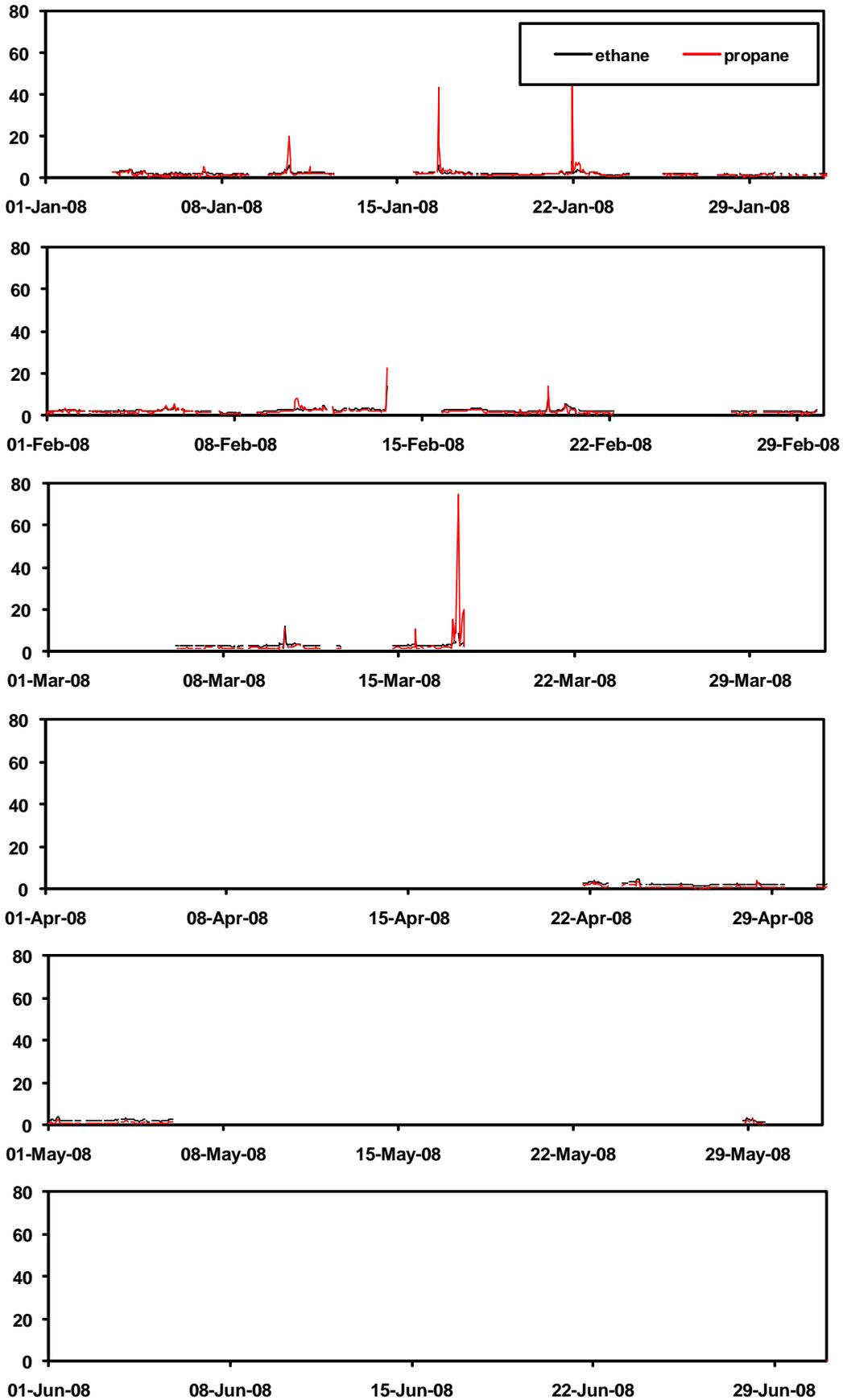
ethene / ethyne $\mu\text{g m}^{-3}$



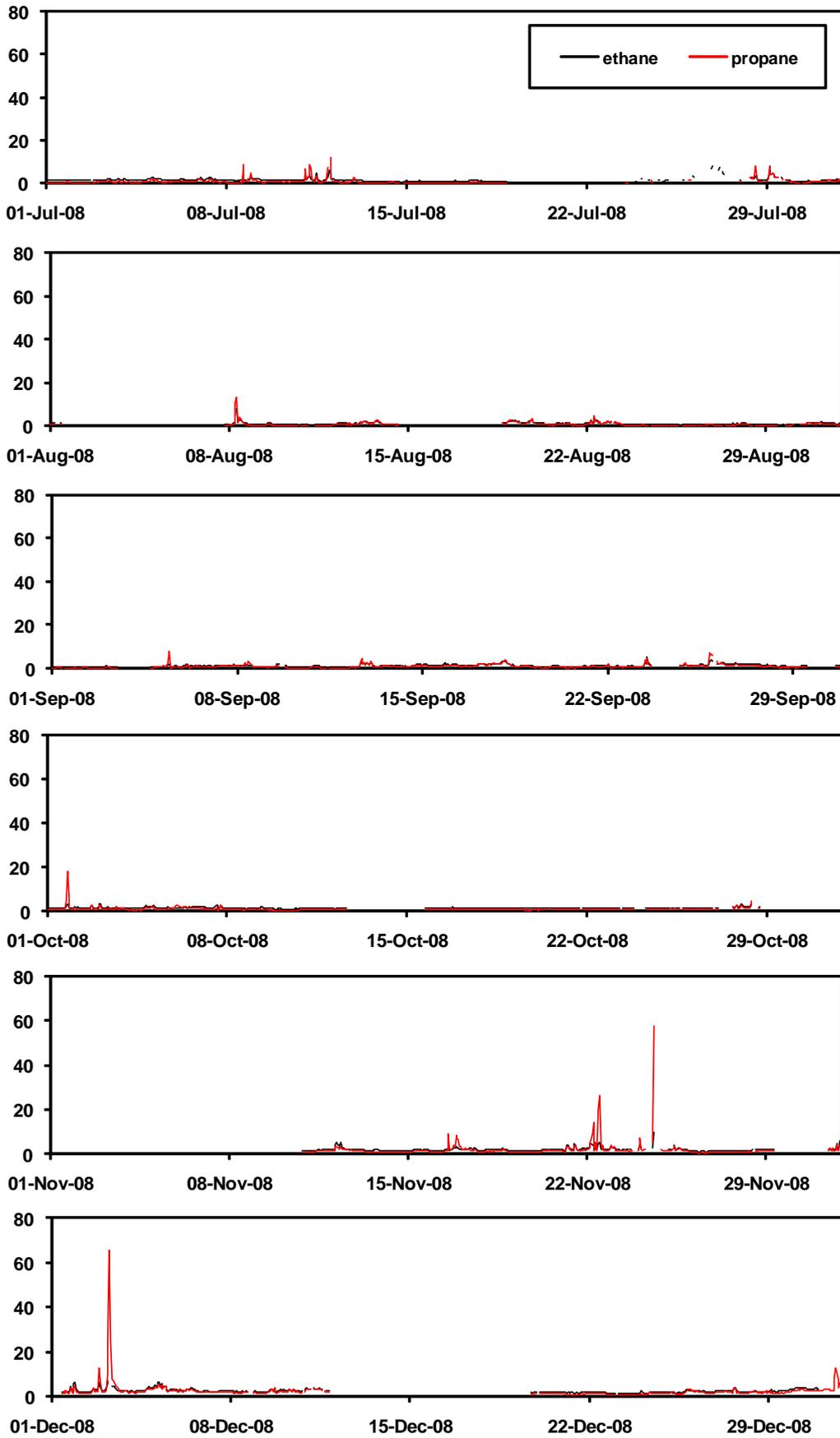
ethene / ethyne $\mu\text{g m}^{-3}$



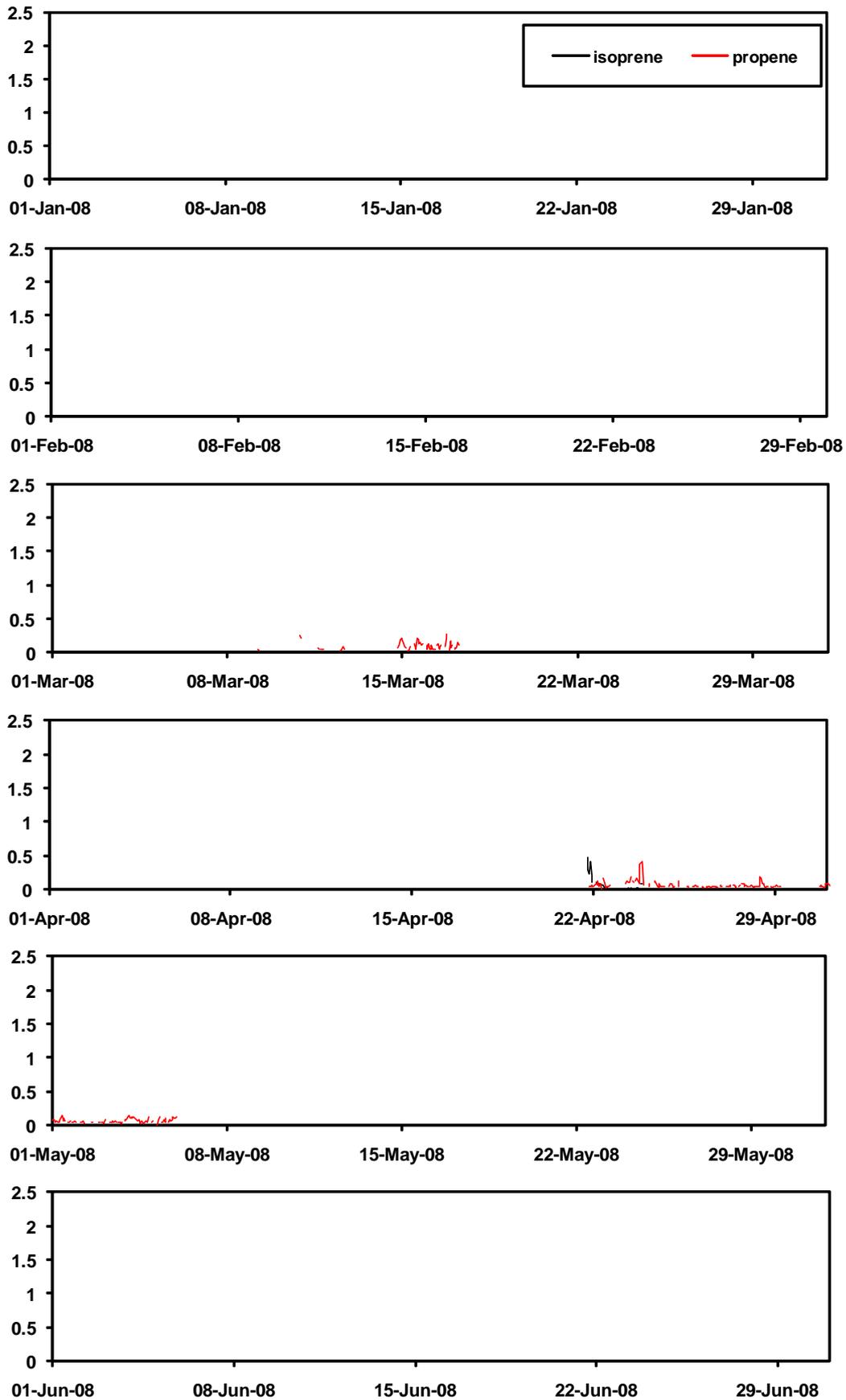
ethane / propane $\mu\text{g m}^{-3}$



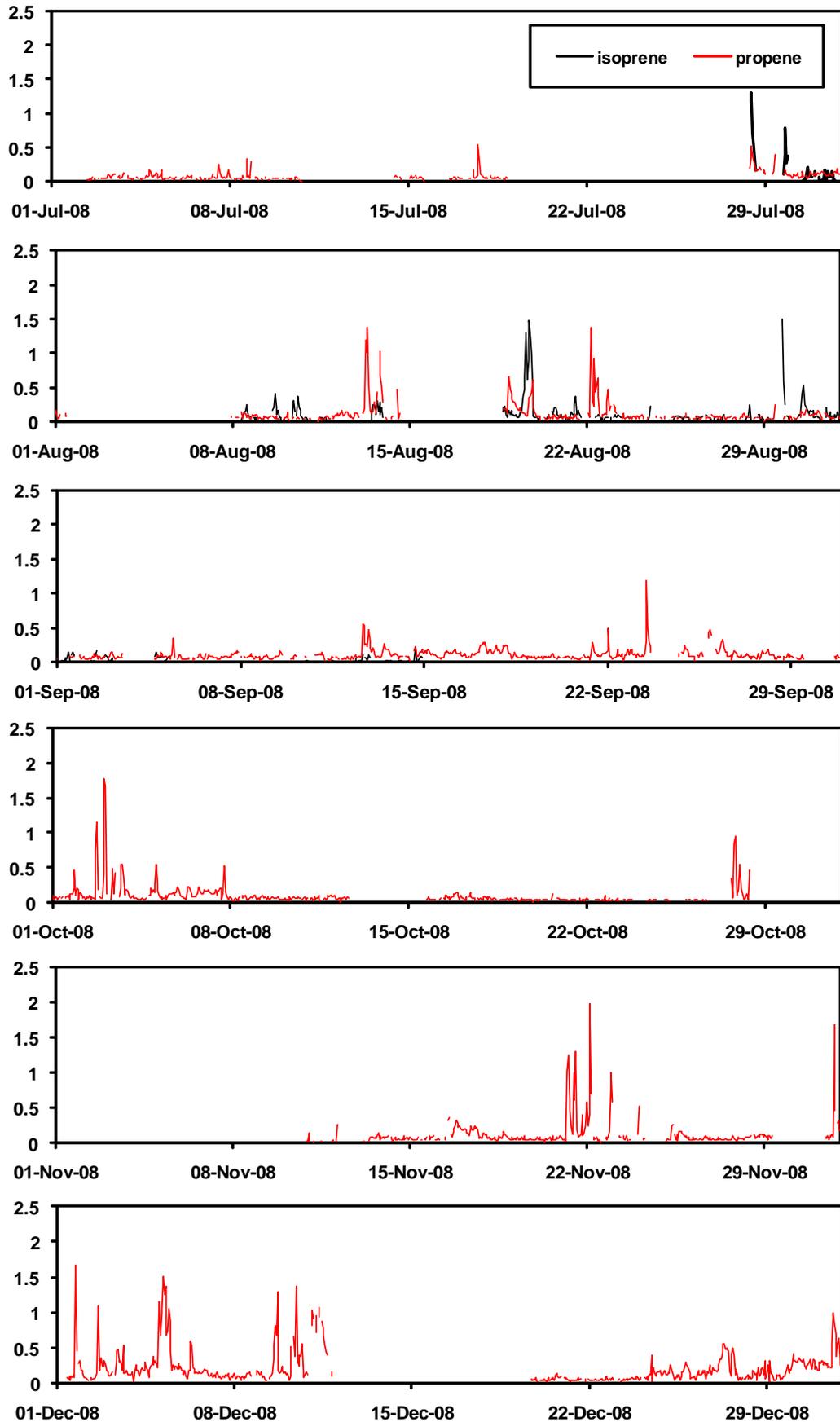
ethane / propane $\mu\text{g m}^{-3}$



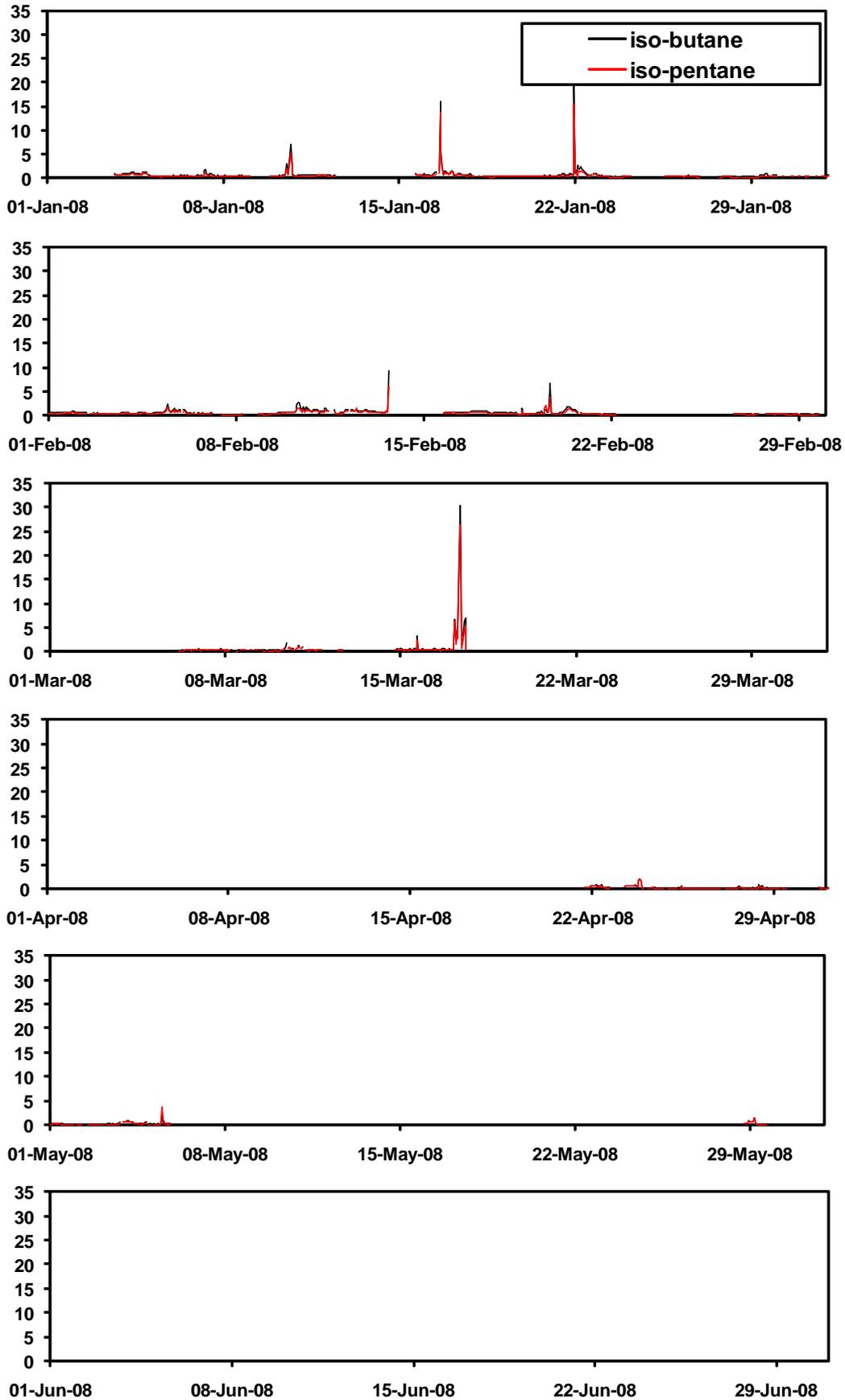
isoprene / propene $\mu\text{g m}^{-3}$



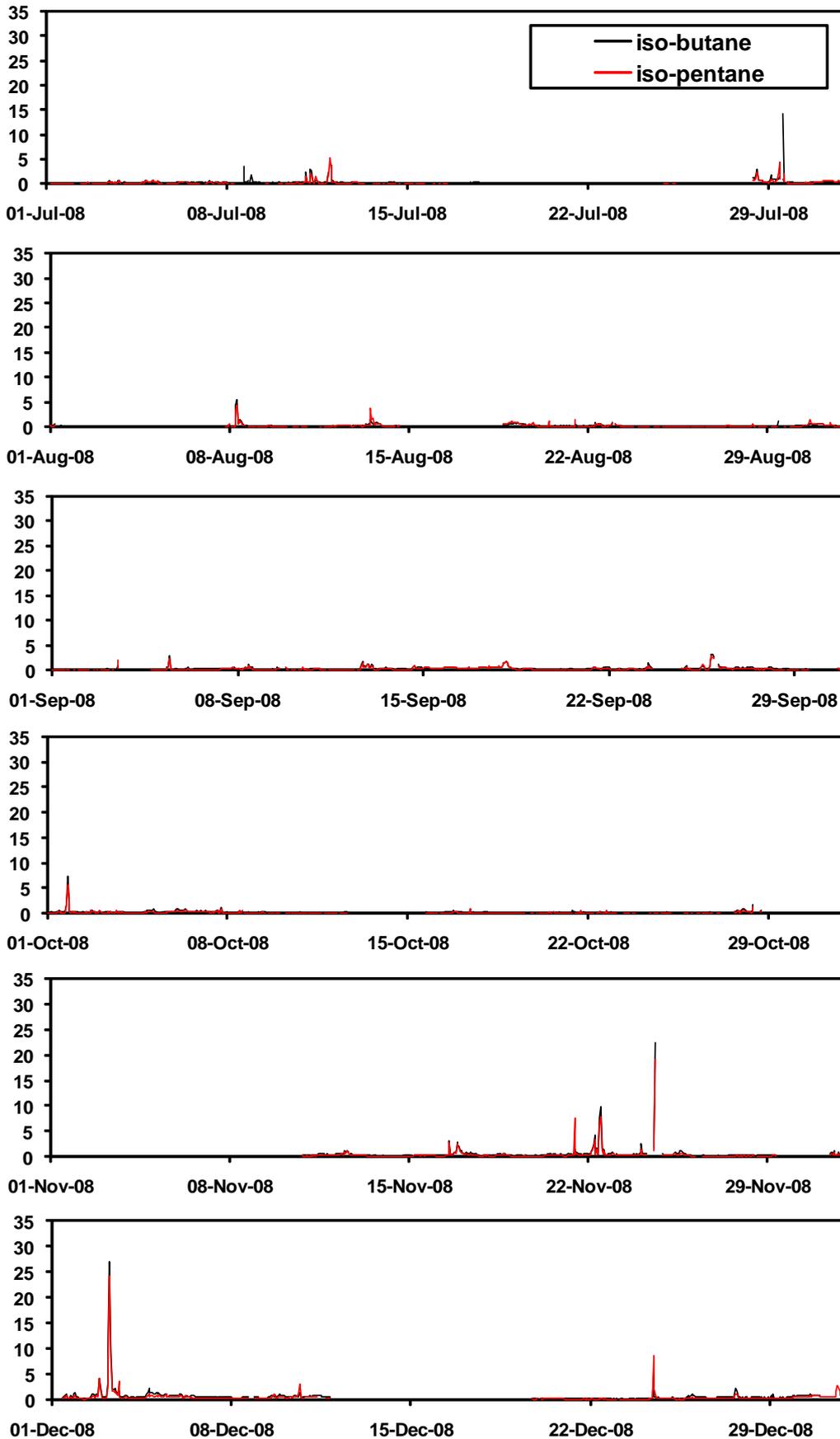
isoprene / propene $\mu\text{g m}^{-3}$



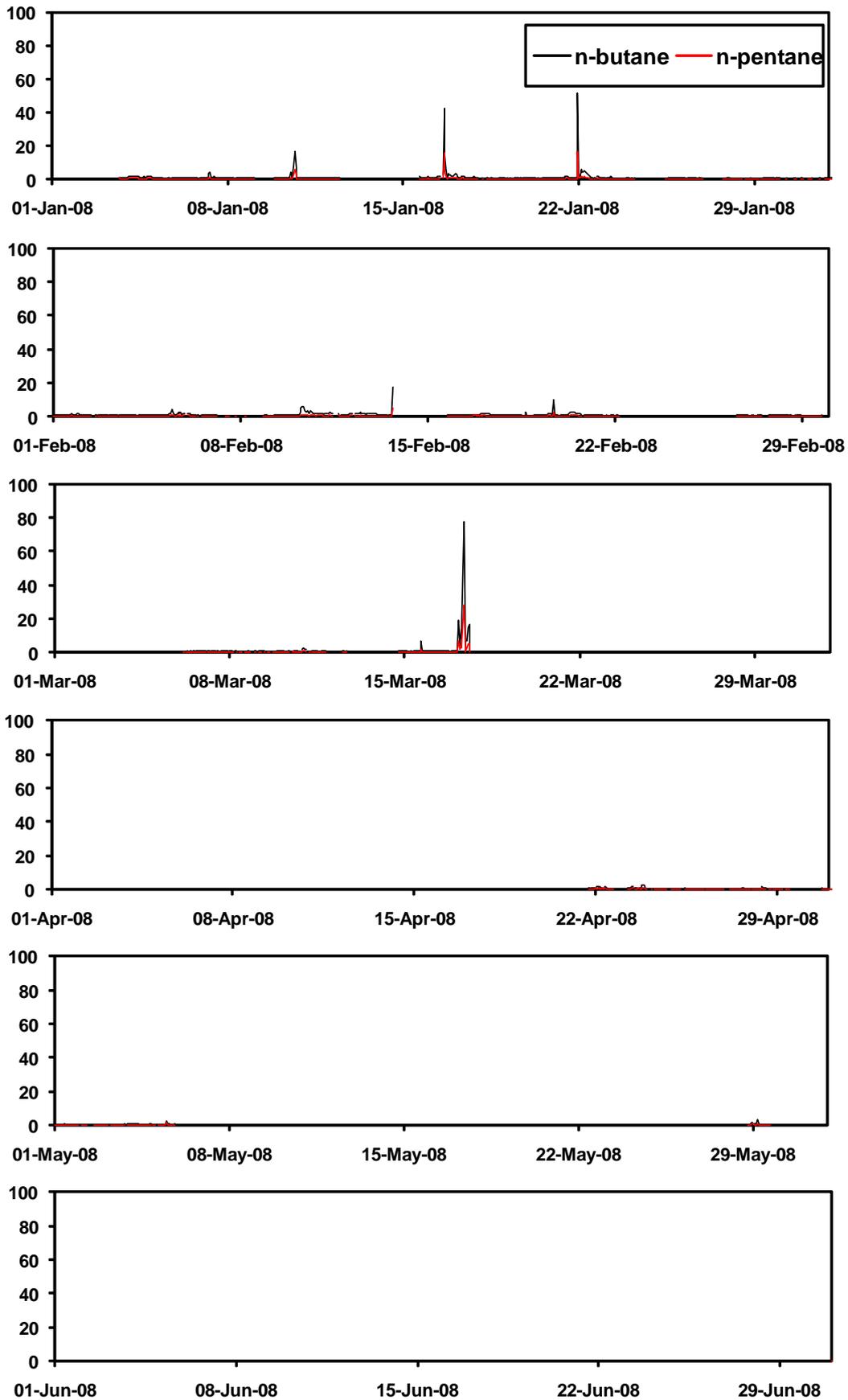
iso-butane / iso-pentane $\mu\text{g m}^{-3}$



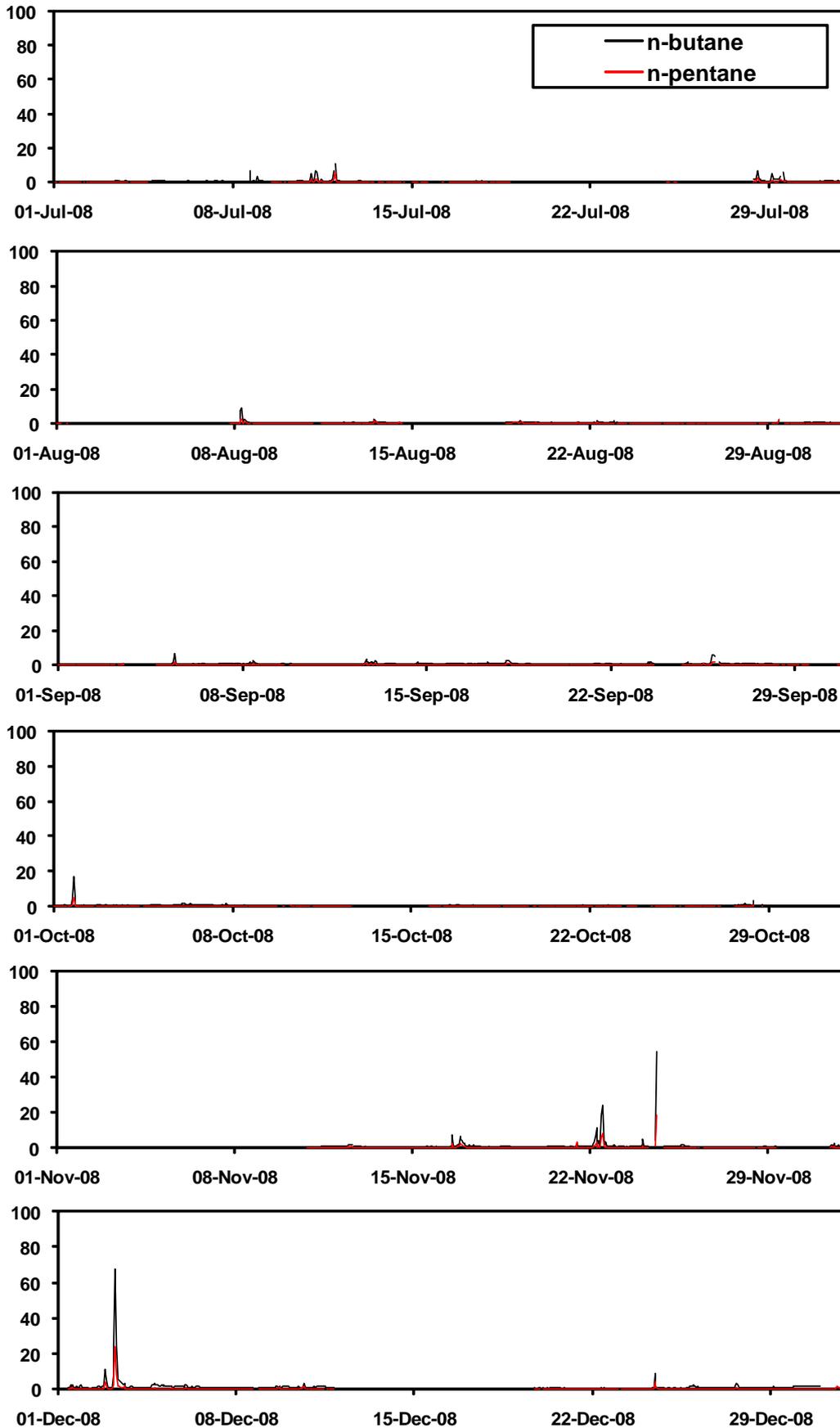
iso-butane / iso-pentane $\mu\text{g m}^{-3}$



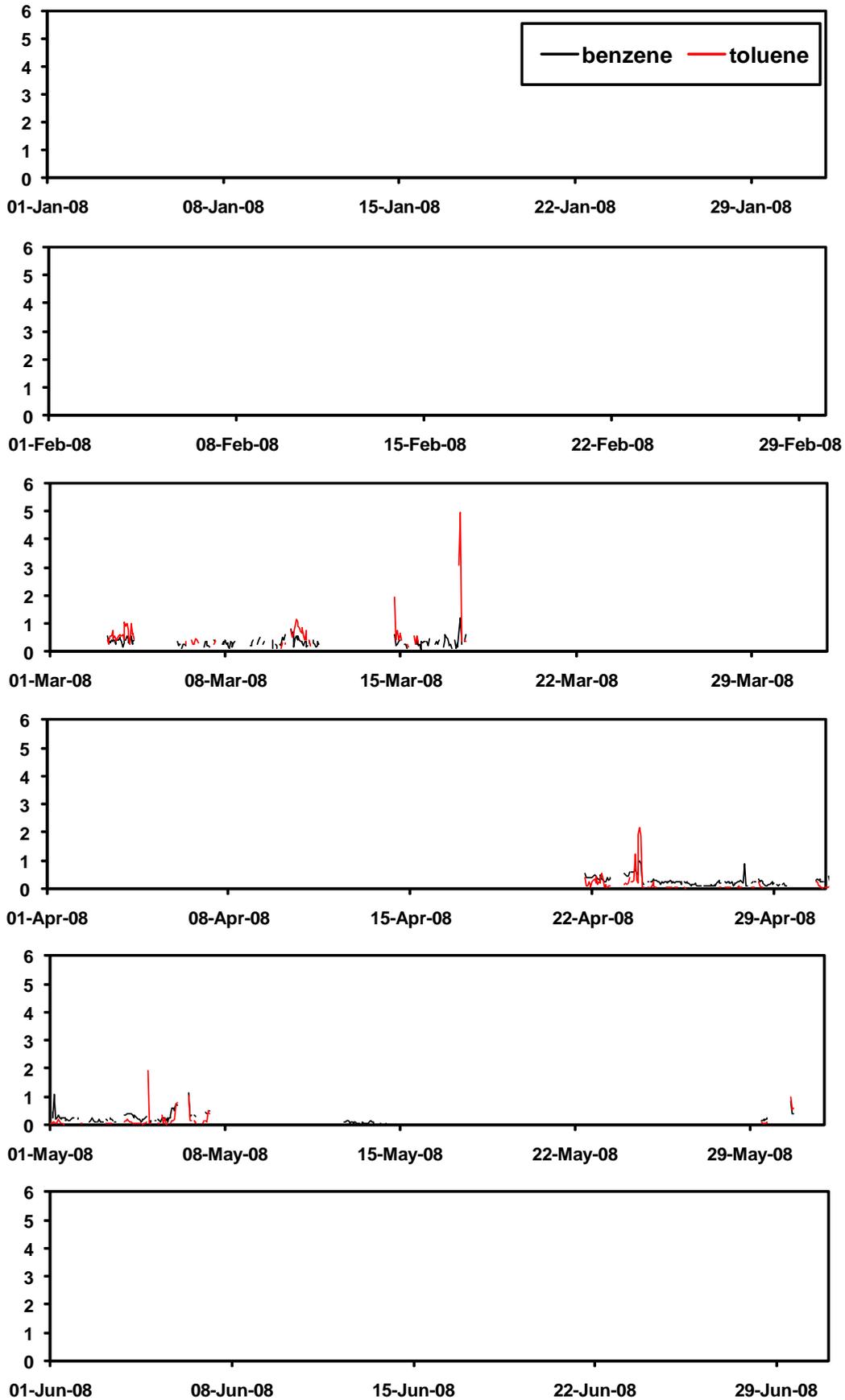
n-butane / n-pentane $\mu\text{g m}^{-3}$



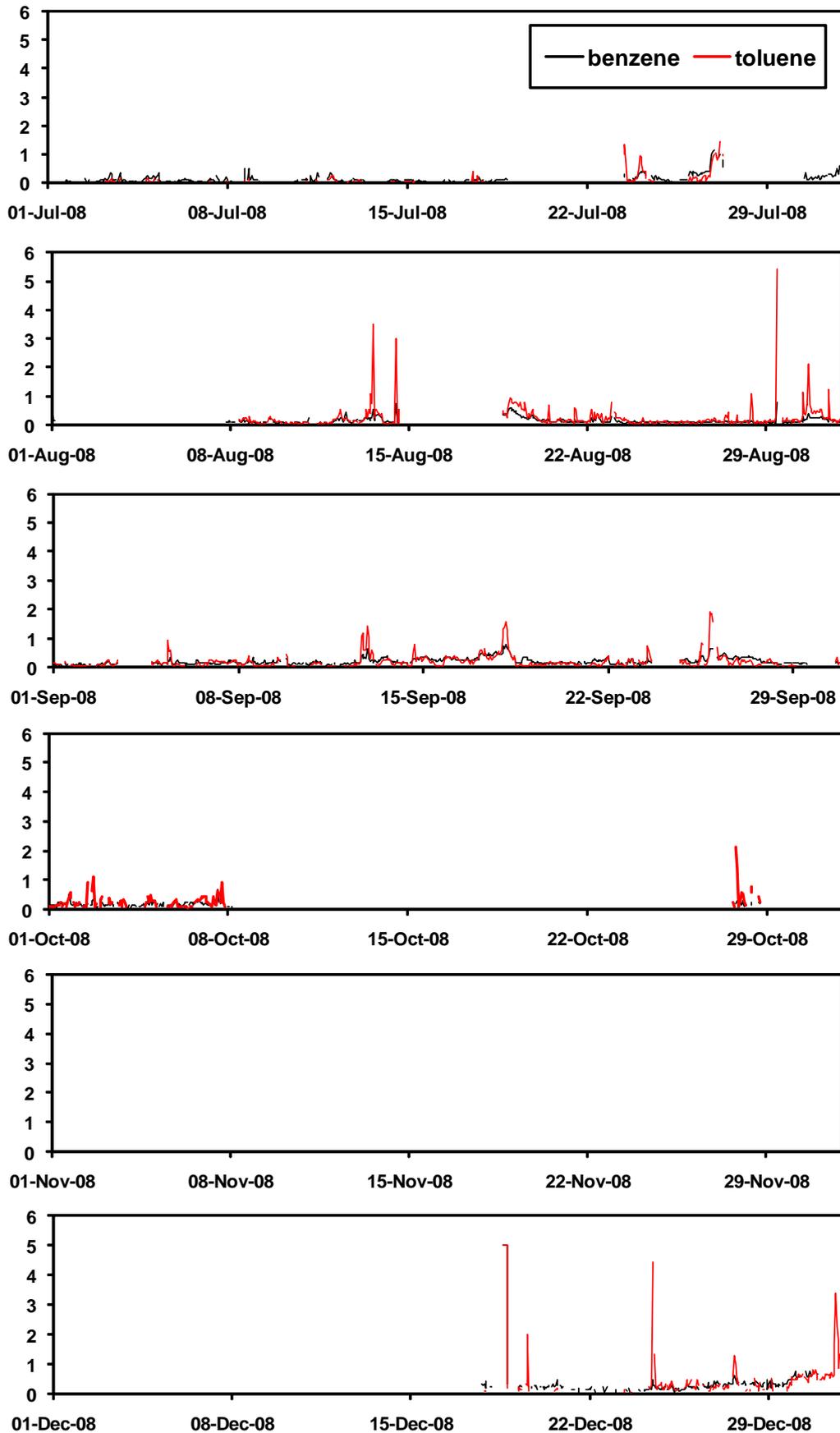
n-butane / n-pentane $\mu\text{g m}^{-3}$



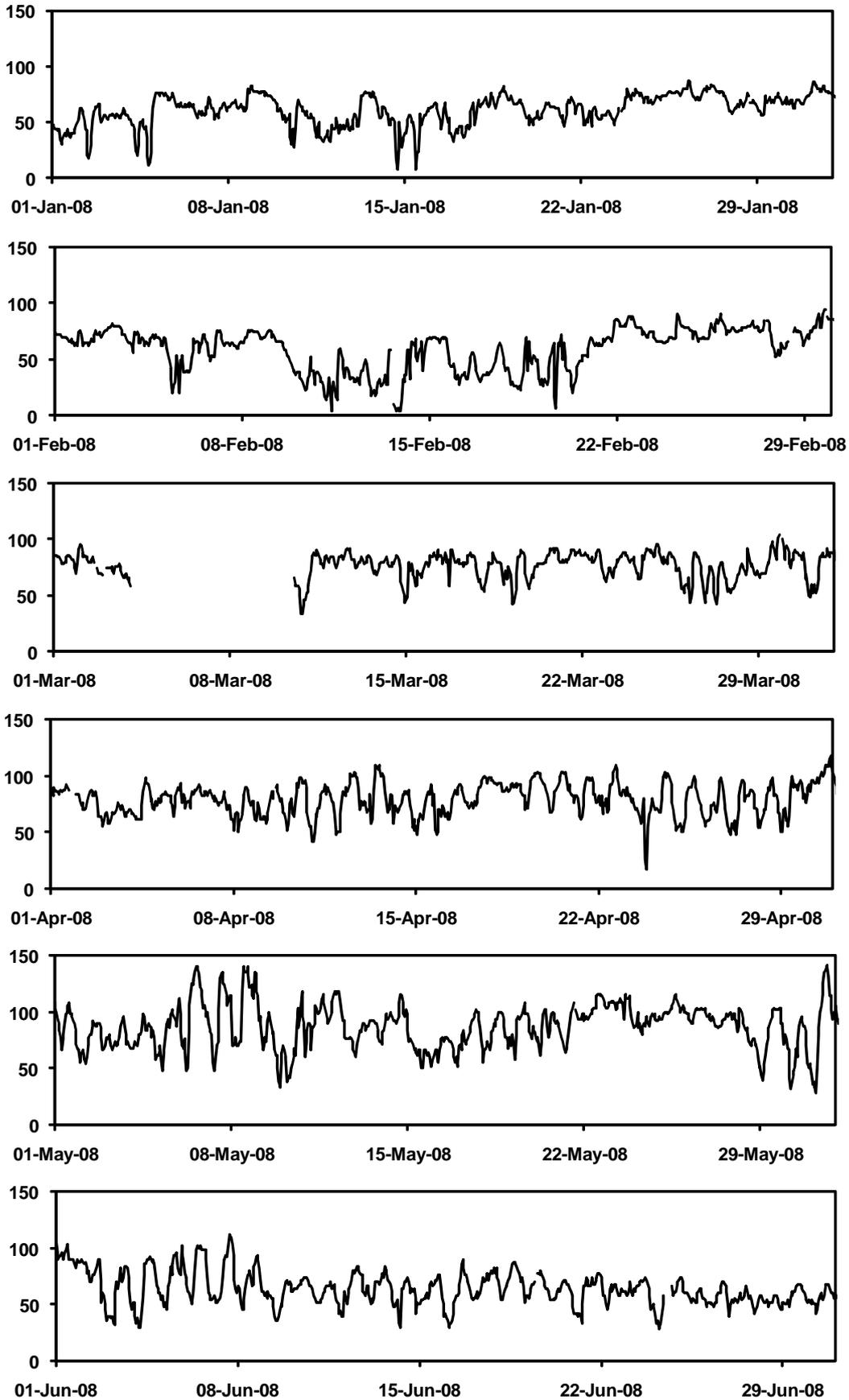
benzene / toluene $\mu\text{g m}^{-3}$



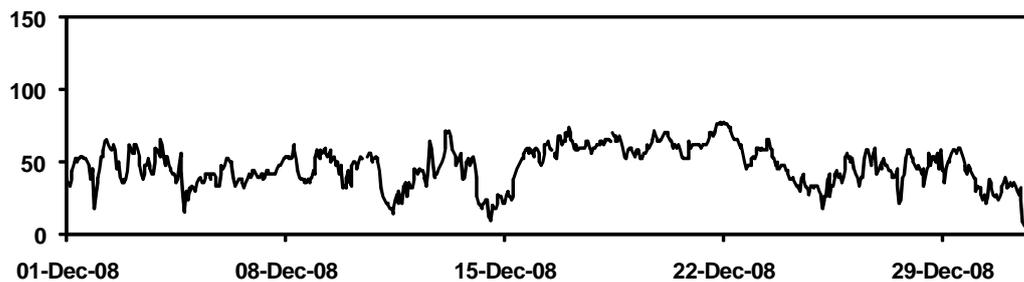
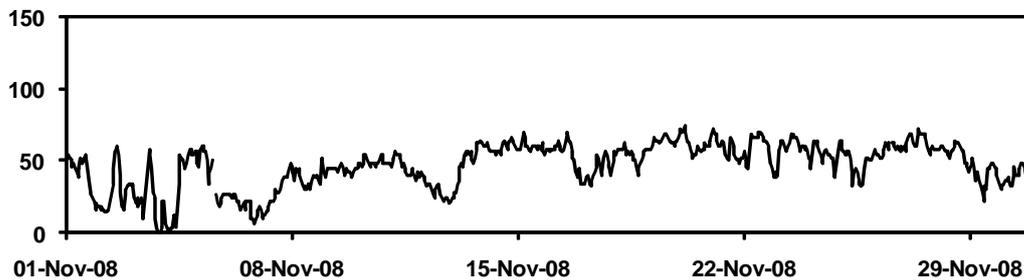
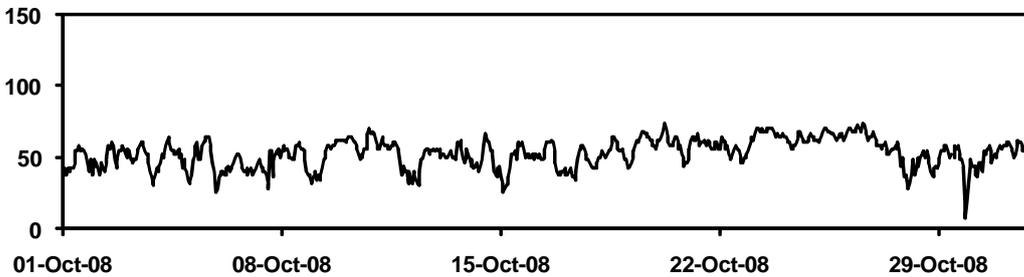
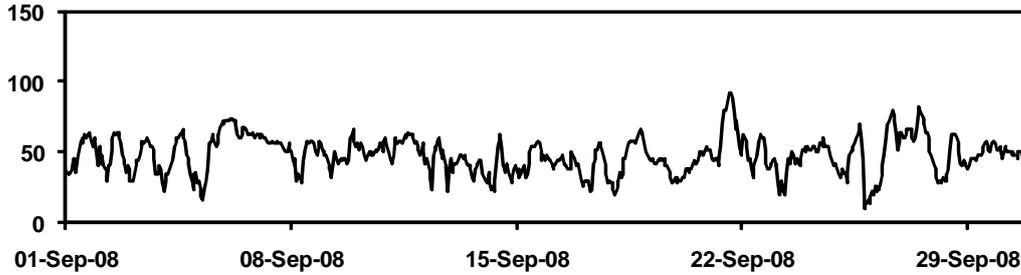
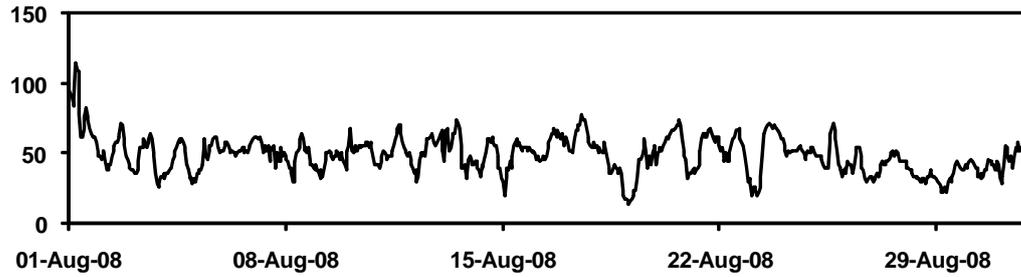
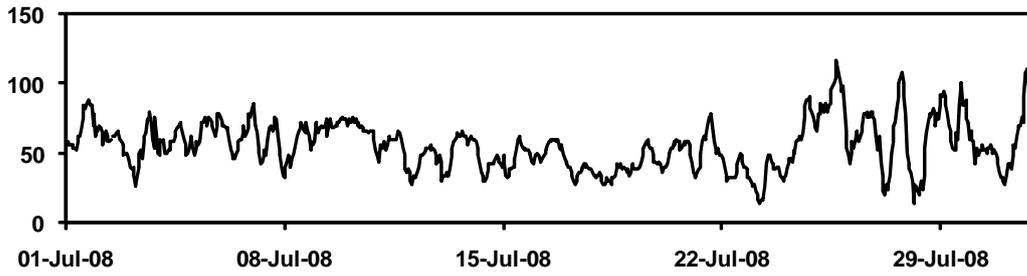
benzene / toluene $\mu\text{g m}^{-3}$



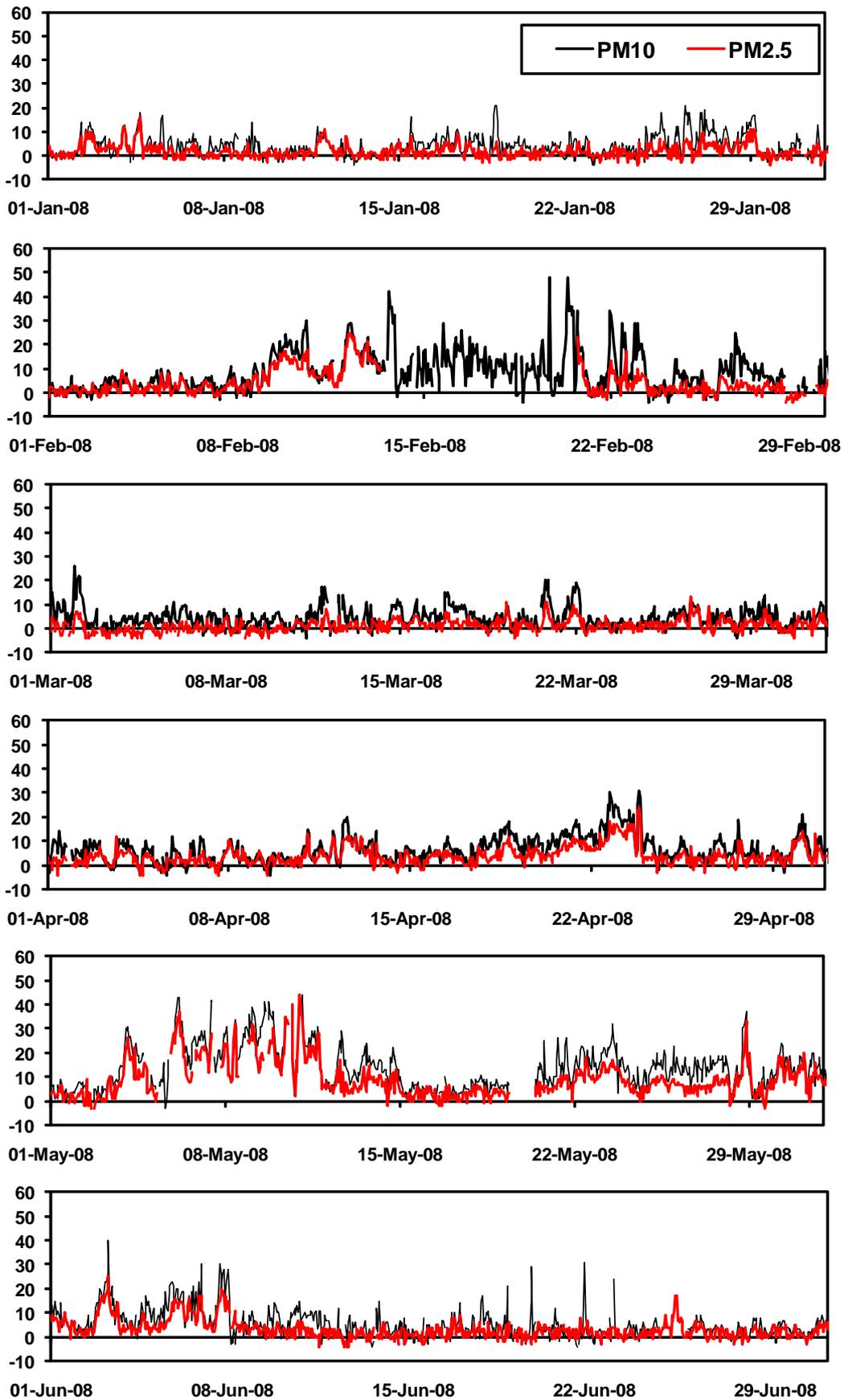
Ozone (AURN) $\mu\text{g m}^{-3}$



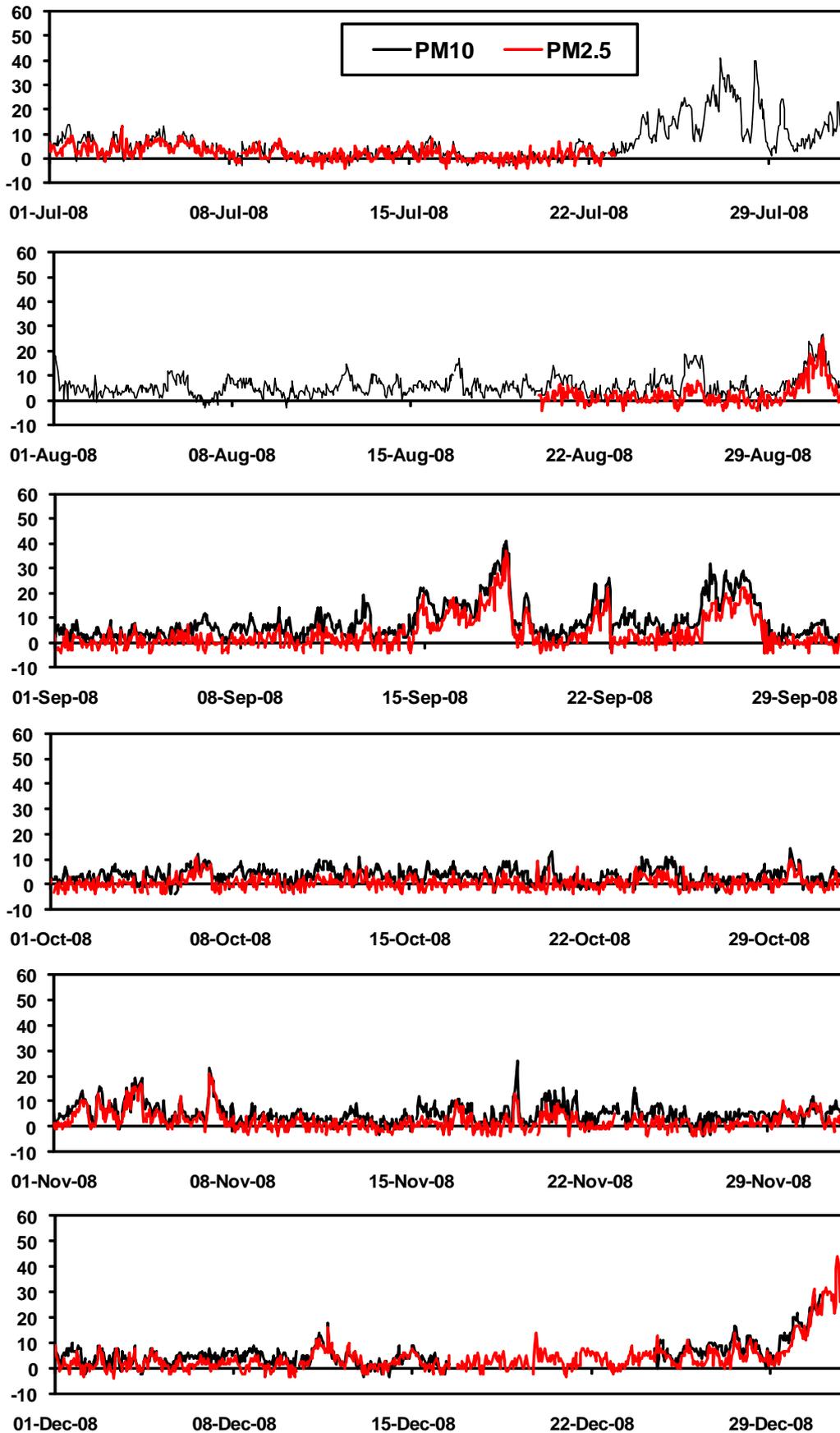
Ozone (AURN) $\mu\text{g m}^{-3}$



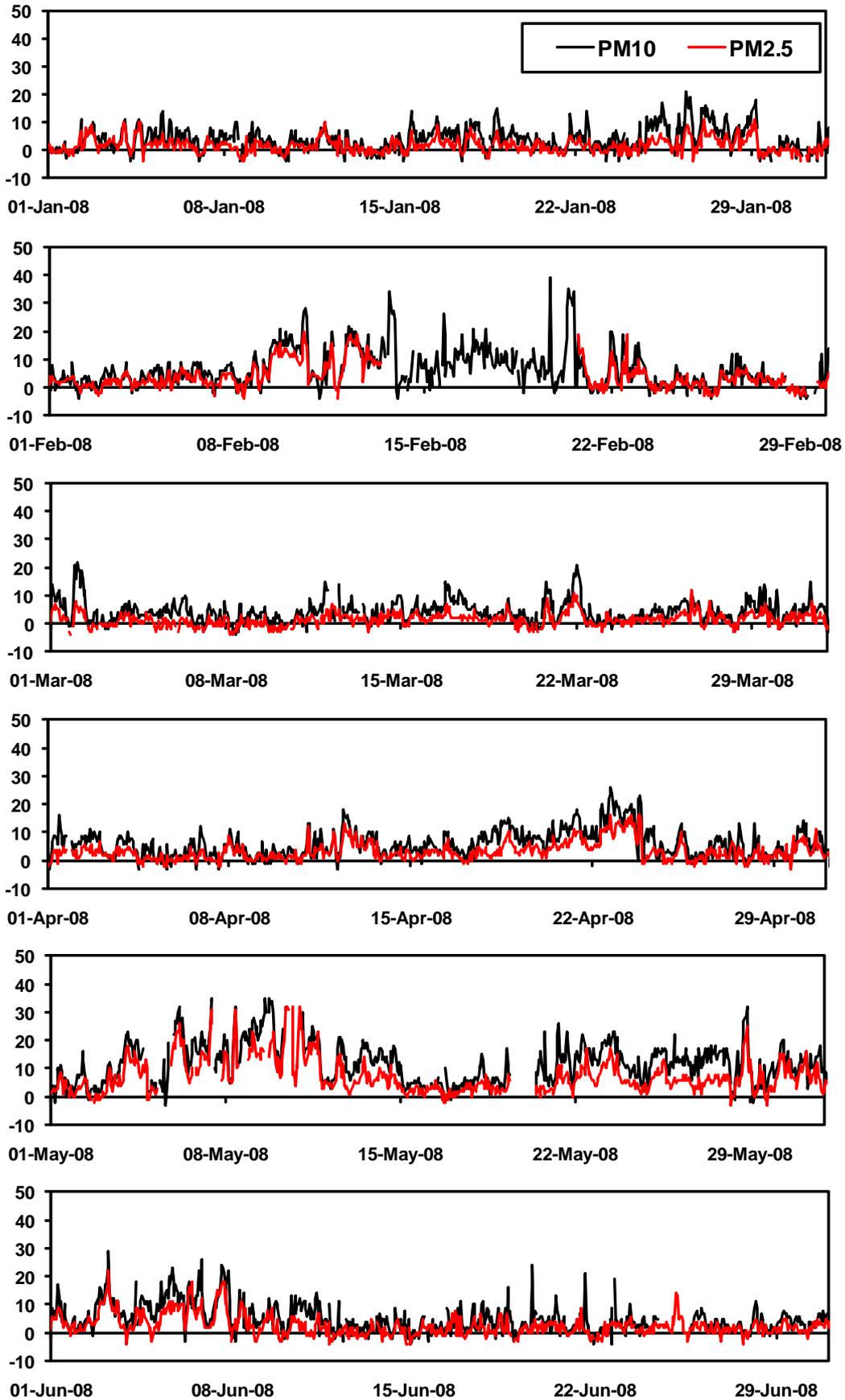
Particulate matter (TEOM/FDMS) $\mu\text{g m}^{-3}$



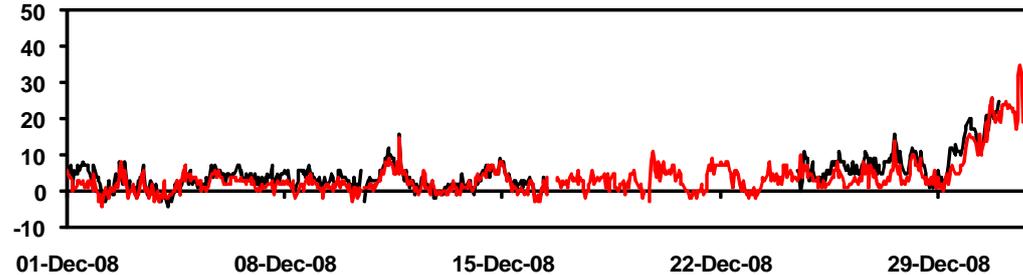
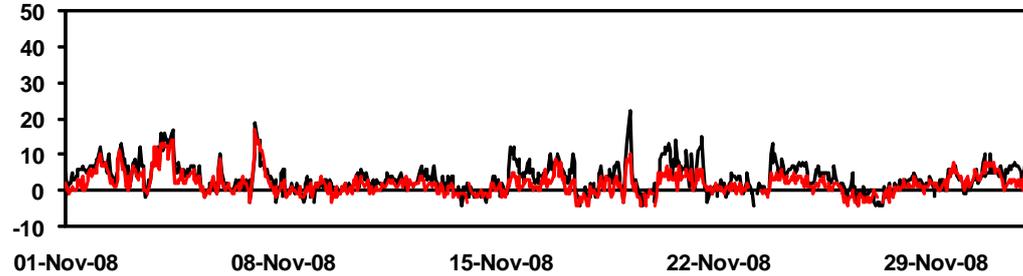
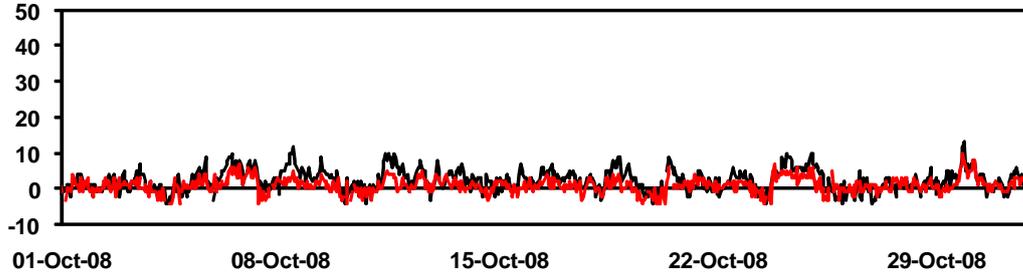
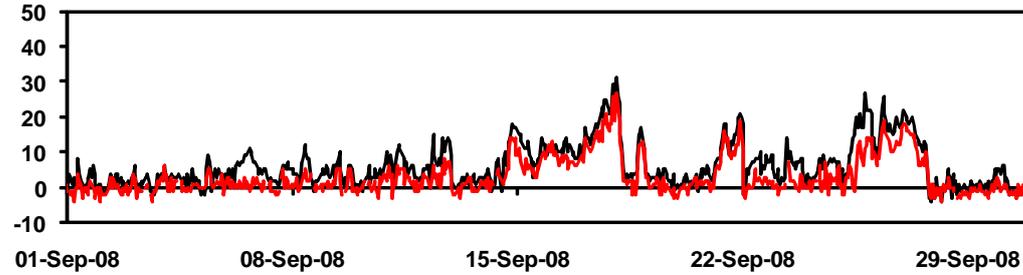
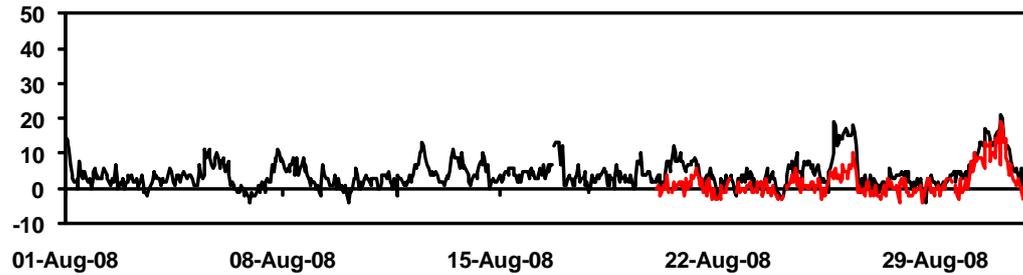
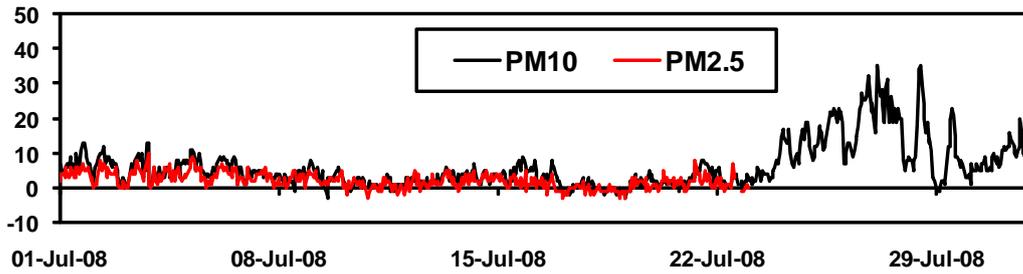
Particulate matter (TEOM/FDMS) $\mu\text{g m}^{-3}$



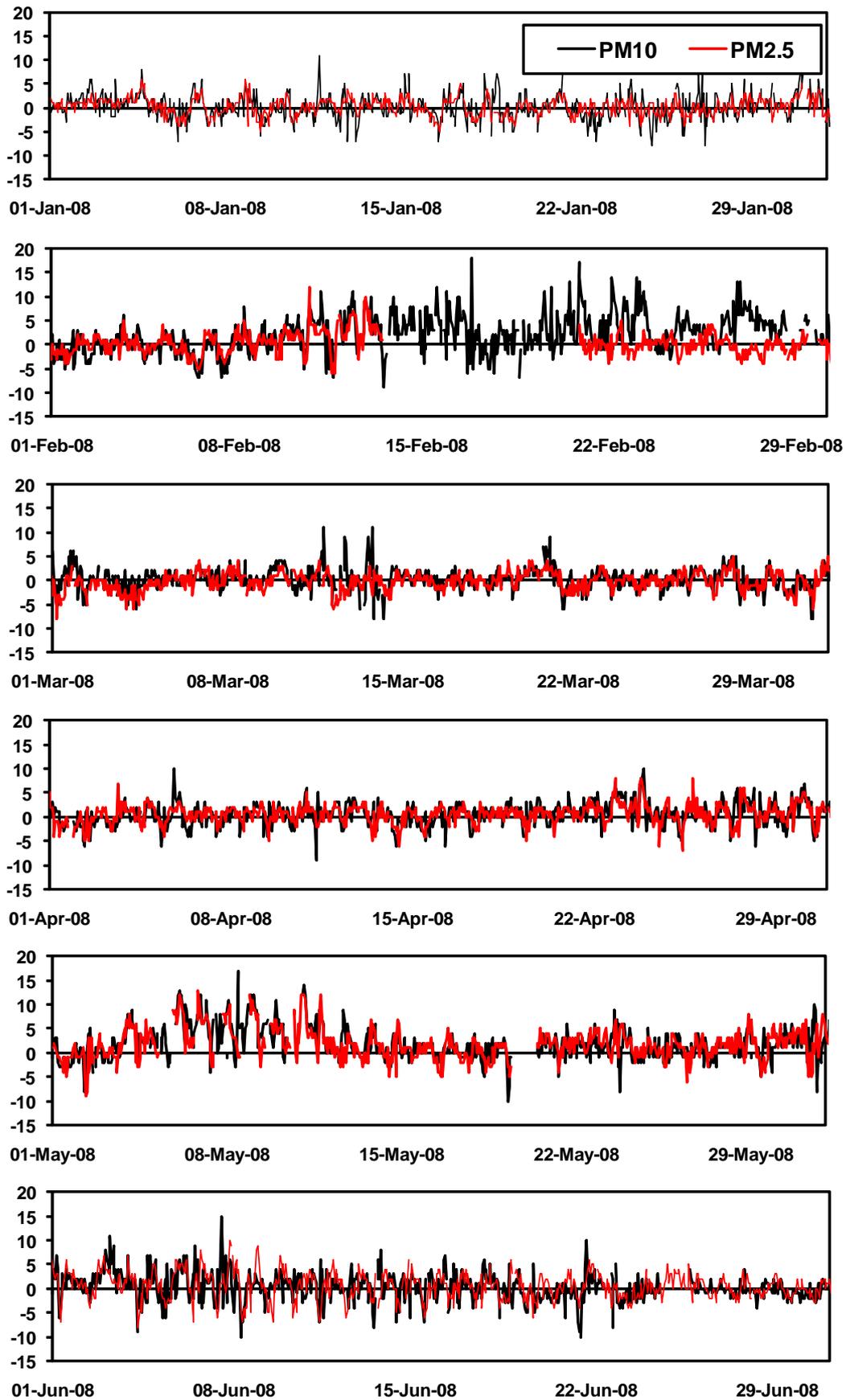
Non-volatile particulate matter (TEOM/FDMS) $\mu\text{g m}^{-3}$



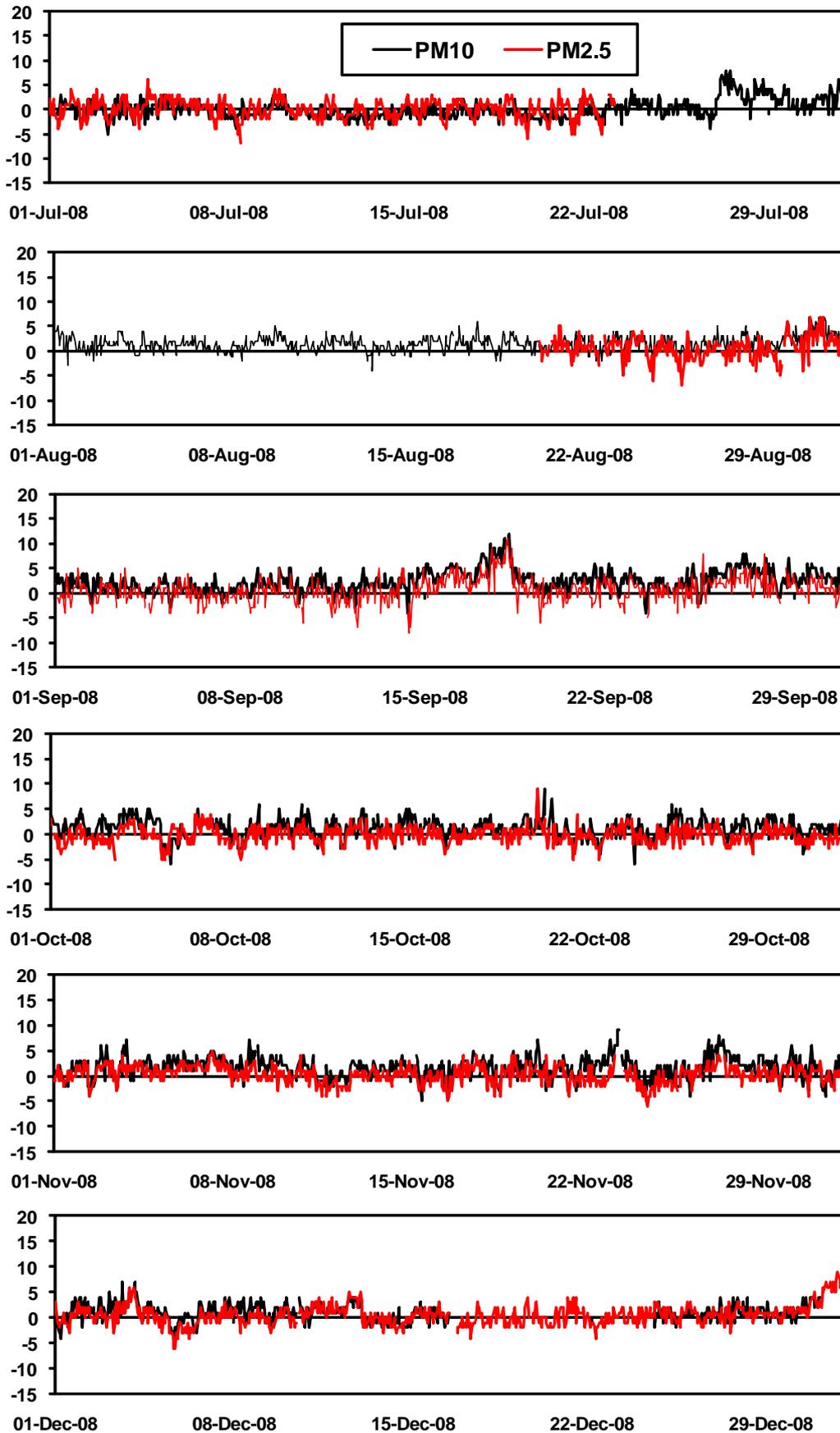
Non-volatile particulate matter (TEOM/FDMS) $\mu\text{g m}^{-3}$



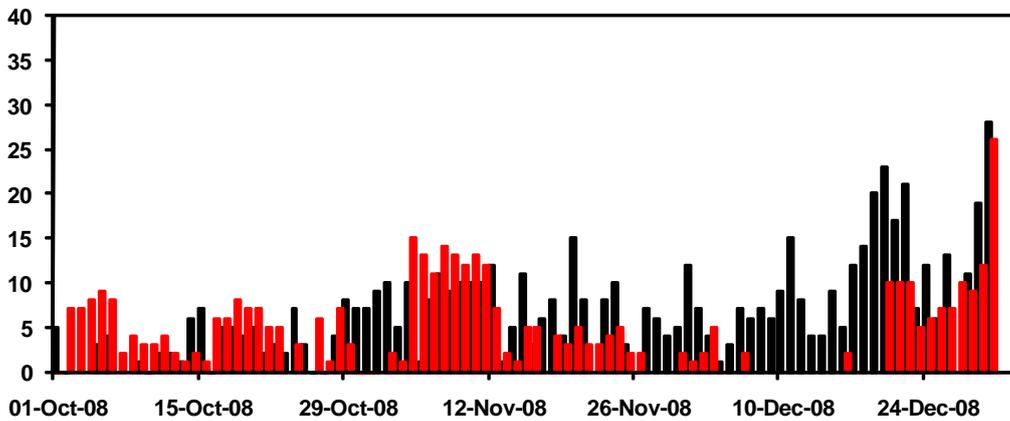
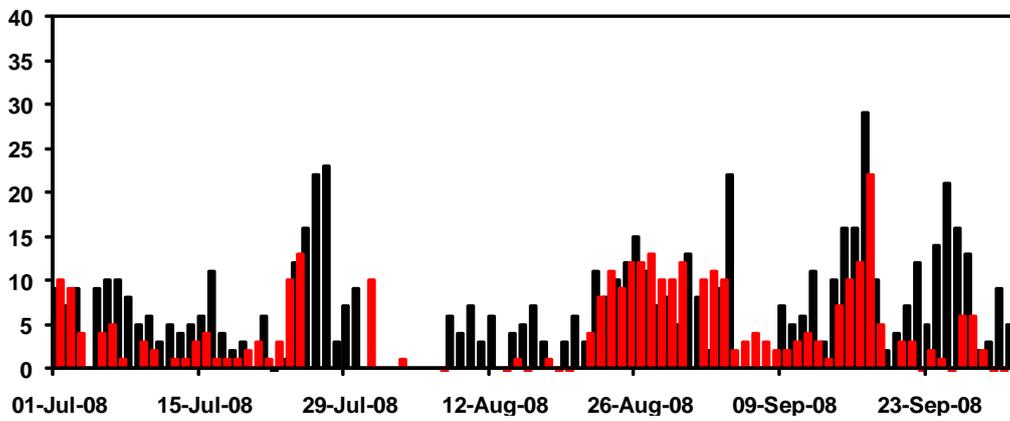
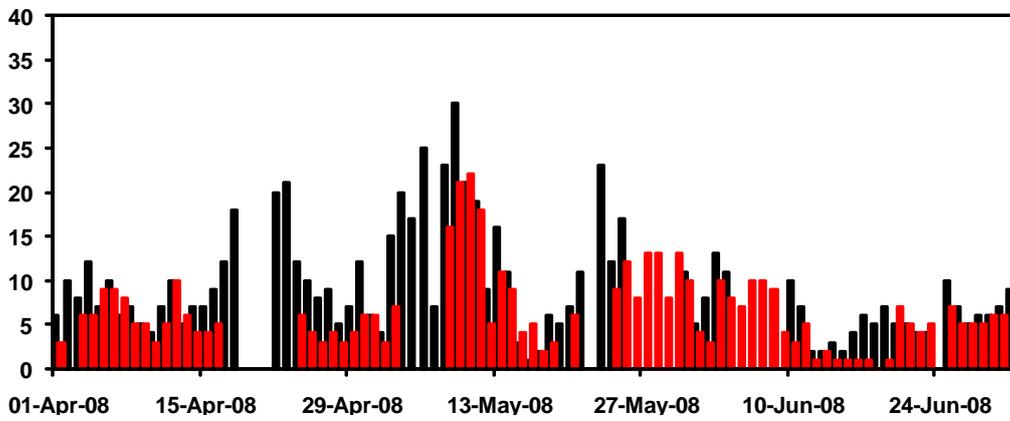
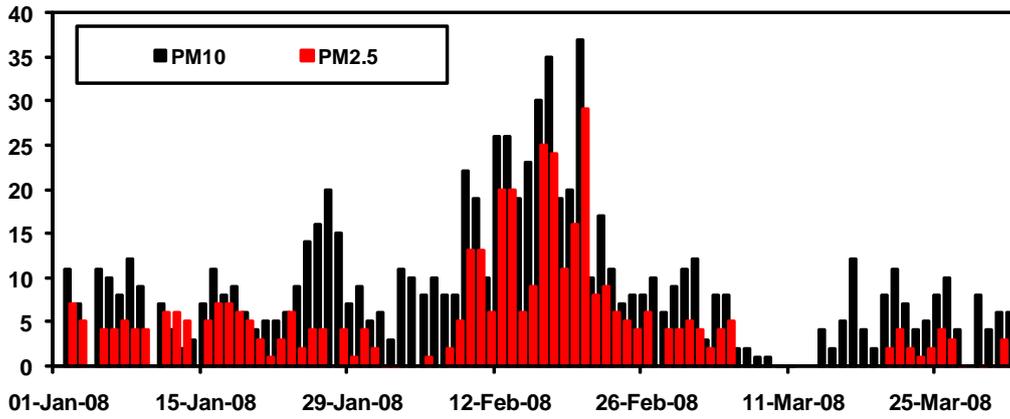
Volatile particulate matter (TEOM/FDMS) $\mu\text{g m}^{-3}$



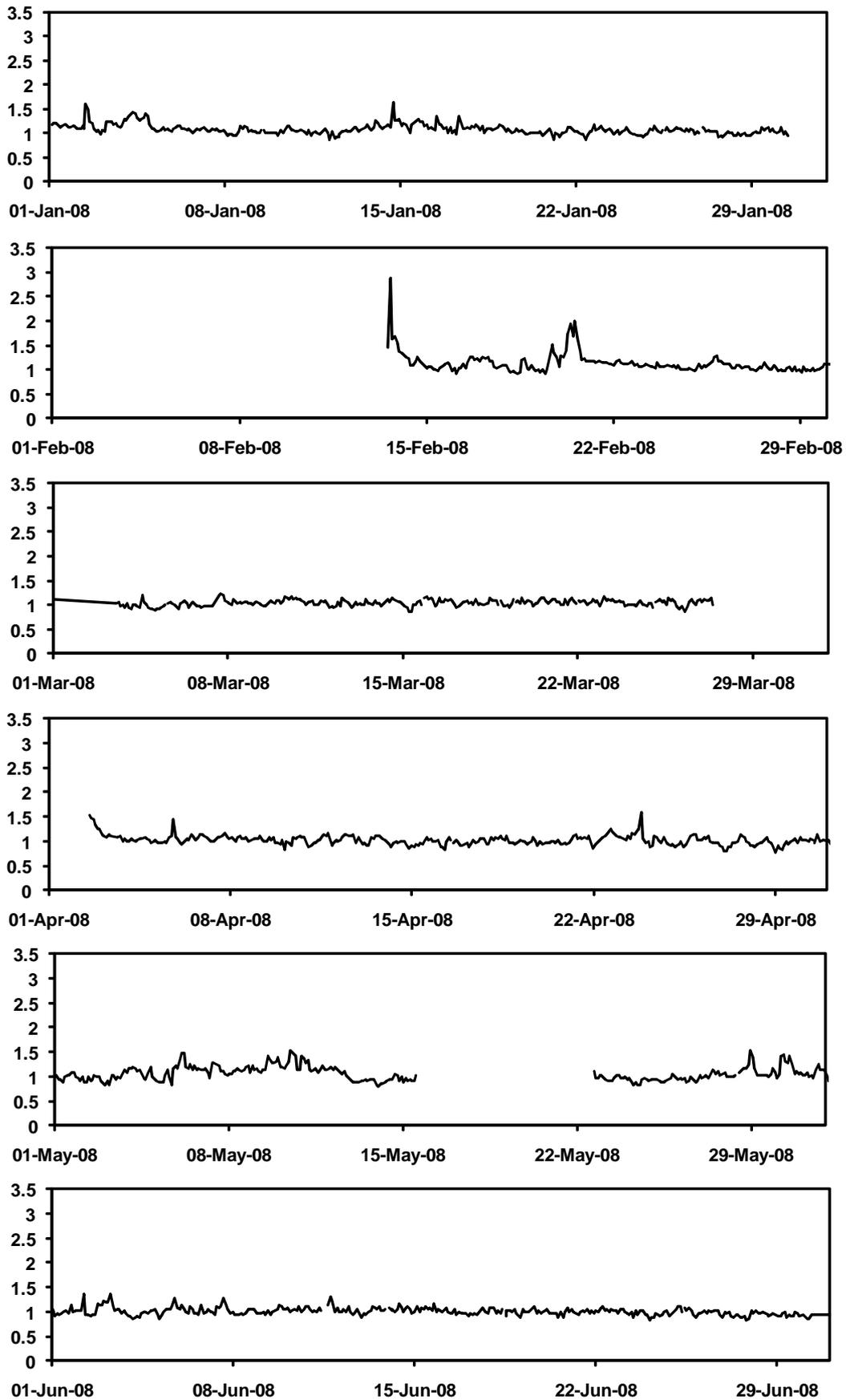
Volatile particulate matter (TEOM/FDMS) $\mu\text{g m}^{-3}$



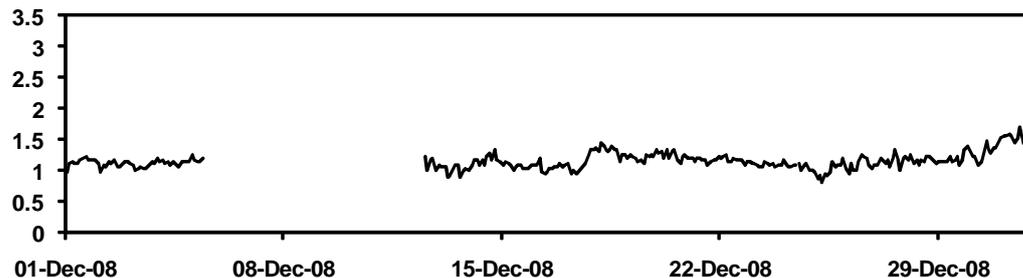
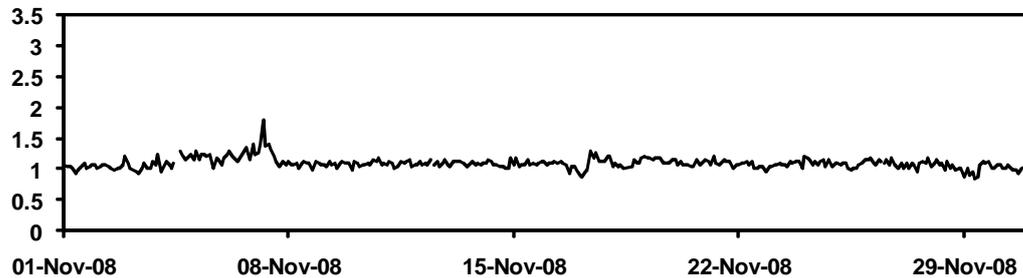
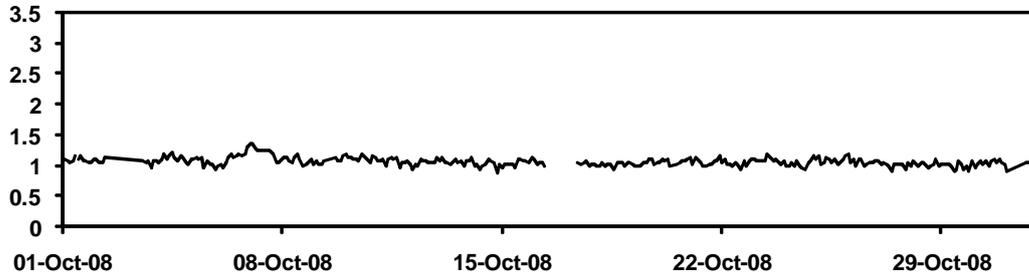
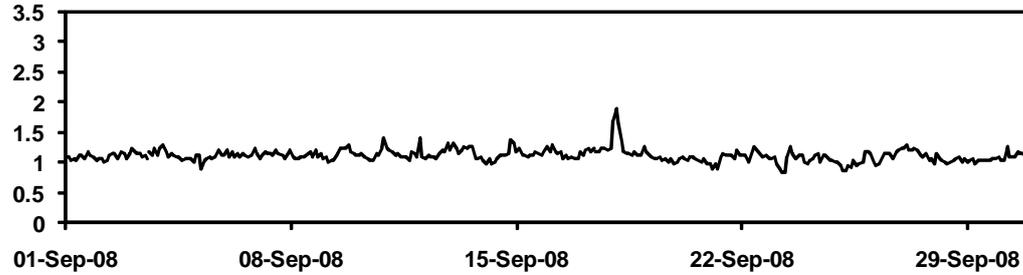
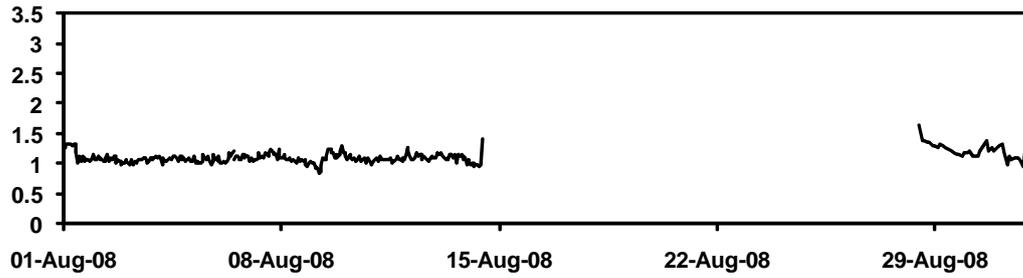
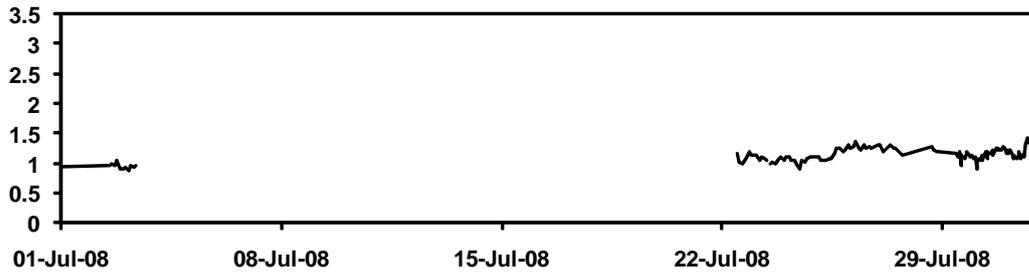
Particulate matter, $\mu\text{g m}^{-3}$



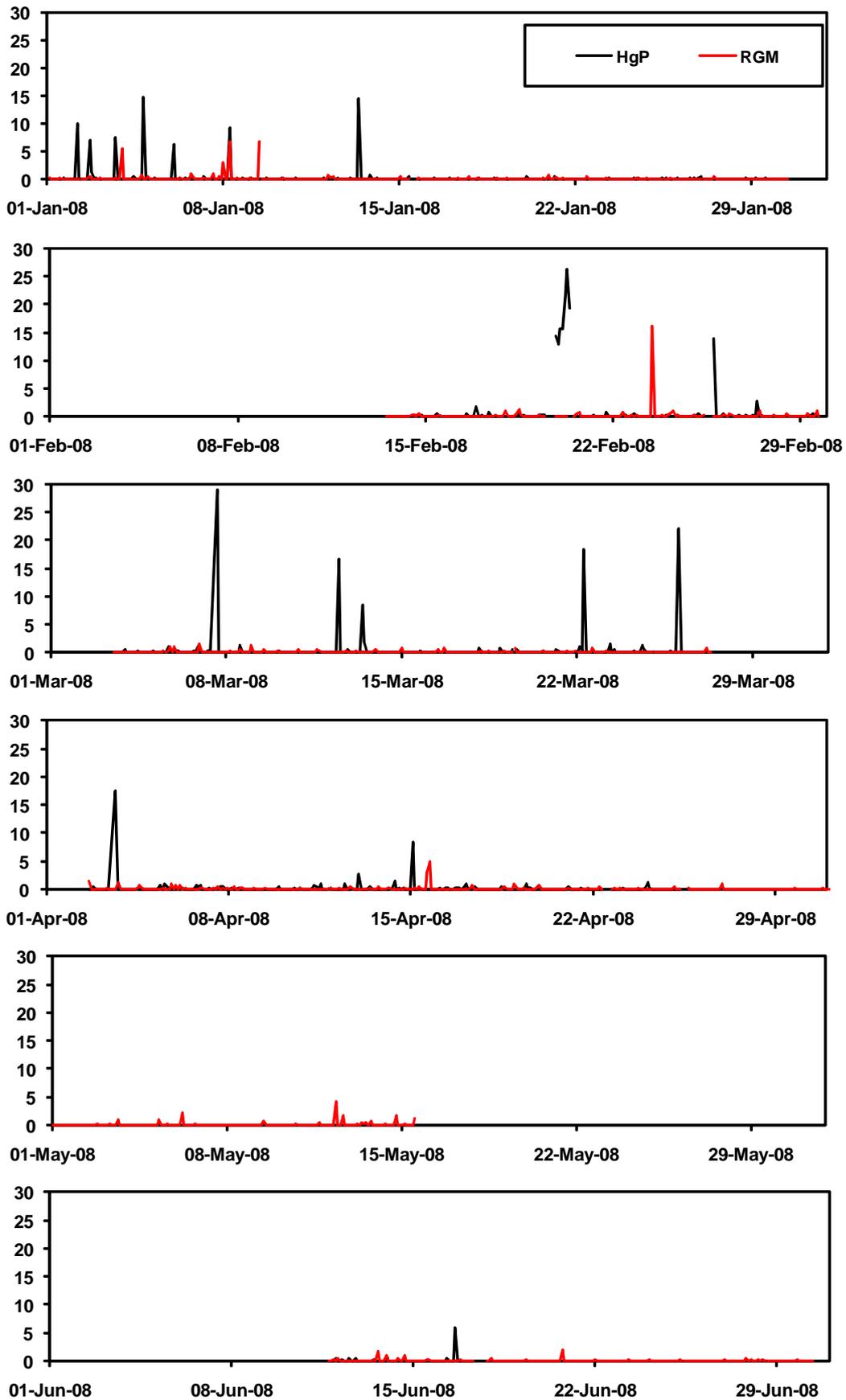
Elemental mercury (Hg^0) ng m^{-3}



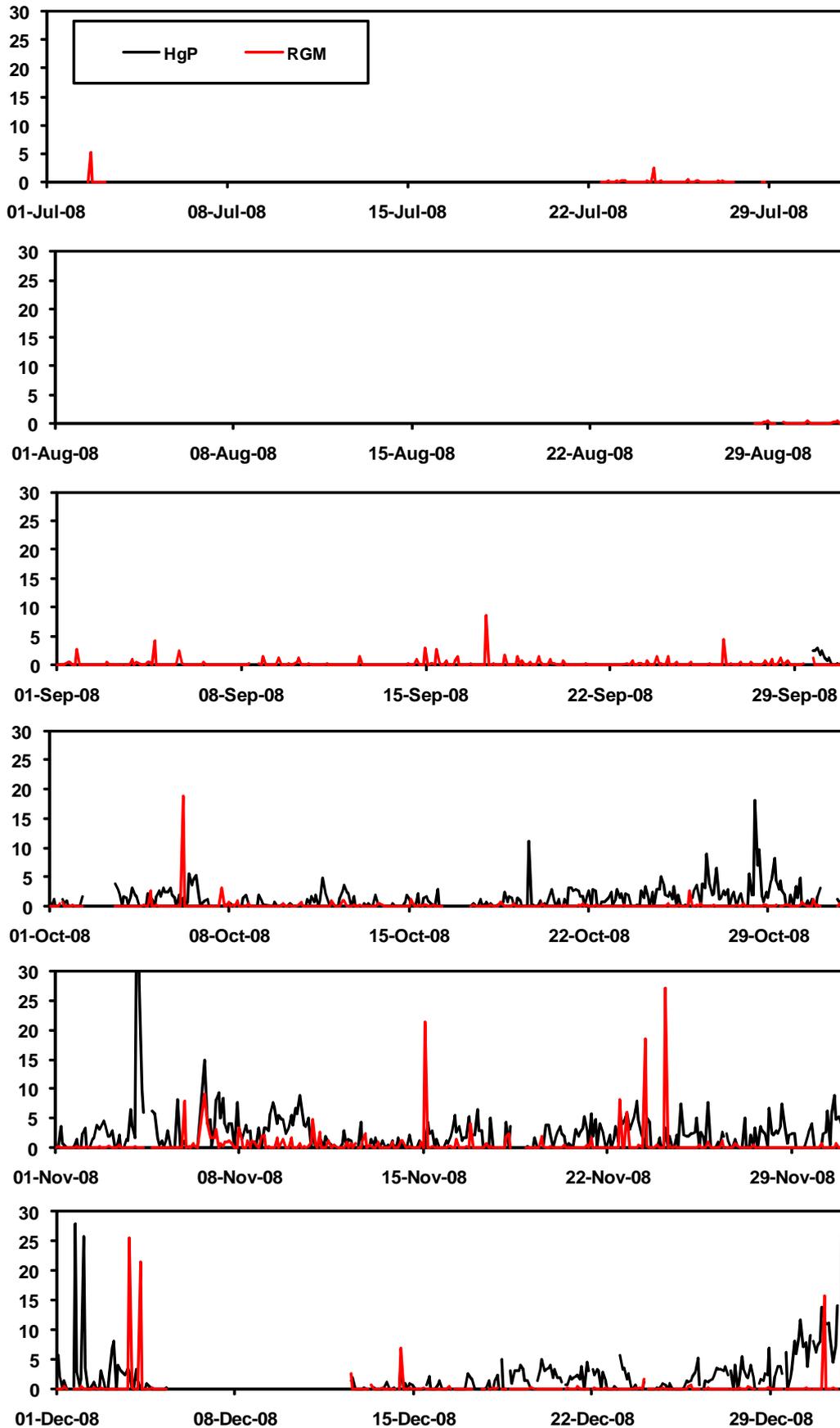
Elemental mercury (Hg^0) ng m^{-3}



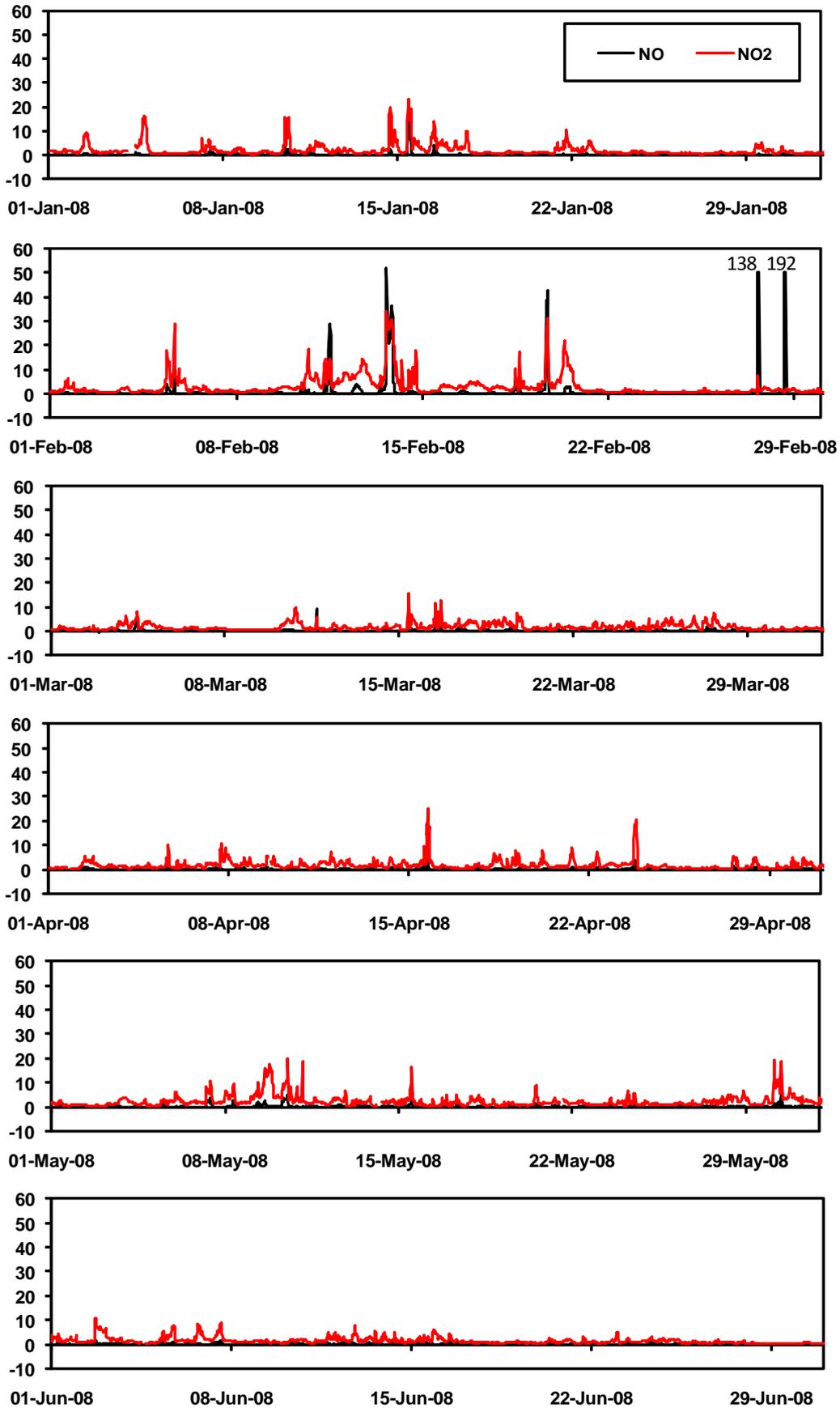
Speciated mercury pg m⁻³



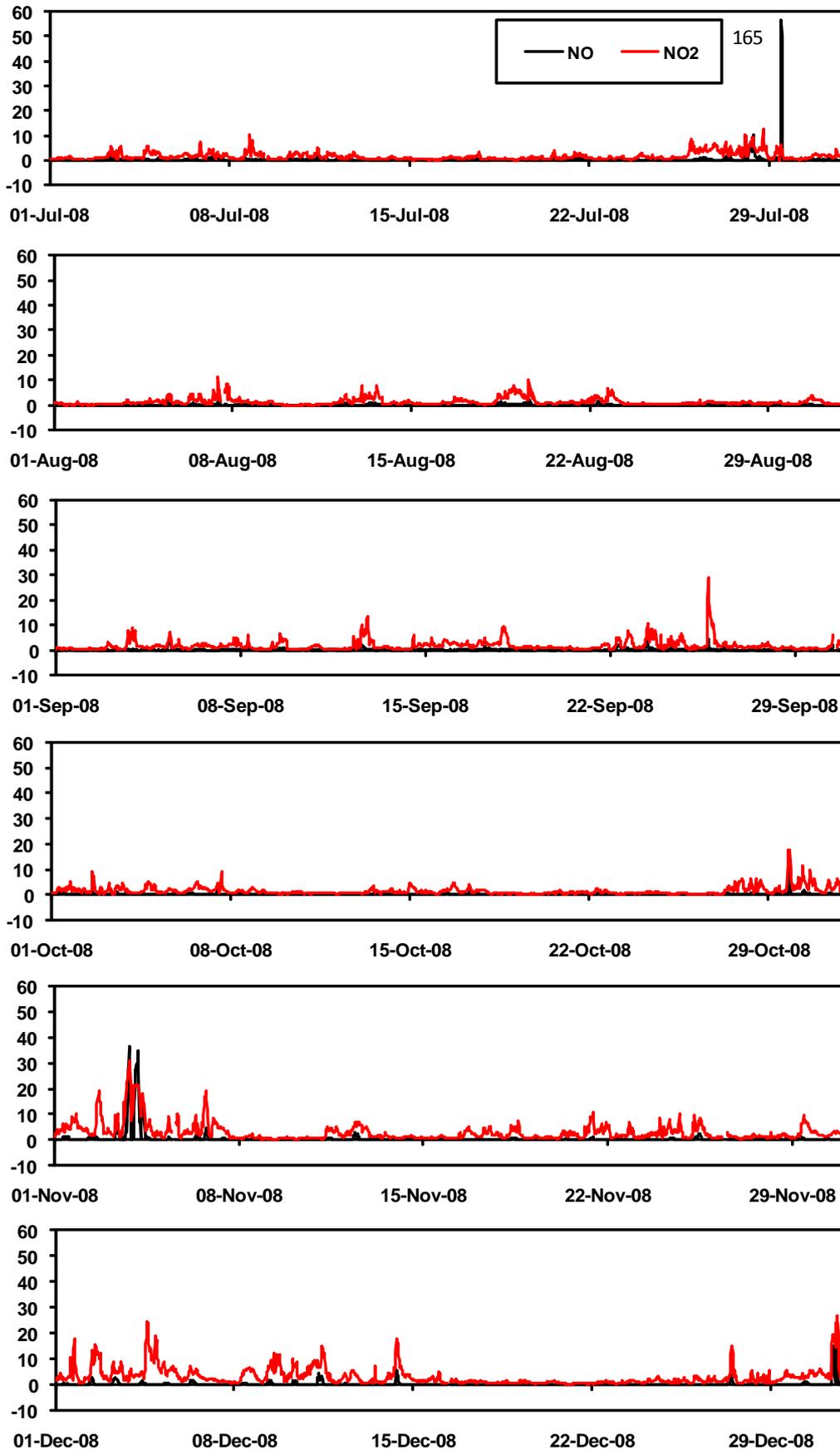
Speciated mercury pg m⁻³



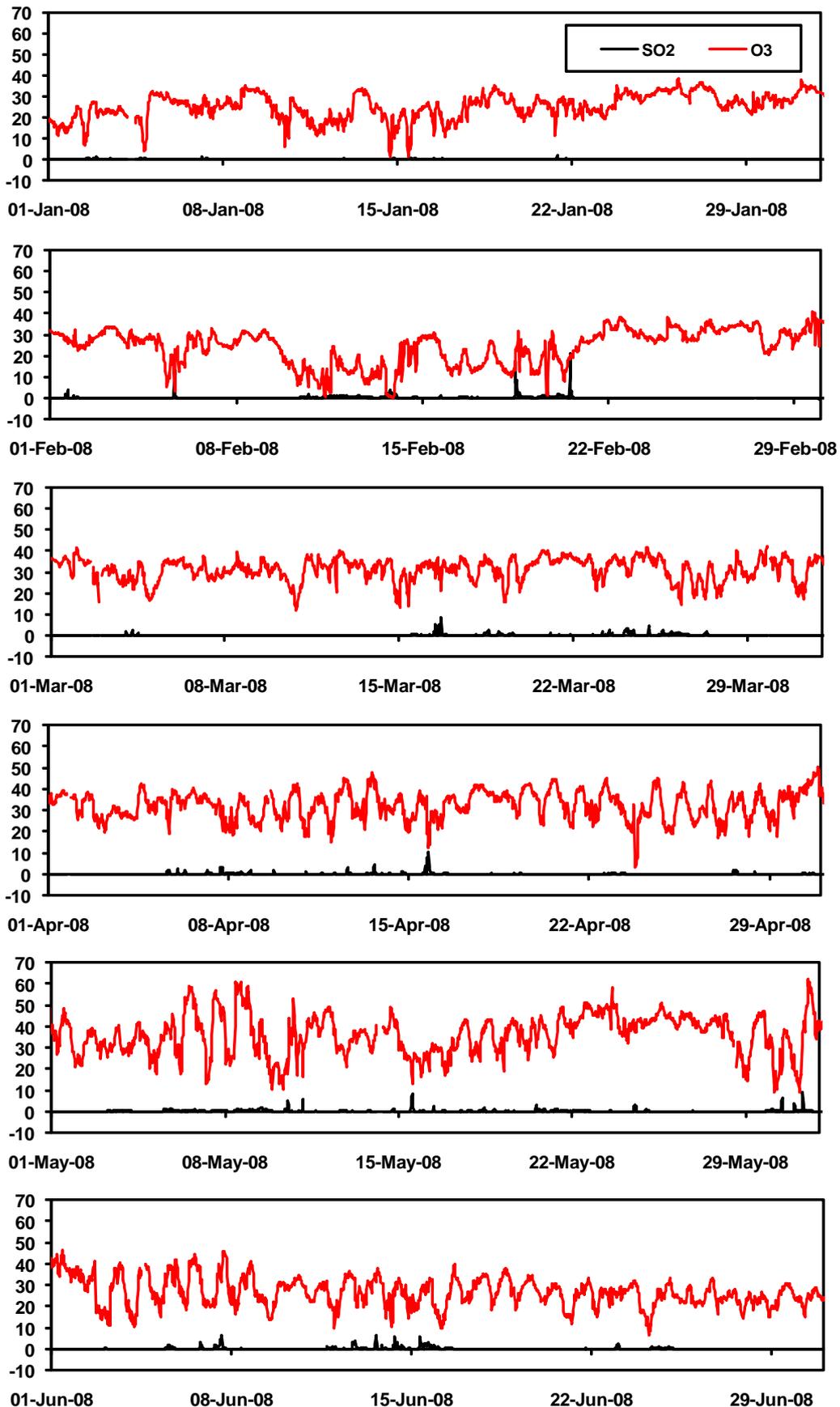
Nitric oxide/nitrogen dioxide (ppb)



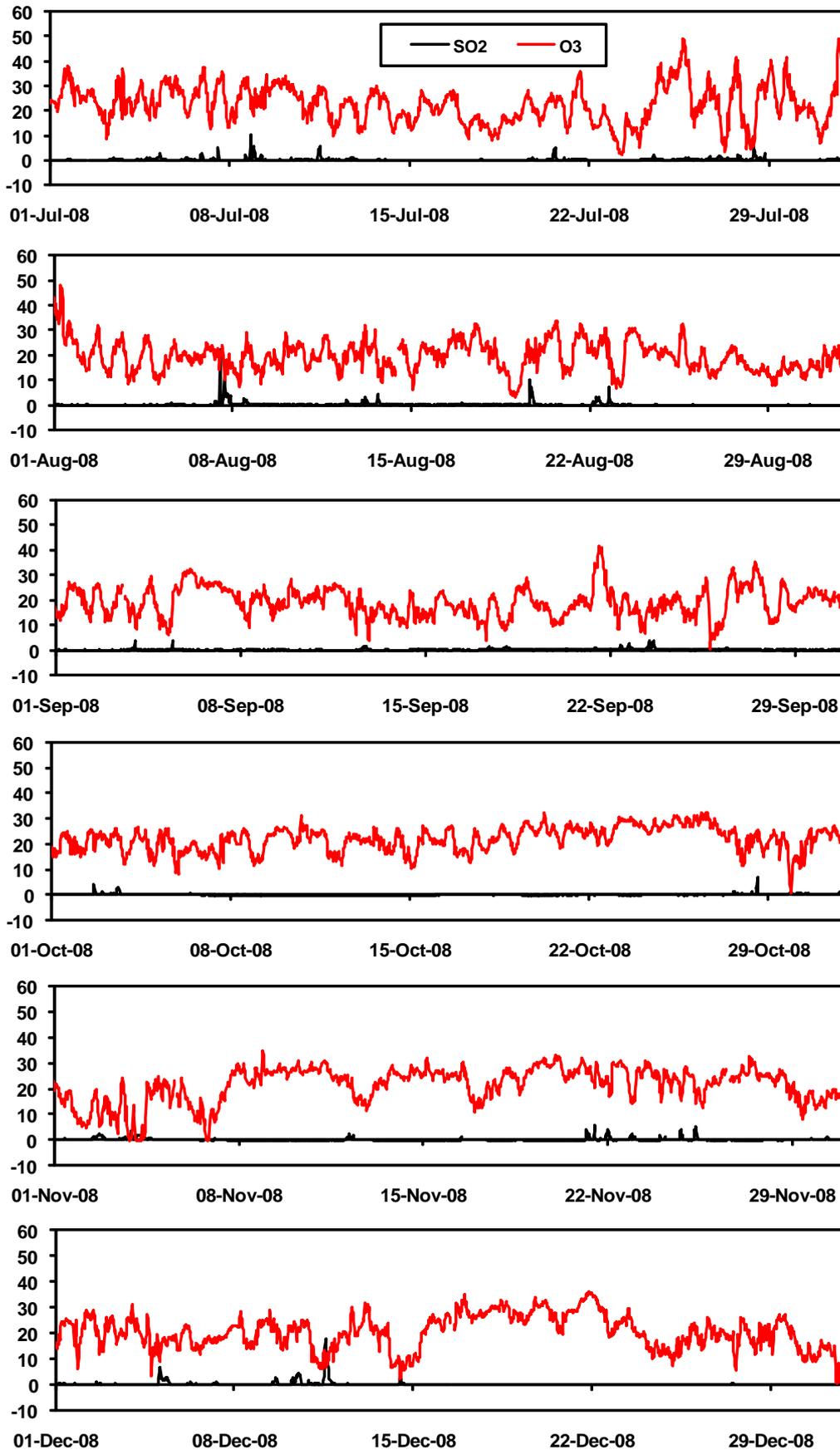
Nitric oxide/nitrogen dioxide (ppb)



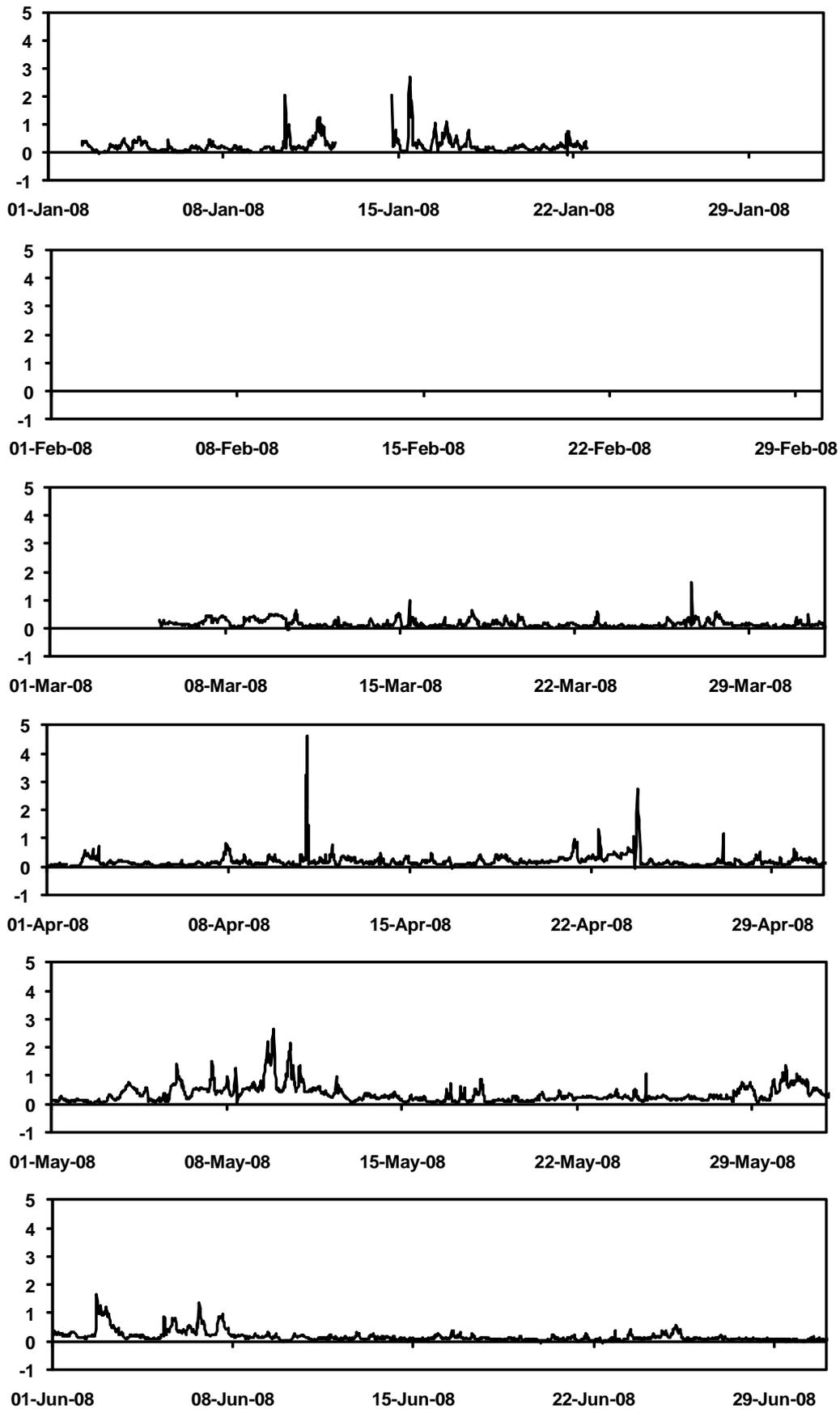
Sulphur dioxide/ozone (ppb)



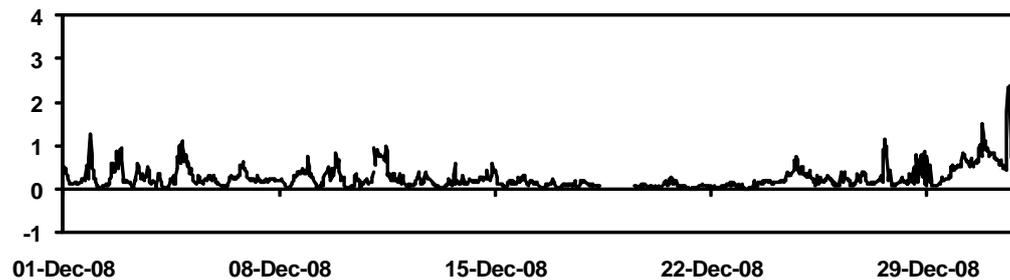
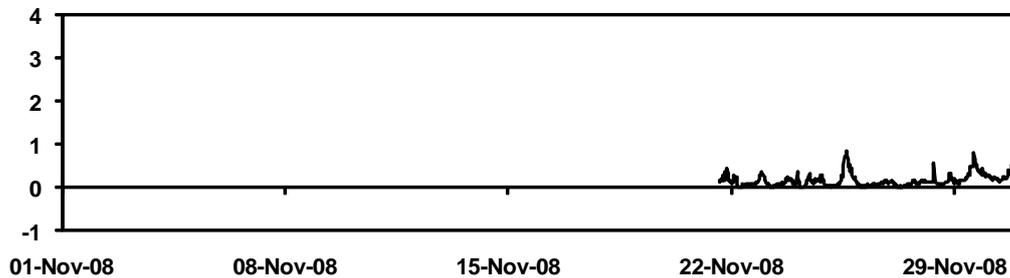
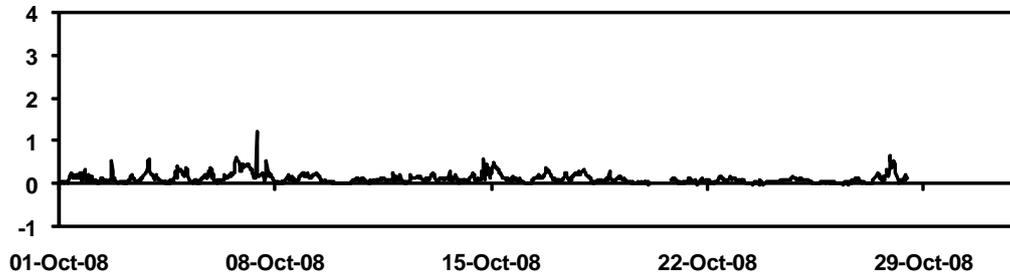
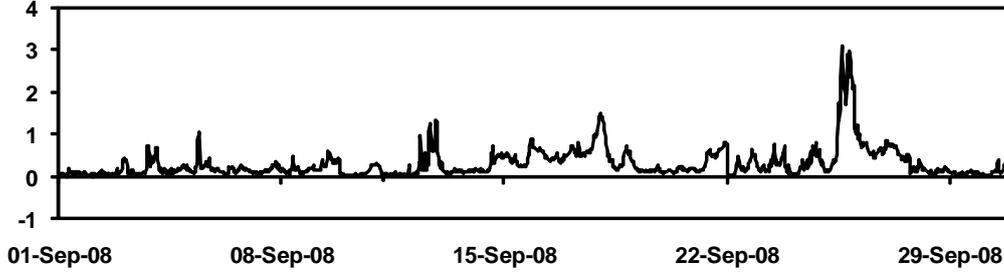
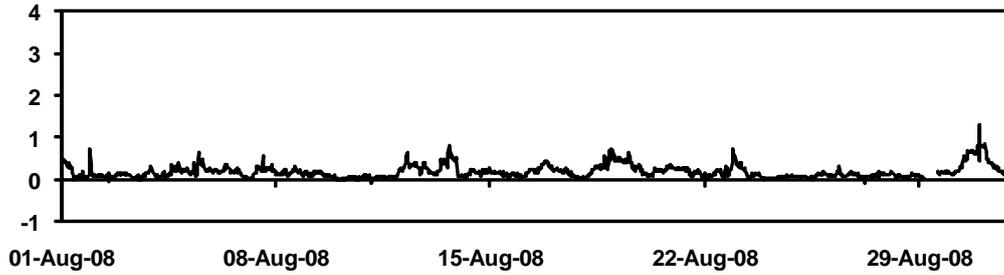
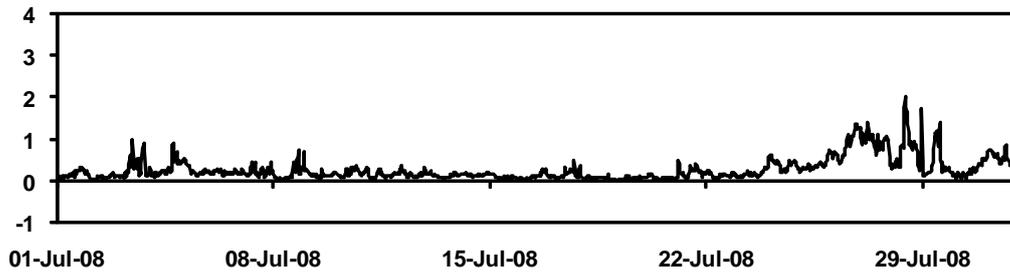
Sulphur dioxide/ozone (ppb)



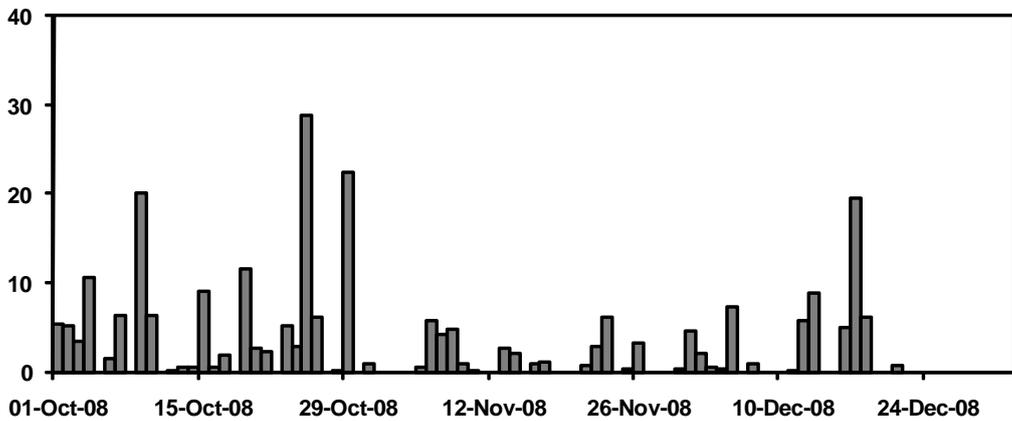
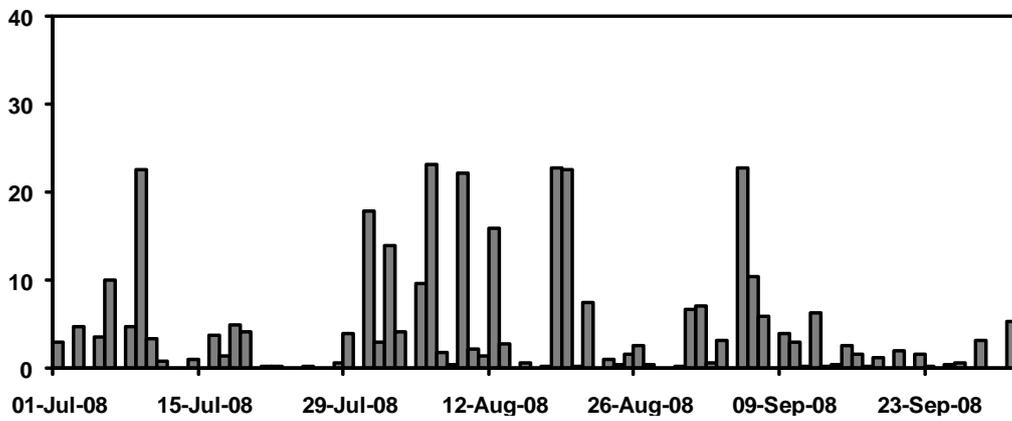
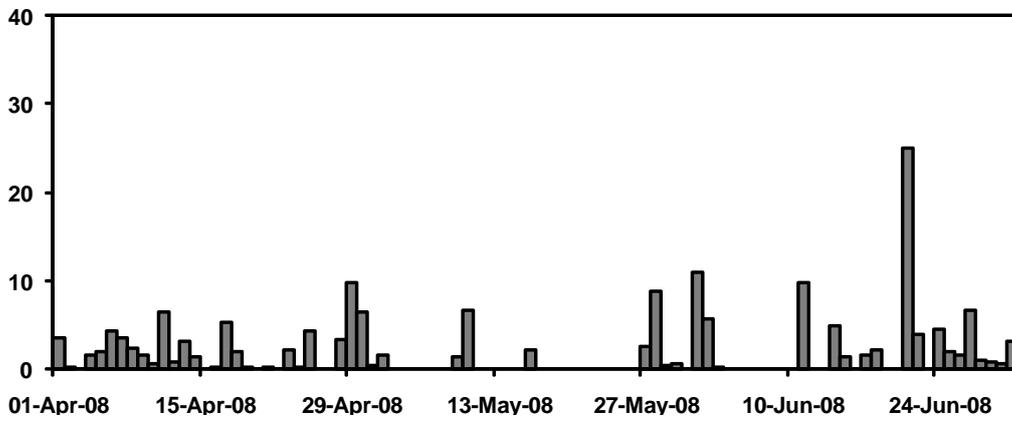
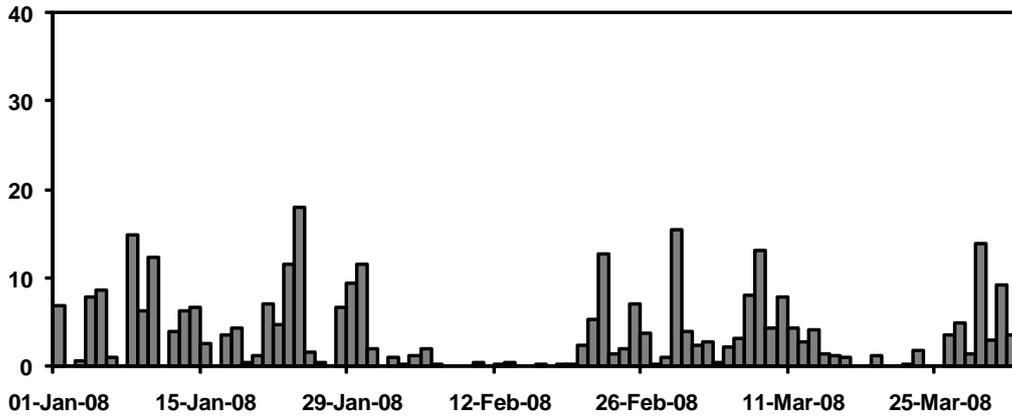
Black carbon (aethalometer) $\mu\text{g m}^{-3}$



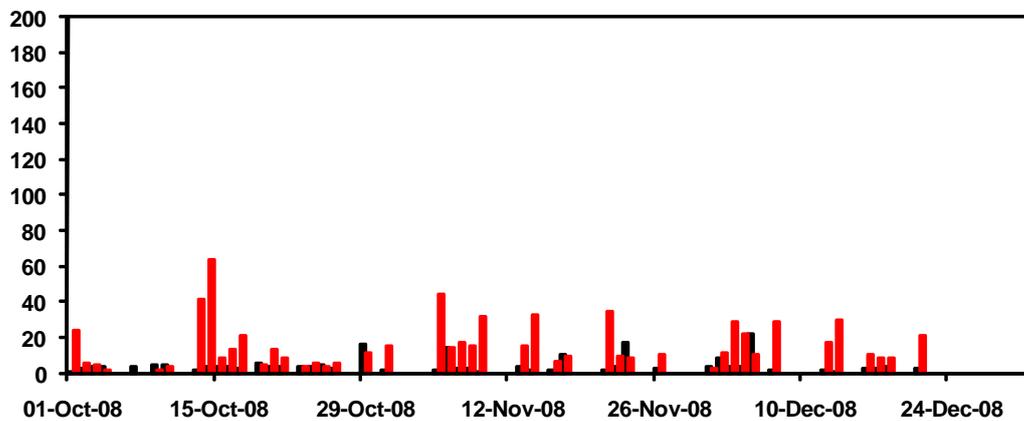
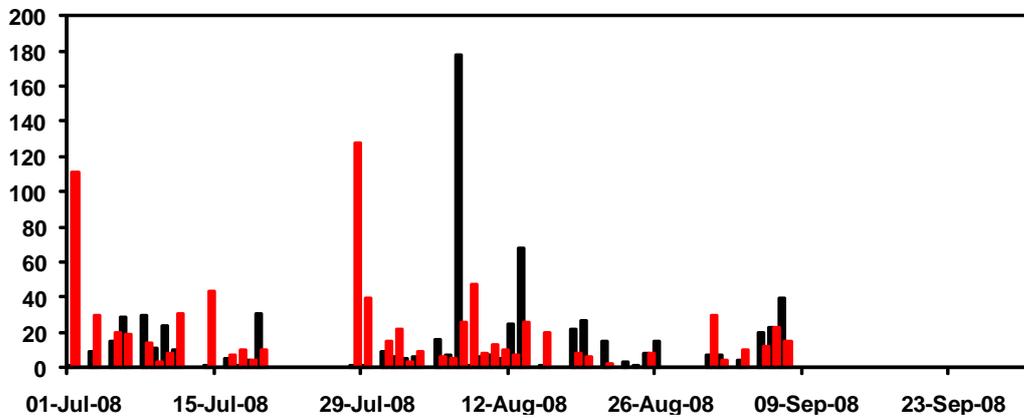
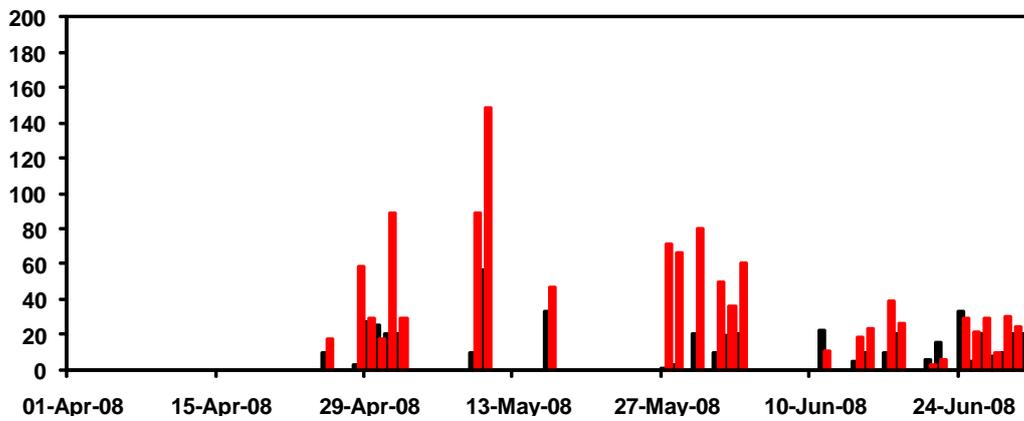
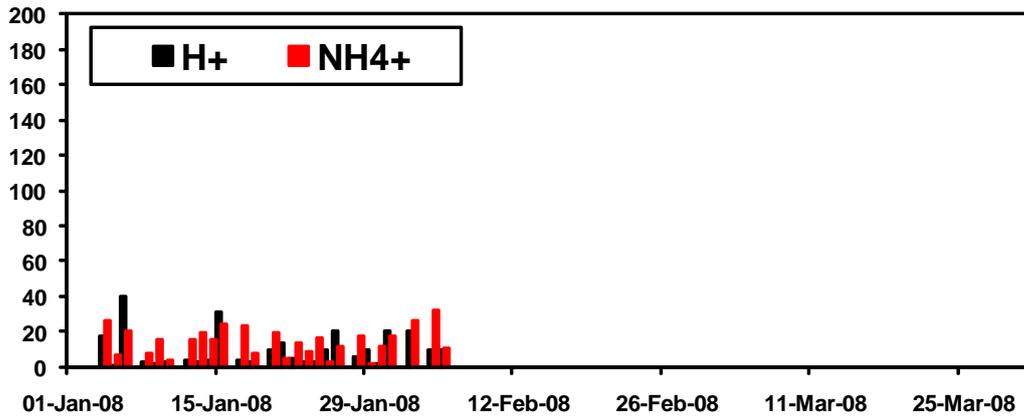
Black carbon (aethalometer) $\mu\text{g m}^{-3}$



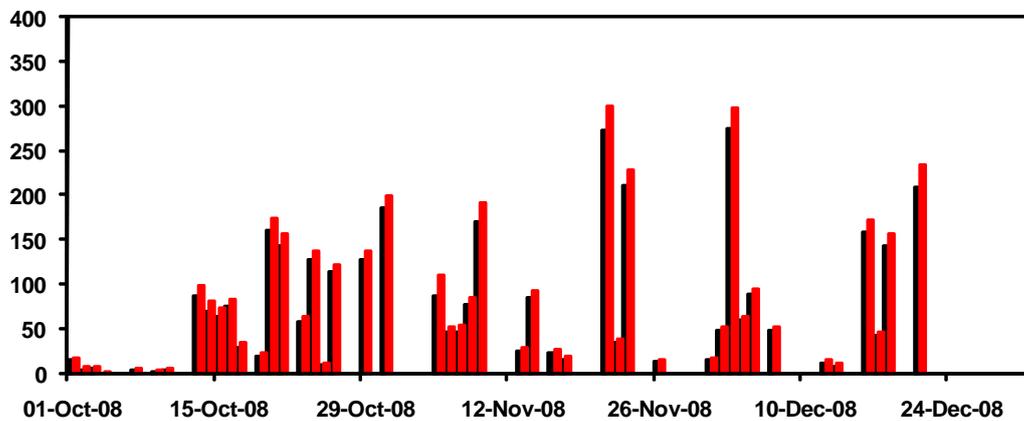
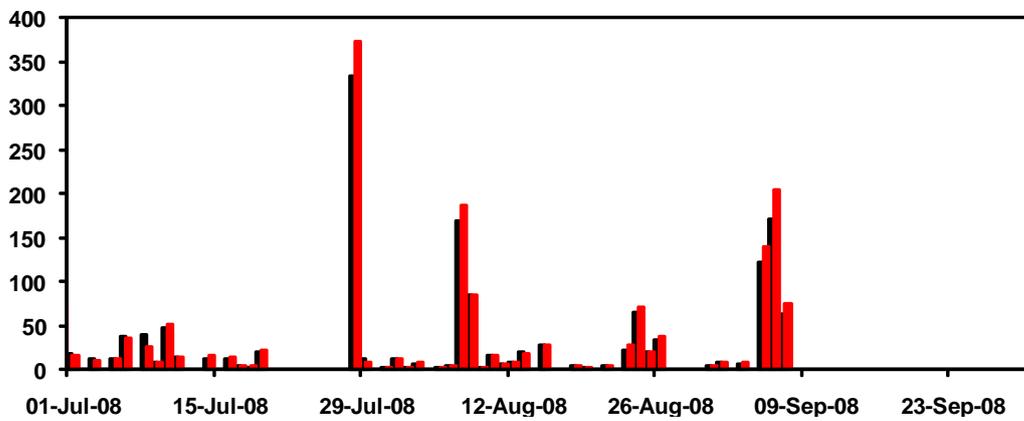
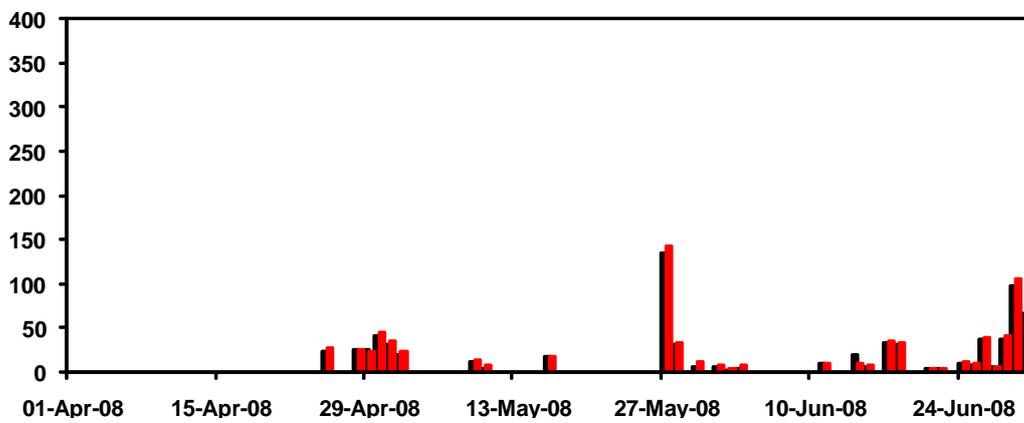
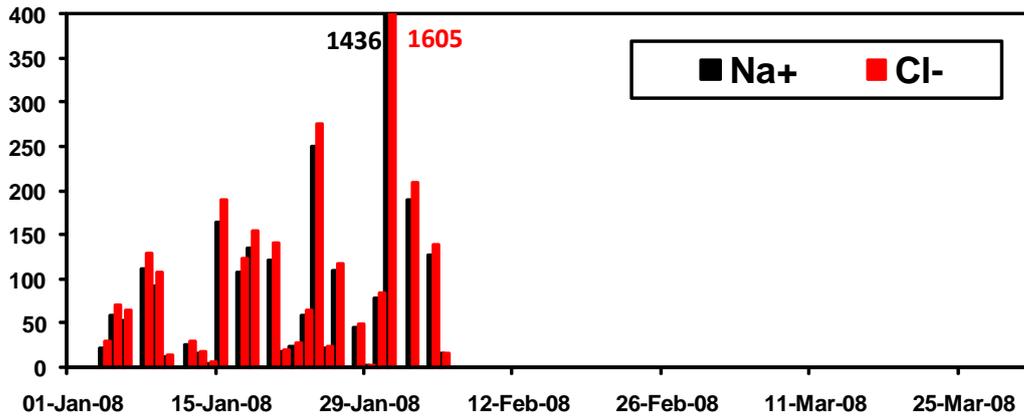
Daily rainfall (mm)



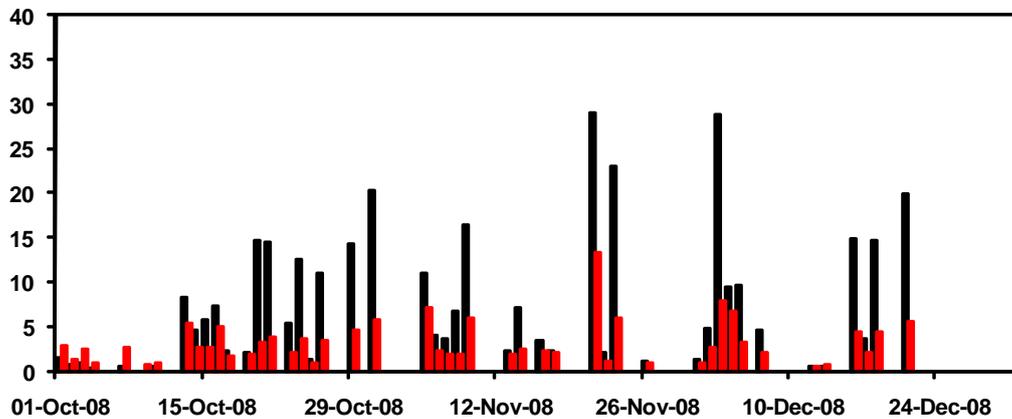
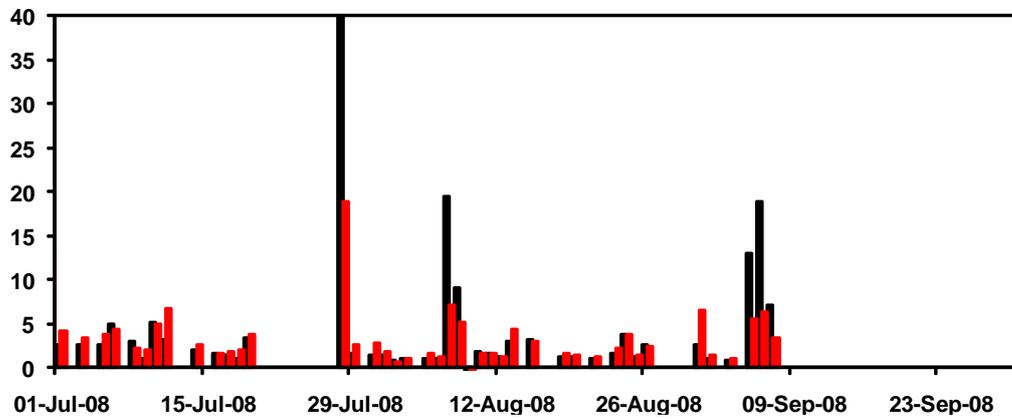
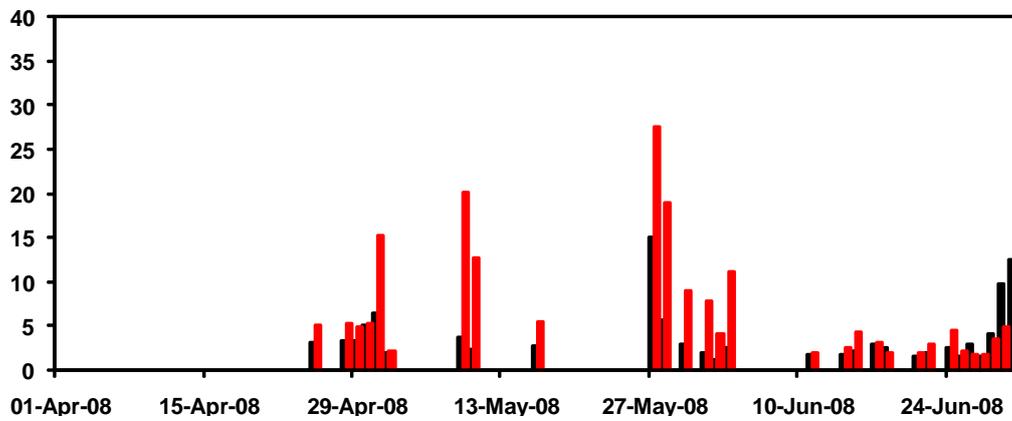
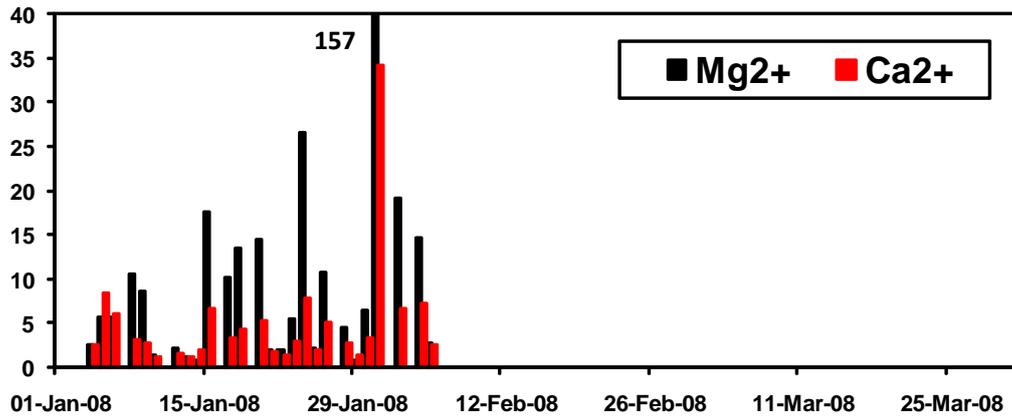
Wet-only rain concentrations (daily) H⁺ and NH₄⁺ μM



Wet-only rain concentrations (daily) Na⁺ and Cl⁻ μM



Wet-only rain concentrations (daily) Mg²⁺ and Ca²⁺ µM



Wet-only rain concentrations (daily) SO_4^{2-} and NO_3^- μM

