



ENVIRONMENTAL
MONITORING

M-1124 | 2018

Monitoring of greenhouse gases and aerosols at Svalbard and Birkenes in 2017

Annual report



COLOPHON

Executive institution

NILU - Norsk institutt for luftforskning
P.O. Box 100, 2027 Kjeller

ISBN: 978-82-425-2952-7 (electronic)
ISSN: 2464-3327

Project manager for the contractor

Cathrine Lund Myhre

Contact person in the Norwegian Environment Agency

Øystein Tennfjord

M-no

1124

Year

2018

Pages

143

Contract number

17078061

Publisher

NILU - Norsk institutt for luftforskning
NILU report 29/2018
NILU project no. O-99093/O-105020/O-113007

The project is funded by

Norwegian Environment Agency and
NILU - Norwegian Institute for Air Research.

Author(s)

C.L. Myhre, T. Svendby, O. Hermansen, C. Lunder, S. M. Platt, M. Fiebig, A.M. Fjæraa, G. Hansen, N. Schmidbauer, T. Krognæs, S.E. Walker

Title - Norwegian and English

Monitoring of greenhouse gases and aerosols at Svalbard and Birkenes in 2017 - Annual report
Overvåking av klimagasser og partikler på Svalbard og Birkenes i 2017: Årsrapport

Summary - sammendrag

The report summarizes the activities and results of the greenhouse gas monitoring at the Zeppelin Observatory situated on Svalbard in Arctic Norway during the period 2001-2017, and the greenhouse gas monitoring and aerosol observations from Birkenes for 2009-2017.

Rapporten presenterer aktiviteter og måleresultater fra klimagassovertvåkingen ved Zeppelin observatoriet på Svalbard for årene 2001-2017 og klimagassmålinger og klimarelevante partikkelmålinger fra Birkenes for 2009-2017.

4 emneord

Drivhusgasser, partikler, klimagasser
halokarboner

4 subject words

Greenhouse gases, aerosols, climate gases,
halocarbons

Front page photo

Kjetil Tørseth, NILU

Preface

This report presents the 2017 annual results from the national monitoring of greenhouse gas concentrations and climate-relevant aerosol (particle) properties. The observations are done at two atmospheric supersites; one regional background site in southern Norway and one Arctic site. The observations made are part of the national monitoring programme conducted by NILU- Norsk institutt for luftforskning (NILU) on behalf of The Norwegian Environment Agency. Additionally, the report includes results from Trollhaugen observatory in Antarctica. These measurements are not a part of the national monitoring programme, but receive direct support from Ministry of Climate and Environment.

The national monitoring programme comprises measurements of 46 greenhouse gases at the Zeppelin Observatory in the Arctic; and this includes a long list of halocarbons, which are not only greenhouse gases, but most of them are also ozone depleting substances. NILU initiated measurements of CH₄-isotope (of $\delta^{13}\text{C}_{\text{CH}_4}$) at Zeppelin in the year 2012 to provide more insight into Arctic sources of CH₄, and potential changes. The continuation of these measurements was implemented in the Norwegian Monitoring programme, and the results are included for the first time. NILU upgraded and extended the observational activity at the Birkenes Observatory in Aust-Agder and from 2010, the national monitoring programme was extended to include aerosol properties (size, number, scattering and absorption properties) relevant for understanding the effects of aerosols on radiation.

The present report is the third of a series of annual reports in 2018, which cover the national monitoring of atmospheric composition in the Norwegian rural background environment. The other three reports focus on the atmospheric composition and deposition of air pollution of particulate and gas phase of inorganic constituents, particulate carbonaceous matter, ground level ozone and particulate matter, the second presents the monitoring of the ozone layer and UV.

Participation in international programmes is crucial for quality assurance and quality control of the Norwegian measurement data and instruments. Data from this report are contributing to, and benefiting from, the European Research Infrastructure ACTRIS (*Aerosols, Clouds, and Trace gases Research InfraStructure*) and ICOS (*Integrated Carbon Observation System*). Additionally, data and results support and benefit from EMEP (European Monitoring and Evaluation Programme) under CLTRAP (Convention on Long-range Transboundary Air Pollution), and AGAGE (Advanced Global Atmospheric Gases Experiment).

All measurement data presented in the current report are public and can be received by contacting NILU, or they can be downloaded directly from the database: <http://ebas.nilu.no>.

A large number of persons at NILU have contributed to the current report, including those responsible for sampling, technical maintenance, chemical analysis and quality control and data management, in particular, Tove Svendby, Ove Hermansen, Chris Lunder, Markus Fiebig, Georg Hanssen, Stephen M. Platt, Terje Krognnes, Ann Mari Fjæraa Norbert Schmidbauer, and Sam E. Walker.

NILU, Kjeller, 1 October 2018

Cathrine Lund Myhre
Senior Scientist, Dep. Atmospheric and Climate Research

Content

Preface	2
Sammendrag (Norwegian)	5
Summary.....	7
1 Introduction to monitoring of greenhouse gases and aerosols.....	11
1.1 The monitoring programme in 2017	11
1.2 The measurements in relation to research and policy agreements	12
1.3 The ongoing monitoring programme and the link to networks and research infrastructures	13
1.4 Greenhouse gases, aerosols and their climate effects	18
1.4.1 Revisions of forcing estimates of carbon dioxide, methane, and nitrous oxide	22
2 Observations of climate gases.....	24
2.1 Climate gases with natural and anthropogenic sources	27
2.1.1 Carbon dioxide (CO ₂).....	27
2.1.2 Methane (CH ₄).....	30
2.1.3 Nitrous Oxide (N ₂ O)	38
2.1.4 Volatile organic compounds (VOC)	39
2.1.5 Carbon monoxide (CO)	44
2.1.6 Chloromethane at the Zeppelin Observatory.....	46
2.1.7 Bromomethane - CH ₃ Br at the Zeppelin Observatory	47
2.2 Greenhouse gases with solely anthropogenic sources	50
2.2.1 Chlorofluorocarbons (CFCs) at Zeppelin Observatory	50
2.2.2 Hydrochlorofluorocarbons (HCFCs) at Zeppelin Observatory	53
2.2.3 Hydrofluorocarbons (HFCs) at Zeppelin Observatory	55

2.2.4	Halons measured at Zeppelin Observatory.....	59
2.2.5	Other chlorinated hydrocarbons at Zeppelin Observatory	61
2.2.6	Perfluorinated compounds at Zeppelin Observatory	64
2.3	Summary of measurements of climate gases.....	68
3	Aerosols and climate.....	71
3.1	Observed optical properties of aerosols.....	75
3.1.1	Optical aerosol properties measured at the Birkenes Observatory.....	75
3.1.2	Optical aerosol properties measured the Zeppelin Observatory.....	79
3.1.3	Optical aerosol properties measured at the Trollhaugen Observatory	81
3.2	Measurements of aerosol number and size	84
3.2.1	Physical aerosol properties measured at the Birkenes Observatory.....	84
3.2.2	Physical aerosol properties measured in situ at the Zeppelin Observatory	86
3.2.3	Physical aerosol properties measured in situ at the Trollhaugen Observatory ..	88
3.3	Summary of physical and optical aerosol properties.....	91
3.4	Column optical aerosol properties measured by ground-based remote sensing.....	95
3.4.1	Column optical aerosol properties measured by ground-based remote sensing at Birkenes Observatory.....	95
3.4.2	Column optical aerosol properties measured by ground-based remote sensing at Ny-Ålesund	99
4	References	103
	APPENDIX I: Data Tables.....	109
	APPENDIX II Description of instruments and methodologies.....	121
	APPENDIX III: Abbreviations.....	139

Sammendrag (Norwegian)

Denne årsrapporten beskriver aktivitetene og hovedresultatene fra delprogrammet "Overvåking av klimagasser og aerosoler på Zeppelin-observatoriet på Svalbard og Birkenes-observatoriet i Aust-Agder, Norge". Rapporten omfatter målinger av 46 klimagasser og inkluderer de viktigste naturlig forekommende drivhusgassene, syntetiske klimagasser og ulike partikkelegenskaper som har høy relevans for stråling og klima. Mange av gassene har også sterk ozonnedbrytende effekt. For de fleste klimagassene er utvikling og trender for perioden 2001-2017 rapportert, i tillegg til daglige og årlige gjennomsnittsverdier. Programmet ble utvidet med 16 nye gasser i 2015, og videre med seks nye gasser i 2016. For de nye komponentene er data innhentet og analysert tilbake til 2010. Resultatene er oppsummert i tabell 1 på side 9.

Målingene på Zeppelin-observatoriet gir informasjon om utviklingen i bakgrunnsnivåkonsentrasjonene av klimagasser i Arktis, og alle de 46 gassene i overvåkningsprogrammet blir målt her. Birkenes-observatoriet ligger i et område i Sør-Norge som kan være sterkt påvirket av langtransportert luftforurensning, i tillegg til påvirkning fra lokal vegetasjon. På Birkenes gjøres observasjoner av karbondioksid (CO₂) og metan (CH₄), samt omfattende målinger av klimarelevante egenskaper til aerosoler (partikler). Aerosoler måles også på Zeppelin og Trollhaugen (Antarktis), men måleprogrammene her er ikke like omfattende som på Birkenes.

Konsentrasjonene av alle de viktigste klimagassene har økt kraftig siden 2001, bortsett fra ozonnedbrytende klorfluorkarboner (KFK-er) og noen få halogenerte gasser som er regulert gjennom Montrealprotokollen. De fleste av disse har hatt en reduksjon, som vist i rapporten.

Observasjonene fra 2017 viser nye rekordhøye nivåer for de fleste klimagassene i overvåkningsprogrammet. Årlig midlere CO₂ konsentrasjon i 2017 var 408.0 ppm (parts per million) på Zeppelin og 411.3 ppm på Birkenes. Det er økninger på henholdsvis 3.6 ppm (0.89%) og 1.5 ppm (0.37%) fra 2016.

Konsentrasjonene av metan var også rekordhøye i 2017, med årlige middelerverdier på 1939 ppb på Zeppelin og 1945 ppb på Birkenes. I forhold til 2016-nivået representerer dette økninger på 7 ppb (0.35%) og 3 ppb (0.18%) på henholdsvis Zeppelin og Birkenes. Endringene i løpet av de siste ti årene er store i forhold til utviklingen av metannivået i perioden 1998-2005, da endringene var tilnærmet null både på Zeppelin og globalt. Ulike metankilder (for eksempel naturgass og våtmarker) har ulik isotopsammensetning. Isotopmålinger av CH₄ på Zeppelin viser en klar trend i $\delta^{13}\text{C}_{\text{CH}_4}$ etter 2012. En negativ trend i $\delta^{13}\text{C}_{\text{CH}_4}$, samt en økning av metankonsentrasjonene, tyder på at det har vært endringer i metankildene eller opptaksprosessene i løpet av de siste 6 årene. Endringene i $\delta^{13}\text{C}_{\text{CH}_4}$ på Zeppelin tyder på økte utslipp fra biosfæren og/eller jordbruk. En annen forklaring kan være endringer av opptaket.

Lystgass (N₂O) har store menneskeskapt kilder (som jordbruk og bergverk), men også naturlige kilder. N₂O nådde også nytt rekordnivå i 2017. Den årlige middelkonsentrasjonen var på 330.0 ppb, en økning på 0.9 ppb siden 2016.

De syntetiske, menneskeskapt klimagassene som inngår i overvåkningsprogrammet på Zeppelin er fire klorfluorkarboner (KFK-er), tre hydroklorfluorkarboner (HKFK-er), og 11 hydrofluorkarboner (HFK-er), de to sistnevnte gruppene er KFK-erstatninger. I tillegg

inngår tre haloner, en gruppe med åtte andre halogenerte klimagasser, samt fire perfluorerte karboner (PFK-er) med svært høyt globalt oppvarmingspotensial (GWP). Videre rapporteres sulfurylfluorid og nitrogentrifluorid, som begge er ekstremt sterke drivhusgasser.

Generelt, avtar konsentrasjonen av KFK-gasser, mens de øvrige øker da disse ofte er erstatningsgasser for KFK-ene. Det ble nylig oppdaget ukjente og urapporterte utslipp av KFK-11 i Asia, noe som har redusert den markante, negative trenden i de globale målingene av KFK-11. Utviklingen i KFK-gassene gir likevel grunn til optimisme, fordi konsentrasjonen for de fleste av disse gassene er synkende. På den annen side økte konsentrasjonene av KFK-erstatningsstoffene, HKFK-er og HFK-er, i perioden 2001-2017. For HKFK-er har det riktignok vært en avtagende økning de siste årene, mens konsentrasjonen av HFK-gassene har økt kraftig siden 2001, og den økende trenden ser ut til å fortsette. Konsentrasjonene av disse menneskeskapte HFK-ene er fortsatt svært lave, noe som betyr at bidraget til den globale oppvarmingen per i dag er lite, men gitt den ekstremt sterke økningen og bruken av disse gassene, er det viktig å følge utviklingen nøye i fremtiden.

Konsentrasjonene av PFK-er og svovelfluorider (SF_6 og SO_2F_2) er også lave. Et unntak er PFK-14, som hadde en årlig middelværdi på 84.3 ppt i 2017, en økning på 1.0 ppt fra 2016. Konsentrasjonen av SF_6 bør også følges nøye, ettersom denne forbindelsen har en atmosfærisk levetid på 3 200 år og et GWP på hele 23 500. Denne forbindelsen har økt med hele 91% siden målingene startet på Zeppelin i 2001.

Aerosoler er små partikler i atmosfæren. Partiklenes klimapåvirkning avhenger av mengden partikler og absorpsjonsegenskapene til enkeltpartiklene. Konsentrasjonen av partikler på Birkenes bestemmes i hovedsak av den langtransporterte luftforurensningen fra det kontinentale Europa, i tillegg til regionale og lokale vegetasjonskilder (biogene kilder). I 2017 ser vi også en kraftig episode i september som kan spores tilbake til branner i den sydlige russiske steppen. Siden oppstart av målingene på Birkenes i 2010, ser vi ingen klar trend i utviklingen av partikkelegenskapene (antall, absorpsjons- og strålingsspredningsegenskaper), men tidsseriene er foreløpig for korte for pålitelige trendberegninger.

På Zeppelin er situasjonen litt annerledes. Ved å sammenstille våre aerosolmålinger med resultater fra andre samarbeidspartnere, finner vi en nedadgående trend i aerosolabsorpsjonen, noe som viser at konsentrasjonen av sot eller såkalt «*black carbon*» går ned. Observasjoner av den totale kolonnen av aerosoler over Ny-Ålesund (aerosol optisk dybde, AOD) viser økte konsentrasjoner om våren sammenlignet med resten av året. Dette fenomenet, som kalles arktisk dis (*Arctic haze*), skyldes transport av forurensning fra lavere breddegrader i løpet av vinteren/våren, hovedsakelig transport fra Europa og Russland.

Summary

This annual report describes the activities and main results of the programme “*Monitoring of greenhouse gases and aerosols at the Zeppelin Observatory, Svalbard, and Birkenes Observatory, Aust-Agder, Norway*”. The report comprises the measurements of 46 climate gases up to December 31st 2017; including the most important naturally occurring well-mixed greenhouse gases, synthetic greenhouse gases, reactive short lived gases as volatile organic compounds (VOCs), and CO in addition to various particle properties with high relevance to climate. Many of the gases also have strong ozone depleting effects. The development and trends for the period 2001-2017 are reported, in addition to daily and annual mean observations. In 2015, the programme was extended to include 16 new gases, all with measurements analysed back to 2010. In 2016 the programme was further extended with six more species after modification of the instrumentation at Zeppelin: three hydrofluorocarbons (HFCs), sulphuryl fluoride (SO₂F₂), halon H-2402 and nitrogen trifluoride (NF₃). The main results for 2017 are summarized in Table 1.

The measurements at Zeppelin Observatory provide the trend in background level concentrations of climate gases in the Arctic, and all the 46 climate gases in the programme are monitored at this site. Birkenes Observatory is located in an area of Southern Norway highly influenced by long-range transport of pollutants. At Birkenes the influence of local vegetation/terrestrial interactions is also important. Carbon dioxide (CO₂) and methane (CH₄) measurements are undertaken at Birkenes, in addition to a comprehensive aerosol measurement programme. Aerosol measurements are also performed at Zeppelin and Trollhaugen (Antarctica), albeit less extensively than the Birkenes measurements.

The concentration of all main greenhouse gases have been increasing since 2001, except for the ozone-depleting chlorofluorocarbons (CFCs) and a few halogenated gases regulated through the successful Montreal Protocol.

The observations from 2017 show new record high levels for most climate gases measured. The annual average CO₂ concentration in 2017 was 408.0 ppm at Zeppelin and 411.3 ppm at Birkenes. The increases from 2016 are 3.6 ppm (0.89%) and 1.5 ppm (0.37%), respectively.

The concentration of CH₄ also reached new record high levels in 2017 with annual mean concentrations of 1939 ppb at Zeppelin and 1945 ppb at Birkenes. These levels represent increases of 7 ppb (0.35%) and 3 ppb (0.18%) at Zeppelin and Birkenes, respectively. The changes over the last 10 years are large compared to the evolution of the methane levels in the period 1998-2005, when the change was close to zero both at Zeppelin and globally. For the isotopic signature of CH₄, a clear trend in $\delta^{13}\text{C}_{\text{CH}_4}$ is evident after 2012. The observed negative shifts in ambient $\delta^{13}\text{C}_{\text{CH}_4}$, and increases in the CH₄ ambient mixing ratio, imply changes in the balance of sources and sinks. The developments in $\delta^{13}\text{C}_{\text{CH}_4}$ observed at Zeppelin suggest increases in biosphere and/or agriculture (wetland or ruminant) emissions. Another explanation is a change in the sink processes.

N₂O at Zeppelin was also record high in 2017. The annual mean concentration was 330.0 ppb, an increase of 0.9 ppb since 2016.

The synthetic manmade greenhouse gases included in the monitoring programme at Zeppelin are 4 chlorofluorocarbons (CFCs), 3 hydrochlorofluorocarbons (HCFCs), and 11 HFCs - the last

two being CFC substitutes. In addition three halons, a group of 8 halogenated gases, and 4 perfluorinated carbons (PFCs) with very high global warming potentials are included. Furthermore, sulphuryl fluoride and nitrogen trifluoride were reported for the first time in 2016. Both are extremely strong greenhouse gases.

New and previously unreported CFC-11 emission sources were recently discovered in Asia, which have slowed down the rate of CFC-11 decreases globally. Still, the development of the CFC gases measured at Zeppelin is very positive as the concentrations of the dominating species are declining. Contrary to the CFCs, the CFC substitutes HCFCs and HFCs have increased over the period 2001-2017. For the HCFCs, a relaxation in the upward trend has been observed the last years, but for HFCs a strong increase is observed since 2001, and the increasing trend is continuing. The contribution from these manmade gases to global warming is small today, as the concentrations of HFCs are still very low. But given the extremely rapid increase in the use of these gases, it is crucial to follow their development in the future.

Atmospheric concentrations of PFCs and sulphur hexafluoride (SF_6) are also low. An exception is PFC-14, which had a mixing ratio of 84.3 ppt in 2017, an increase of 1.0 ppt since 2016. SF_6 should also be followed closely, as this compound has an atmospheric life time of 3 200 years and an extremely high GWP of 23 500. This compound has increased by 91% since 2001.

Aerosols are small particles in the atmosphere. They can have warming or cooling effects on climate, depending on their properties. Aerosol loads and properties at Birkenes are mainly caused by long-range transport of air pollution from continental Europe, combined with regional sources like biogenic particle formation. Since starting the measurements at Birkenes, no trends in the aerosol physical properties can be detected. This is in agreement with results from other Nordic stations, but the time series are too short for firm conclusions.

For Zeppelin the situation is different. By comparing our aerosol absorption data collected at Zeppelin with data from collaborating institutes, a decreasing trend is observed, indicating a decrease of “black carbon” at Zeppelin. Observations of the total amount of aerosol particles in the atmosphere above Ny-Ålesund (aerosol optical depth) show high concentration levels during springtime compared to the rest of the year. This phenomenon, called Arctic haze, is due to transport of pollution from lower latitudes accumulating in the Arctic atmosphere during winter/spring.

Table 1a: Key findings; Greenhouse gases measured at Zeppelin, Ny-Ålesund; lifetimes in years¹, global warming potential (GWP over 100 years, when available), annual mean concentrations for 2017 and their long term trends per year over the measurement period. Concentrations are in ppm (parts per million) for CO₂, ppb (part per billion) for CH₄, CO, and N₂O, and ppt (parts per trillion) for the other gases. The trend method is described in appendix II.

Component		Life-time /Yr	GWP	Annual mean 2017	Absolute change last year	Long term trend /yr
Carbon dioxide - Zeppelin	CO ₂	.**	1	408.0	3.6	2.6
Carbon dioxide - Birkenes				411.3	1.5	2.5
Methane - Zeppelin	CH ₄	12.4	28	1938.9	6.8	5.9
Methane - Birkenes				1945.3	3.4	8.2
Carbon monoxide	CO	few months	-	114.3	1.4	-1.3
Nitrous oxide	N ₂ O	121	265	330.0	0.94	0.93
Chlorofluorocarbons						
CFC-11	CCl ₃ F	45	4 660	230.2	-1.14	-1.89
CFC-12	CF ₂ Cl ₂	640	10 200	512.5	-3.51	-2.36
CFC-113	CF ₂ ClCFCl ₂	85	13 900	71.1	-0.54	-0.65
CFC-115	CF ₃ CF ₂ Cl	1 020	7 670	8.6	0.06	0.02
Hydrochlorofluorocarbons						
HCFC-22	CHClF ₂	11.9	1 760	252.0	3.49	6.21
HCFC-141b	C ₂ H ₃ FCl ₂	9.2	782	25.9	-0.12	0.59
HCFC-142b	CH ₃ CF ₂ Cl	17.2	1 980	23.4	-0.06	0.63
Hydrofluorocarbons						
HFC-125	CHF ₂ CF ₃	28.2	3 170	25.9	3.00	1.46
HFC-134a	CH ₂ FCF ₃	13.4	1 300	103.2	6.57	5.01
HFC-152a	CH ₃ CHF ₂	1.5	506	10.2	0.02	0.50
HFC-23	CHF ₃	228	12 400	30.7	1.09	1.00
HFC-365mfc	CH ₃ CF ₂ CH ₂ CF ₃	8.7	804	1.24	0.05	0.08
HFC-227ea	CF ₃ CHFCF ₃	38.9	3 350	1.47	0.13	0.11
HFC-236fa	CF ₃ CH ₂ CF ₃	242	8 060	0.17	0.01	0.01
HFC-245fa	CHF ₂ CH ₂ CF ₃	7.7	858	3.04	0.24	0.20
HFC-32	CH ₂ F ₂	5.2	677	18.3	3.14	1.80
HFC-4310mee	C ₅ H ₂ F ₁₀	16.1	1 650	0.28	0.01	0.01
HFC-143a	CH ₃ CF ₃	47.1	4 800	22.5	1.78	1.52
Fluorinated compounds						
PFC-14	CF ₄	50 000	6 630	84.26	0.99	0.39
PFC-116	C ₂ F ₆	10 000	11 100	4.74	0.10	0.09
PFC-218	C ₃ F ₈	2600	8 900	0.66	0.02	0.01
PFC-318	c-C ₄ F ₈	3200	9 540	1.66	0.07	0.05

* The measurements of these components have higher uncertainty. See Appendix I for more details.

¹ From Scientific Assessment of Ozone Depletion: 2010 (WMO, 2011b) and the 4th Assessment Report of the IPCC

** Carbon dioxide does not have a specific lifetime because it is continuously cycled between the atmosphere, oceans and land biosphere and its net removal from the atmosphere involves a range of processes with different time scales.

Component		Life-time /Yr	GWP	Annual mean 2017	Absolute change last year	Long term trend /yr
Sulphurhexafluoride	SF ₆	3 200	23 500	9.46	0.37	0.28
Nitrogen trifluoride	NF ₃	500	16 100	1.76	0.15	-
Sulphuryl fluoride	SO ₂ F ₂	36	4,090	2.46	0.13	0.11
Halons						
H-1211	CBrClF ₂	16	1 750	3.55	-0.11	-0.06
H-1301	CBrF ₃	65	7 800	3.39	0.001	0.02
H-2402	CBrF ₂ CBrF ₂	20	1 470	0.41	-0.01	-0.01
Halogenated compounds						
Chloromethane	CH ₃ Cl	1	12	517.33	-5.45	-0.17
Bromomethane	CH ₃ Br	0.8	2	6.54	-0.15	-0.18
Dichloromethane	CH ₂ Cl ₂	0.4	9	61.40	4.65	1.95
Trichloromethane	CHCl ₃	0.4	16	15.39	0.97	0.27
Carbon tetrachloride	CCl ₄	26	1730	79.53	-1.13	-0.98
Trichloroethane	CH ₃ CCl ₃	5	160	2.33	-0.48	-2.05
Trichloroethene	CHClCCl ₂	-	-	0.32	-0.09	-0.01
Tetrachloroethene	CCl ₂ CCl ₂	-	-	2.48	-0.08	-0.12
Volatile Organic Compounds (VOC)						
Ethane	C ₂ H ₆	Ca 78 days*		1573.78	-9.58	-
Propane	C ₃ H ₈	Ca 18 days*		583.95	14.18	-
Butane	C ₄ H ₁₀	Ca 8 days*		193.00	23.20	-
Pentane	C ₅ H ₁₂	Ca 5 days*		60.77	1.84	-
Benzene	C ₆ H ₆	Ca 17 days*		59.05	-7.65	-
Toluene	C ₆ H ₅ CH ₃	Ca 2 days*		17.85	-7.64	-

1 Introduction to monitoring of greenhouse gases and aerosols

1.1 The monitoring programme in 2017

The purpose of the monitoring programme is to study the long-term development of climate gases and aerosols (particles). Measurements are performed at three sites and the results are used as input to European and global observation networks.

The atmospheric monitoring programme presented in this report focuses on the concentrations of atmospheric greenhouse gases and on selected aerosol physical and optical properties relevant for the understanding of climate change. Sampling sites are at Svalbard in the Norwegian Arctic (Zeppelin Observatory), where observations are considered to be representative for well-mixed background concentration levels. A second site is in southern Norway (Birkenes Observatory), where observations are more influenced by regional and local sources. A third site is in the Antarctic (Troll station).

The main objectives are to quantify the levels of greenhouse gases including ozone depleting substances, describe the relevant optical and physical properties of aerosols, and document their development over time. Measurements of the greenhouse gas concentrations and aerosol properties are core data for studies and assessments of climate change, and also crucial in order to evaluate mitigation strategies and their effectiveness. The Norwegian monitoring sites are located in areas where the influence of local sources are minimal, hence the sites are representative for a wider region allowing detection of long-term changes in atmospheric composition.



Figure 1: Location of NILU's atmospheric supersites measuring greenhouse gases and aerosol properties.

1.2 The measurements in relation to research and policy agreements

The Norwegian greenhouse gas and aerosol monitoring programme is set up to meet national and international needs for greenhouse gas and aerosol measurement data, both for the scientific community, national environmental authorities and global policy making.

Greenhouse gases: The targets set by the Kyoto protocol for its first and second commitment periods are to reduce the total emissions of greenhouse gases by the industrialized countries. Later, the Paris Agreement was negotiated and adopted by consensus at the 21st Conference of the Parties of the UNFCCC in Paris on 12 December 2015. The Paris Agreement entered into force on 4 November 2016. Currently 182 of the 197 Parties to the Convention, have ratified the Protocol². The central aim of the Paris agreement is to keep the increase in the global average temperature to well below 2 °C compared to pre-industrial levels and to pursue efforts to limit the temperature increase to 1.5 °C. The EU Heads of State and Governments agreed in October 2014 on the headline targets and the architecture for the EU framework on climate and energy for 2030. The agreed targets include a cut in greenhouse gas emissions by at least 40% by 2030 compared to 1990 levels³.

Ozone depleting substances and their replacement gases: In 1987 the Montreal Protocol was signed and entered into force in 1989 in order to reduce the production, use and eventually emission of the ozone-depleting substances (ODS). The amount of most ODS in the troposphere is now declining slowly and is expected to be back to pre-1980 levels around year 2050. It is central to follow the development of the concentration of these ozone depleting substances in order to verify that the Montreal Protocol and its amendments work as expected. The development of the ozone layer above Norway is monitored closely, and the results of the national monitoring of ozone and UV is presented in “*Monitoring of the atmospheric ozone layer and natural ultraviolet radiation: Annual report 2017*” (Svendby et al. 2018). The ozone depleting gases and their replacement gases are also strong greenhouse gases making it even more important to follow the development of their concentrations.

To control the new replacement gases, a historical agreement was signed on 15 October 2016 when negotiators from 197 countries agreed to reduce the production and consumption of hydrofluorocarbons (HFCs). The agreement was finalized at the United Nations meeting in Kigali, Rwanda, aiming to reduce the use of HFCs by more than 80% over the course of the twenty-first century. The agreement in Kigali represents an amendment of the 1987 Montreal Protocol. The HFCs can be up to 10000 times as effective at trapping heat compared to carbon dioxide. Today HFCs account for a small fraction of the greenhouse-gas emissions and have had limited influence on the global warming up to now. However, the use of HFCs is growing rapidly and the projected HFC emission could contribute up to 0.5 °C of global warming by the end of this century if not regulated (Xu et al., 2013). Because the agreement in Kigali is an amendment to the Montreal Protocol, which was ratified back in the 1990s, this new agreement is legally binding for the countries that have ratified when the amendment enters into force in January 2019.

² <https://unfccc.int/process/the-paris-agreement/status-of-ratification>

³ Details here: <http://ec.europa.eu/clima/policies/strategies/2030/> and here http://www.consilium.europa.eu/uedocs/cms_data/docs/pressdata/en/ec/145397.pdf

⁴ *Norwegian Environment Agency monitoring reports

1.3 The ongoing monitoring programme and the link to networks and research infrastructures

In response to the need for monitoring of greenhouse gases and ozone depleting substances, the *Norwegian Environment Agency* and *NILU - Norwegian Institute for Air Research* signed a contract commissioning NILU to run a programme for monitoring greenhouse gases at the Zeppelin Observatory, close to Ny-Ålesund in Svalbard in 1999. This national programme now includes monitoring of 46 greenhouse gases and trace gases at the Zeppelin Observatory in the Arctic, many of them also ozone depleting substances. In 2009, NILU upgraded and extended the observational activity at the Birkenes Observatory in Aust-Agder. From 2010, the Norwegian Environment Agency/NILU monitoring programme was extended to also include the new observations from Birkenes of the greenhouse gases CO₂ and CH₄ and selected aerosol observations particularly relevant for the understanding of climate change. Relevant components are also reported in “*Monitoring of long-range transported air pollutants in Norway, annual report 2017*”⁵(Aas et al. 2018), this includes particulate and gaseous inorganic constituents, particulate carbonaceous matter, ground level ozone and particulate matter for 2017. This report also includes a description of the weather in Norway in 2017 in Chap. 2, which is relevant for the observed concentrations of greenhouse gases and aerosols.

The location of both sites are shown in Figure 1, and pictures of the sites are shown in Figure 2. The unique location of the Zeppelin Observatory at Svalbard, together with the infrastructure of the scientific research community in Ny-Ålesund, makes it ideal for monitoring the global changes of concentrations of greenhouse gases and aerosols in the atmosphere. There are few local sources of emissions, and the Arctic location is also important as the Arctic is a particularly vulnerable region. The observations at the Birkenes Observatory complement the Arctic site. Birkenes Observatory is located in a forest area with few local sources. However, the Observatory often receives long-range transported pollution from Europe and the site is ideal to analyse the contribution of long range transported greenhouse gases and aerosol properties.

⁵ *Norwegian Environment Agency monitoring reports



Figure 2: The two atmospheric supersites included in this programme, Zeppelin above and Birkenes to the left

Data and results from the national monitoring programme are also included in various international programmes. Both sites contribute to EMEP (European Monitoring and Evaluation Programme) under the CLTRAP (Convention on Long-range Transboundary Air Pollution). Data from the sites are also reported to CAMP (Comprehensive Atmospheric Monitoring Programme) under OSPAR (the Convention for the Protection of the marine Environment of the North-East Atlantic, <http://www.ospar.org>), AMAP (Arctic Monitoring and Assessment Programme <http://www.amap.no>), WMO/GAW (The World Meteorological Organization, Global Atmosphere Watch programme, <http://www.wmo.int>) and AGAGE (Advanced Global Atmospheric Gases Experiment).

Zeppelin and Birkenes are both included in two central European environmental research infrastructures (RI) focusing on climate forcers and air quality. This ensure high quality data with harmonised methods and measurements across Europe and also a global link via GAW, to have comparable data and results. This is essential to reduce the uncertainty in trends and in the observed levels of the wide range of climate forcers. International collaboration and harmonisation of these types of observations are crucial for improved processes understanding and satisfactory quality to assess trends. The two central RIs are ICOS (Integrated Carbon Observation System, <https://www.icos-ri.eu>) focusing on the understanding of carbon cycle, and ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure, www.actris.eu) focusing on short-lived aerosol climate forcers and related reactive gases, and clouds. The networks EMEP and AGAGE, and the research infrastructures ACTRIS and ICOS are crucial for quality assurance and quality control of the Norwegian measurement data and instruments. All measurements included in this report follow the protocols, methodology and recommendations of these frameworks. This is a prerequisite for

harmonised and comparable data on both European and global scale, see Table 2 at page 16. Implementation of Norwegian measurements in ICOS through the ICOS-Norway⁶ project is described in the appendix II.

NILU hosts the data centres of the European Monitoring and Evaluation Programme (EMEP), ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure) and the WMO Global Atmosphere Watch (GAW) World Data Centre for Aerosol (WDCA) and GAW- World Data Centre for Reactive Gases (WDCRG) (from 2015), and numerous other projects and programs (e.g. AMAP, HELCOM) and all data reported are accessible in the EBAS data base:

<http://ebas.nilu.no>. All data from the national monitoring programme and from these frameworks are reported to this database. It is important to highlight that NILUs work in ACTRIS and also hosting the WMO GAW World Data Centres for Aerosol, among many other synergy effects, ensures efficient dissemination of the data on atmospheric aerosol properties collected within the Norwegian climate monitoring programme, to the scientific community. Among others, ACTRIS will develop a primary standard for calibrating instruments measuring aerosol absorption, one of the properties of atmospheric black carbon, and develop quality standards for measuring the aerosol particle size distribution in order to further improve assessments of aerosol climate forcing. Another project relevant in this context is *Environmental Research Infrastructures Providing Shared Solutions for Science and Society* (ENVRI^{plus})⁷ project. ENVRI^{plus} is an umbrella project for all environmental research infrastructures funded or supported by the EU. One of its objectives will be to put data from the atmospheric, marine, tectonic, and biosphere domains into a common context by making the data interoperable, i.e. visible in common services. The efforts started with achieving this goal first within the atmospheric domain.

Compiled key information on the national monitoring programme is listed in Table 2. From 2015 the programme was extended with 16 new greenhouse gases and reactive trace gases, mainly HFCs and non-methane hydrocarbons. From 2016 also NF₃ and 5 PFCs and SO₂F₂ were added. More detailed information on the monitoring programme and measurement frequencies are provided in Appendix II. For the measurements of aerosol properties more details are presented in chapter 3.

⁶ <https://no.icos-cp.eu>

⁷ <http://www.envriplus.eu>

Table 2: Summary of the measurement programme, run by NILU, at Birkenes and Zeppelin Observatory. The green colour indicate the components added to the program on 2016.

Component	Birkenes Start	Zeppelin Start	International network, QA programme in bold	Comment
Trace gases				
CO ₂	2009	2012	ICOS	Measured at Zeppelin since 1988 by Univ. Stockholm. By NILU at Zeppelin since 2012, now included in the programme. Qualified as ICOS class 1 site, and passed first step in September, 2016. ICOS labelling scheduled in 2018 for Birkenes
CH ₄	2009	2001	ICOS, EMEP	ICOS labelling and implementation in 2017 for Zeppelin, 2018 for Birkenes
N ₂ O	-	2009	ICOS	ICOS labelling and implementation in 2017
CO	-	2001	ICOS	ICOS labelling and implementation in 2017
Ozone (surface)	1985	1989	EMEP	Reported in Aas et al. 2018; M-1064/2018.
CFCs		2001/ 2010 and later	AGAGE	<p>*The measurements marked “***”: these components are not within the required precision of AGAGE, but a part of the AGAGE quality assurance programme.</p> <p>The compounds marked in green were included in the national monitoring programme from 2015, with harmonised time series and measurements back to 2010 when the Medusa instrument was installed at Zeppelin.</p>
CFC-11*				
CFC-12*				
CFC-113*				
CFC-115*				
HCFCs				
HCFC-22				
HCFC-141b				
HCFC-142b				
HFC-125				
HFC-134a				
HFC-152a				
HFCs				
HFC-125				
HFC-134a				
HFC-152a				
HFC-23				
HFC-227ea				
HFC-236fa				
HFC-245fa				
HFC-365mfc				
HFC-32				
HFC-4310mee				
HFC-143a				
PFCs				
PFC-14				
PFC-116				
PFC-218				
PFC-318				
Halons				
H-1211				
H-1301				
H-2402				
Other chlorinated				
CH ₃ Cl				
CH ₃ Br				
CH ₂ Cl ₂				
CHCl ₃				
CCl ₄				
CH ₃ CCl ₃				
CHClCCl ₂				
CCl ₂ CCl ₂				
Other fluorinated				
SF ₆				
NF ₃		2016		
SO ₂ F ₂		2010		
VOCs		2010	ACTRIS, EMEP	

C ₂ H ₆ - ethane				VOCs were included in the national monitoring programme from 2015, but the measurements are harmonised back to 2010.
C ₃ H ₈ - propane				
C ₄ H ₁₀ - butane				
C ₅ H ₁₂ - pentane				
C ₆ H ₆ - benzene				
C ₆ H ₅ CH ₃ - toluene				
Aerosol measurements				
Absorption properties	2009	2015	ACTRIS, EMEP	Measured by Univ. of Stockholm at Zeppelin, New from late 2015
Scattering properties	2009	-	ACTRIS, EMEP	Measured by Univ. of Stockholm at Zeppelin
Number Size Distribution	2009	2010	ACTRIS, EMEP	Reported in Aas et al. 2018; M-1064/2018.
Cloud Condensation Nuclei	2012	-	ACTRIS	Zeppelin: In collaboration with Korean Polar Research Institute
Aerosol Optical depth	2010	2007	AERONET, GAW-PFR	Birkenes: AERONET, Ny-Ålesund: GAW-PFR
PM ₁₀	2001		EMEP	Reported in Aas et al. 2018; M-1064/2018.
PM _{2.5}	2001		EMEP	
Chemical composition -inorganic	1978	1979	EMEP	
Chemical composition - carbonaceous matter	2001		EMEP	

1.4 Greenhouse gases, aerosols and their climate effects

The Intergovernmental Panel on Climate Change's (IPCC's) Fifth Assessment Report (IPCC AR5) and the contribution from Working Group I "*Climate Change 2013: The Physical Science Basis*" was published in September 2013. This substantial climate assessment report presents new evidence of past and projected future climate change from numerous independent scientific studies ranging from observations of the climate system, paleoclimate archives, theoretical studies on climate processes and simulations using climate models. Their main conclusion was:

"Warming of the climate system is unequivocal, and since the 1950s, many of the observed changes are unprecedented over decades to millennia. The atmosphere and ocean have warmed, the amounts of snow and ice have diminished, sea level has risen, and the concentrations of greenhouse gases have increased"

(IPCC, Summary for policy makers, WG I, 2013)

Their conclusions are based on a variety of independent indicators, some of them are observations of atmospheric compositional change. The overall conclusion with respect to the development of the concentrations of the main greenhouse gases is:

"The atmospheric concentrations of carbon dioxide, methane, and nitrous oxide have increased to levels unprecedented in at least the last 800,000 years. Carbon dioxide concentrations have increased by 40% since pre-industrial times, primarily from fossil fuel emissions and secondarily from net land use change emissions. The ocean has absorbed about 30% of the emitted anthropogenic carbon dioxide, causing ocean acidification"

(IPCC, Summary for policy makers, 2013)

In particular chapter 2, "*Observations: Atmosphere and Surface*", presents all types of atmospheric and surface observations, including observations of greenhouse gases since the start of the observations in mid-1950s and changes in aerosols since the 1980s. The IPCC AR5 report was the first time long term changes of aerosols were included in the report, based on global and regional measurement networks and satellite observations. The main conclusion with respect to development of the aerosol levels is that "*It is very likely that aerosol column amounts have declined over Europe and the eastern USA since the mid-1990s and increased over eastern and southern Asia since 2000*" (Hartmann et al, 2013). This is important since the net effect of aerosols is atmospheric cooling, counteracting the effect of greenhouse gases. The changes in Europe and the USA are mainly due to mitigation strategies of e.g. sulfur, while the emissions are increasing rapidly in Asia, including increasing emissions of the warming component black carbon.

The basic metric to compare the effect of the various climate change drivers is radiative forcing (RF). RF is the net change in the energy balance of the Earth system due to some imposed change. RF provides a quantitative basis for comparing potential climate response to different changes. Forcing is often presented as the radiative change from one time-period to another, such as pre-industrial to present-day. For many forcing agents the RF is an appropriate way to compare the relative importance of their potential climate effect. However, rapid adjustments in the troposphere can either enhance or reduce the perturbations, leading to large differences in the forcing driving the long-term climate change. In the last IPCC report it was also introduced a new concept, the effective radiative forcing (ERF). The ERF concept aims to take rapid adjustments into account, and is the change in net TOA (Top Of Atmosphere) downward radiative flux after allowing for atmospheric temperatures, water vapour and clouds to adjust, but with surface temperature or a portion of surface conditions unchanged (Myhre et al, 2013b). Figure 3 shows the RF and ERF of the main components referring to a change in the atmospheric level since 1750, pre-industrial time.

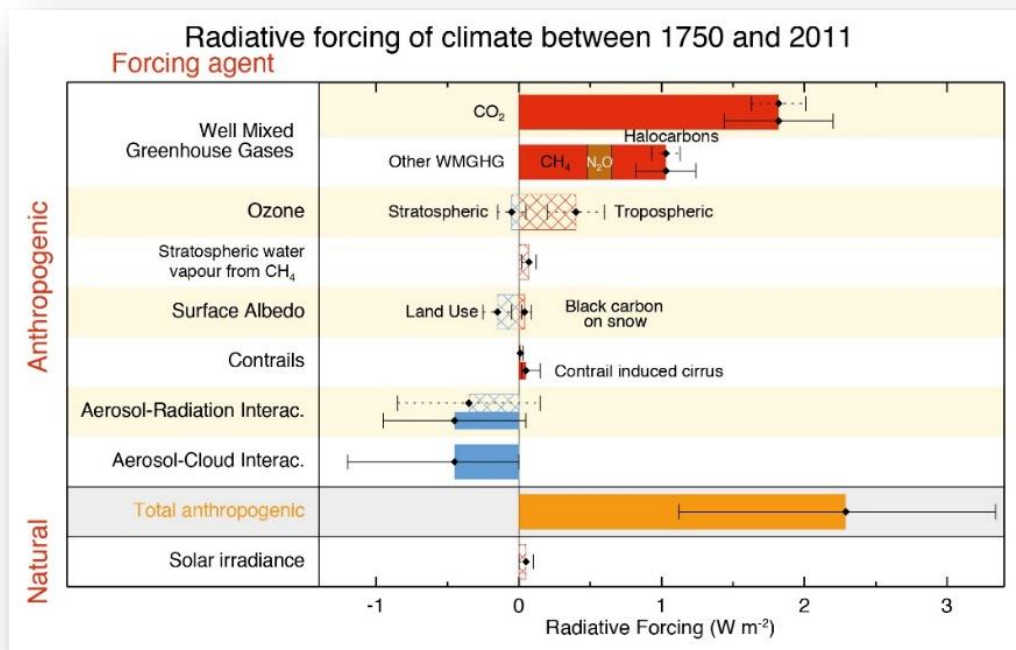


Figure 3: Bar chart for RF (hatched) and ERF (solid) for the period 1750-2011. Uncertainties (5 to 95% confidence range) are given for RF (dotted lines) and ERF (solid lines). (Taken from Myhre et al, 2013b).

Total adjusted anthropogenic forcing is 2.29 W m^{-2} , [1.13 to 3.33], and the main anthropogenic component driving this is CO_2 with a total RF of 1.82 W m^{-2} . The direct and indirect effect of aerosols are cooling and calculated to -0.9 W m^{-2} . The diagram in Figure 4 shows a comparison in percent % of the various contribution from the long-lived greenhouse gases to the total forcing of the well-mixed greenhouse gases, based on 2011 levels.

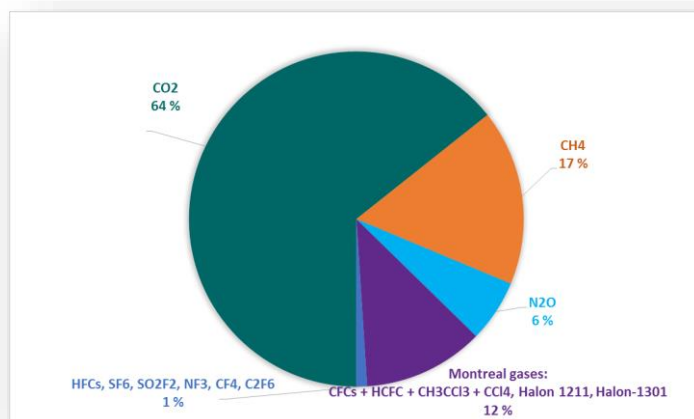


Figure 4: The contribution in % of the well-mixed greenhouse gases to the total forcing of the well-mixed greenhouse gases for the period 1750-2011 based on estimates in Table 8.2 in Chap 8, of IPCC (Myhre et al, 2013b).

An interesting and more detailed picture of the influence of various emissions on the RF is illustrated in Figure 5. This Figure shows the forcing since 1750 by emitted compounds, to better illustrate the effects of emissions and potential impact of mitigations.

As seen, the number of emitted compounds and changes leading to RF is larger than the number of compounds causing RF directly. This is due to indirect effects, in particular components involved in atmospheric chemistry that affect e.g. CH₄ and ozone. Emissions of CH₄, CO, and NMVOC all lead to excess CO₂ as one of the end products, and this is the reason why the RF of direct CO₂ emission is slightly lower than the RF of abundance change of CO₂ in Figure 3. Note also that for CH₄, the contribution from emissions is estimated to be almost twice as large as that from the direct effect of the CH₄ concentration change, 0.97 W m⁻² versus 0.48 W m⁻² shown in Figure 3 and Figure 5 respectively. This is because emission of CH₄ leads to ozone production (shown in green colour in the CH₄ bar in Figure 5), stratospheric water vapour, CO₂ (as mentioned above), and importantly affects its own lifetime. As seen from the Figure, there is also a particularly complex picture of the effects of aerosols. Black carbon heats the atmosphere, originating from both fossil fuel, biofuel and biomass burning. The direct effect of black carbon from fossil and biofuel is +0.4 W m⁻², while black carbon from biomass burning is 0 in total due to co-emitted effects of organic carbon, cooling the atmosphere and cancelling out the heating effect.

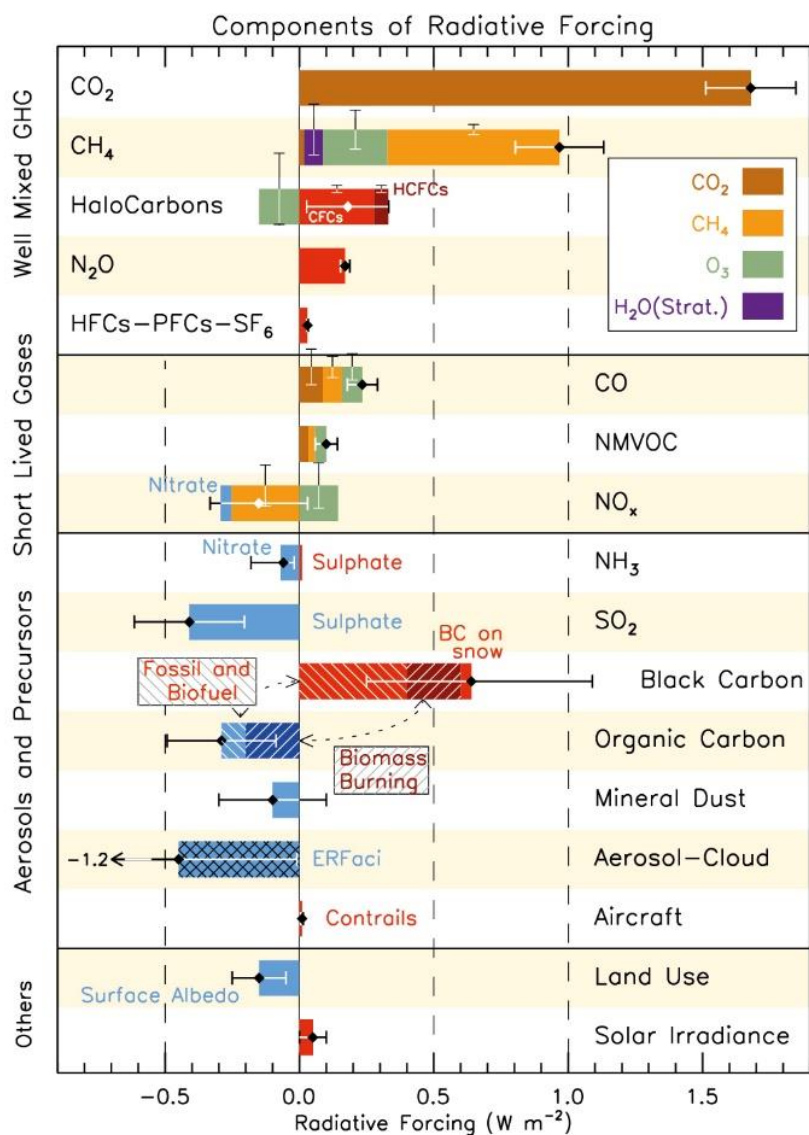


Figure 5: RF bar chart for the period 1750-2011 based on emitted compounds (gases, aerosols or aerosol precursors) or other changes. Red (positive RF) and blue (negative forcing) are used for emitted components which affect few forcing agents, whereas for emitted components affecting many compounds several colours are used as indicated in the inset at the upper part the figure. The vertical bars indicate the relative uncertainty of the RF induced by each component. Their length is proportional to the thickness of the bar, that is, the full length is equal to the bar thickness for a $\pm 50\%$ uncertainty. The net impact of the individual contributions is shown by a diamond symbol and its uncertainty (5 to 95% confidence range) is given by the horizontal error bar. ERFaci is ERF due to aerosol-cloud interaction. BC and OC are co-emitted, especially for biomass burning emissions (given as Biomass Burning in the figure) and to a large extent also for fossil and biofuel emissions (given as Fossil and Biofuel in the figure where biofuel refers to solid biomass fuels) (The Figure is taken from Myhre et al, 2013b).

In addition there is a small heating effect of black carbon on snow (0.04 W m^{-2} since 1750). The total effect of black carbon on snow since 1750 is currently in the order of a one year increase of CO_2 concentration in the atmosphere (around 2 ppm).

1.4.1 Revisions of forcing estimates of carbon dioxide, methane, and nitrous oxide

In 2016 there was an important study publishing revised radiative forcing of carbon dioxide, methane, and nitrous oxide. This resulted in a significant revision of the methane radiative forcing, and minor changes in the others. To reduce uncertainty of the forcing, more accurate forcing estimates were included and the global warming potential, using consistent methodology with IPCC and earlier estimates, was revised. For CH₄ this resulted in **25% stronger forcing than given in the IPCC Fifth Assessment Report**, due to inclusion of the shortwave and near-infrared bands of CH₄, improved knowledge about water vapour absorption, more detailed models (Etminan *et al.*, 2016). Figure 6 is taken from Etminan *et al.* (2016) and show the revised estimates, compared to the old as included in IPCC AR5. The estimates are based on concentration development as in representative concentration pathway (RCP⁸) RCP8.5 from IPCC AR5.

To put this in perspective, a paper in Nature Geoscience is using data from Zeppelin and this monitoring programme, together with other data to calculate radiative forcing due to CO₂ and all greenhouse gases over time, (Myhre *et al.*, 2017) with comparison to future RCP pathways. The paper shows that we are now halfway to a doubling of CO₂ in terms for radiative forcing but not in concentrations, since the preindustrial time (1750). According to the observed concentrations, we are now following path between RCP8.5 and other RCP scenarios.

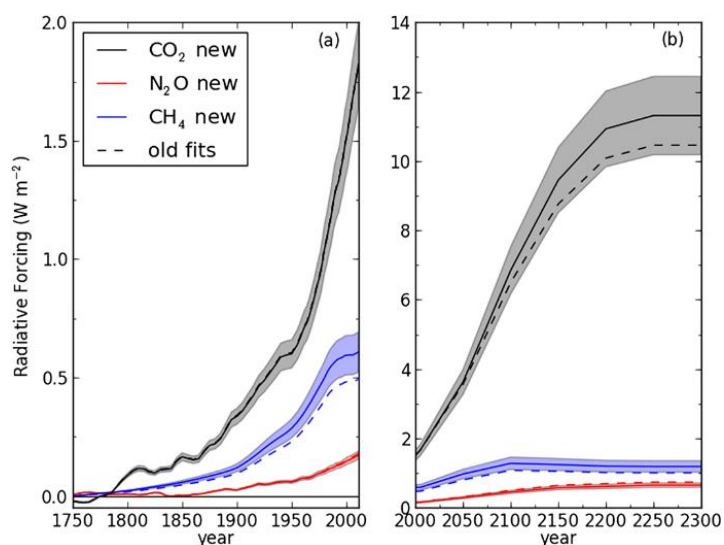


Figure 6: Radiative forcing of CO₂, N₂O, and CH₄ concentration change: (a) from 1755 to 2011 and (b) from 2000 to 2300, using RCP8.5 concentrations from IPCC AR5 relative to preindustrial value (280 ppm of CO₂, 275 ppb of N₂O, and 750 ppb of CH₄) using old and new expressions. Shading for the new expressions indicates the estimated radiative uncertainty in the forcing (Etminan *et al.*, 2016).

⁸ https://en.wikipedia.org/wiki/Representative_Concentration_Pathways

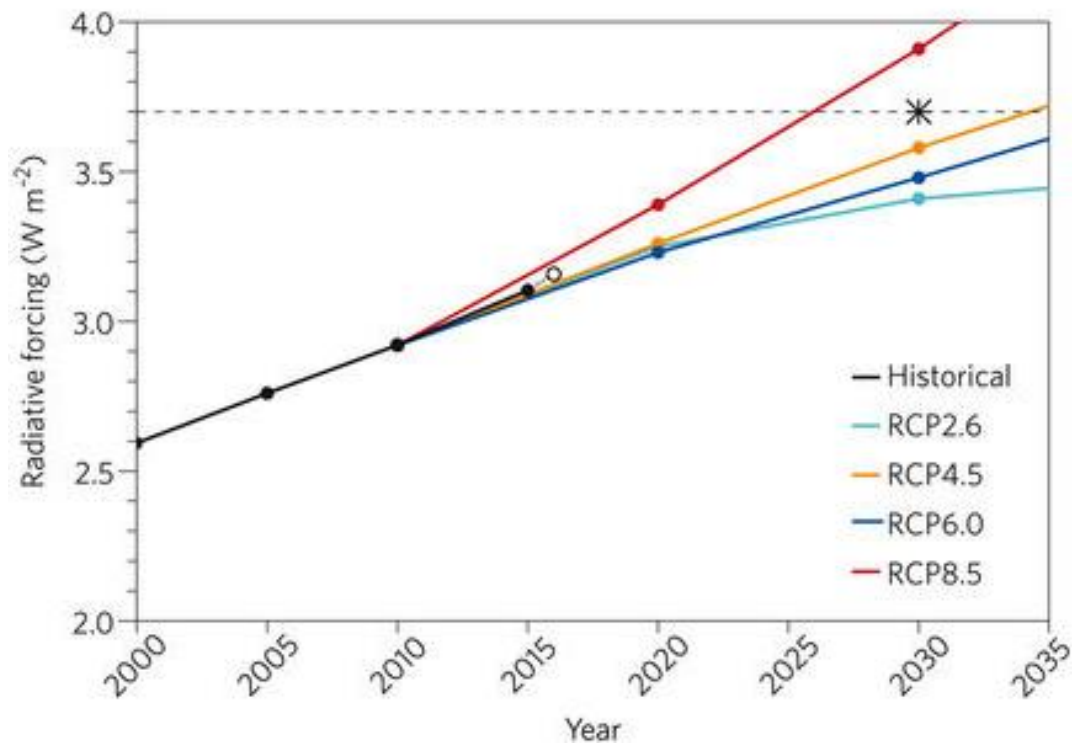


Figure 7: Radiative forcing for all WMGHGs calculated. Historical values are based on observed concentrations. CH₄ concentrations are also from the IPCC. For 2015 the global annual mean concentrations of CO₂, CH₄ and N₂O are from the National Oceanic and Atmospheric Administration, and for halocarbons the relative increase since 2010 are from the Arctic Zeppelin observatory. Preliminary data for 2016 are included, which may be subject to small changes, taken from (Myhre et al., 2017) Figure 8

2 Observations of climate gases

NILU measures 46 climate gases at the Zeppelin Observatory at Svalbard and 2 at Birkenes, in addition to surface ozone reported in Aas et al., 2018. The results and analyses of the climate gas measurements are presented in this chapter. Furthermore, observations of CO₂ for the time period 1988-2012 at Zeppelin performed by the Stockholm University - Department of Environmental Science and Analytical Chemistry (ACES), are included in the report.

Table 3 summarises the main results for 2017 and the trends over the period 2001-2017. The table also includes 2017 global annual mean values given in BAMS (Dlugokencky et al., 2018).

Table 3: Greenhouse gases measured at Zeppelin and Birkenes; lifetimes in years, global warming potential (GWP) for 100 year horizon and annual mean for 2017, change last year, and trends per year over the measurement period. Red is increasing and blue is decreasing trends. Global means for 2017 taken from BAMS (Dlugokencky et al., 2018) are included for comparison. All concentrations are mixing ratios in ppt, except for methane, nitrous oxide and carbon monoxide (ppb) and carbon dioxide (ppm).

Component		Life-time	GWP	Global mean BAMS 2017	Annual mean 2017	Absolute change last year	Trend /yr	
Carbon dioxide - Zeppelin	CO ₂	-	1	-	408.0	3.6	2.6	
Carbon dioxide - Birkenes					411.3	1.5	2.5	
Methane - Zeppelin	CH ₄	12.4	28	-	1938.9	6.8	5.9	
Methane - Birkenes					1945.3	3.4	8.2	
Carbon monoxide	CO	few months	-	-	114.3	1.4	-1.3	
Nitrous oxide	N ₂ O	121	265	-	330.0	0.94	0.93	
Chlorofluorocarbons								
CFC-11	CCl ₃ F	45	4,660	228.9	230.2	-1.14	-1.89	
CFC-12	CF ₂ Cl ₂	640	10,200	509.3	512.5	-3.51	-2.36	
CFC-113	CF ₂ ClCFCl ₂	85	13,900	70.9	71.1	-0.54	-0.65	
CFC-115	CF ₃ CF ₂ Cl	1,020	7,670	-	8.6	0.06	0.02	
Hydrochlorofluorocarbons								
HCFC-22	CHClF ₂	11.9	1,760	240.8	252.0	3.49	6.21	
HCFC-141b	C ₂ H ₃ FCl ₂	9.2	782	24.5	25.9	-0.12	0.59	
HCFC-142b	CH ₃ CF ₂ Cl	17.2	1,980	22.1	23.4	-0.06	0.63	
Hydrofluorocarbons								
HFC-125	CHF ₂ CF ₃	28.2	3,170	22.8	25.9	3.00	1.46	
HFC-134a	CH ₂ FCF ₃	13.4	1,300	95.7	103.2	6.57	5.01	
HFC-152a	CH ₃ CHF ₂	1.5	506	6.8	10.2	0.02	0.50	
Hydrofluorocarbons cont.								
HFC-23	CHF ₃	228	12,400	29.9	30.7	1.09	1.00	
HFC-365mfc	CH ₃ CF ₂ CH ₂ CF ₃	8.7	804	0.93	1.24	0.05	0.08	
HFC-227ea	CF ₃ CHFCF ₃	38.9	3,350	1.29	1.47	0.13	0.11	
HFC-236fa	CF ₃ CH ₂ CF ₃	242	8,060	-	0.17	0.01	0.01	

HFC-245fa	CHF ₂ CH ₂ CF ₃	7.7	858	-	3.04	0.24	0.20
HFC-32	CH ₂ F ₂	5.2	677	13	18.3	3.14	1.80
HFC-4310mee	C ₅ H ₂ F ₁₀	16.1	1,650	-	0.28	0.01	0.01
HFC-143a	CH ₃ CF ₃	47.1	4,800	20.6	22.5	1.78	1.52
Perfluorinated compounds							
PFC-14	CF ₄	50,000	6,630	83.6	84.26	0.99	0.39
PFC-116	C ₂ F ₆	10,000	11,100	4.66	4.74	0.10	0.09
PFC-218	C ₃ F ₈	2600	8,900	-	0.66	0.02	0.01
PFC-318	c-C ₄ F ₈	3200	9,540	-	1.66	0.07	0.05
Sulphurhexafluoride	SF ₆	3,200	23,500	9.26	9.46	0.37	0.28
Nitrogen trifluoride	NF ₃	500	16,100	-	1.76	0.15	-
Sulphuryl fluoride	SO ₂ F ₂	36	4,090	-	2.46	0.13	0.11
Halons							
H-1211	CBrClF ₂	16	1,750	3.43	3.55	-0.11	-0.06
H-1301	CBrF ₃	65	7,800	3.26	3.39	0.001	0.02
H-2402	CBrF ₂ CBrF ₂	20	1,470	0.4	0.41	-0.01	-0.01
Halogenated compounds							
Chloromethane	CH ₃ Cl	1	12	547.3	517.33	-5.45	-0.17
Bromomethane	CH ₃ Br	0.8	2	6.60	6.54	-0.15	-0.18
Dichloromethane	CH ₂ Cl ₂	0.4	9	-	61.40	4.65	1.95
Trichloromethane	CHCl ₃	0.4	16	-	15.39	0.97	0.27
Carbon tetrachloride	CCl ₄	26	1730	80.2	79.53	-1.13	-0.98
Trichloroethane	CH ₃ CCl ₃	5	160	2.2	2.33	-0.48	-2.05
Trichloroethene	CHClCCl ₂	-	-	-	0.32	-0.09	-0.01
Tetrachloroethene	CCl ₂ CCl ₂	-	-	-	2.48	-0.08	-0.12
Volatile Organic Compounds (VOC)							
Ethane	C ₂ H ₆	Ca 78 days [#]	-	-	1573.78	-9.58	-
Propane	C ₃ H ₈	Ca 18 days [#]	-	-	583.95	14.18	-
Butane	C ₄ H ₁₀	Ca 8 days [#]	-	-	193.00	23.20	-
Pentane	C ₅ H ₁₂	Ca 5 days [#]	-	-	60.77	1.84	-
Benzene	C ₆ H ₆	Ca 17 days [#]	-	-	59.05	-7.65	-
Toluene	C ₆ H ₅ CH ₃	Ca 2 days [#]	-	-	17.85	-7.64	-

[#]The lifetimes of VOC are strongly dependant on season, sunlight, other components etc. The estimates are global averages given in C. Nicholas Hewitt (ed.): *Reactive Hydrocarbons in the Atmosphere*, Academic Press, 1999, p. 313. The times series for these are short and the trend is very uncertain.

Greenhouse gases and other climate gases have numerous sources, both anthropogenic and natural. Trends and future changes in concentrations are determined by their sources and sinks, and in section 2.1 observations and trends of the monitored greenhouse gases with both natural and anthropogenic sources are presented in more detail. In section 2.2 the detailed results of the ozone depleting substances with purely anthropogenic sources are presented.

For annual trend calculations we have used the method described in Appendix II. In order to estimate uncertainties we have applied an updated method taking autocorrelated errors in

the fitted model residuals into account. Appendix II also includes a description of the measurements at the Zeppelin and Birkenes Observatories in more detail. Generally, Zeppelin Observatory is a unique site for observations of changes in the background level of atmospheric components. All peak concentrations of the measured gases are significantly lower here than at other sites at the Northern hemisphere, due to the station's remote location. Birkenes is closer to the main source areas. Further, the regional vegetation is important for regulating the carbon cycle, resulting in much larger variability in the concentration level compared to the Arctic region.

2.1 Climate gases with natural and anthropogenic sources

The annual mean concentrations for all gases included in the programme for all years are given in Appendix I, Table A 1 at page 111. All the trends, uncertainties and regression coefficients are found in Table A 2 at page 114. Section 2.1 focuses on the measured greenhouse gases that have both natural and anthropogenic sources.

2.1.1 Carbon dioxide (CO₂)

Carbon dioxide (CO₂) is the most important anthropogenic greenhouse gas with a radiative forcing of 1.82 W m⁻² since the year 1750, and an increase since the previous IPCC report (AR4, 2007) of 0.16 Wm⁻² (Myhre et al., 2013b). Etminan et al. (2016) presented revised forcing estimates for all main greenhouse gases (see section 1.4.1 at page 22), and confirmed the forcing estimates for CO₂, with only minor changes.

The increase in forcing is due to the increase in concentrations over the last years. CO₂ is the end product in the atmosphere of the oxidation of all main organic compounds, and it has shown an increase of as much as 40 % since the pre industrial times (Hartmann et al, 2013). This is mainly due to emissions from combustion of fossil fuels and land use change. CO₂ emissions from fossil fuel burning and cement production increased by 2.3% in 2013 since 2012, with a total of 9.9±0.5 GtC (billion tonnes of carbon) equal to 36.2 GtCO₂ emitted to the atmosphere, 62% above 1990 emissions (the Kyoto Protocol reference year). Emissions are projected to increase considerably (1.7%) in 2017 according to Global Carbon Project estimates <http://www.globalcarbonproject.org>, though it is still too early to assess and evaluate this.

NILU started CO₂ measurements at the Zeppelin Observatory in 2012 and the results are presented in Figure 9, together with the time series provided by ITM, University of Stockholm, back to 1988. ITM provides all data up till 2012 and we acknowledge the effort they have been doing in monitoring CO₂ at the site. After upgrading Birkenes in 2009, there are continuous measurements of CO₂ and CH₄ from mid May 2009 also at this site.

The atmospheric daily mean CO₂ concentration measured at the Zeppelin Observatory for the period mid 1988-2017 is presented in Figure 9 (upper panel), together with the 2009-2017 time series for Birkenes in the lower panel.

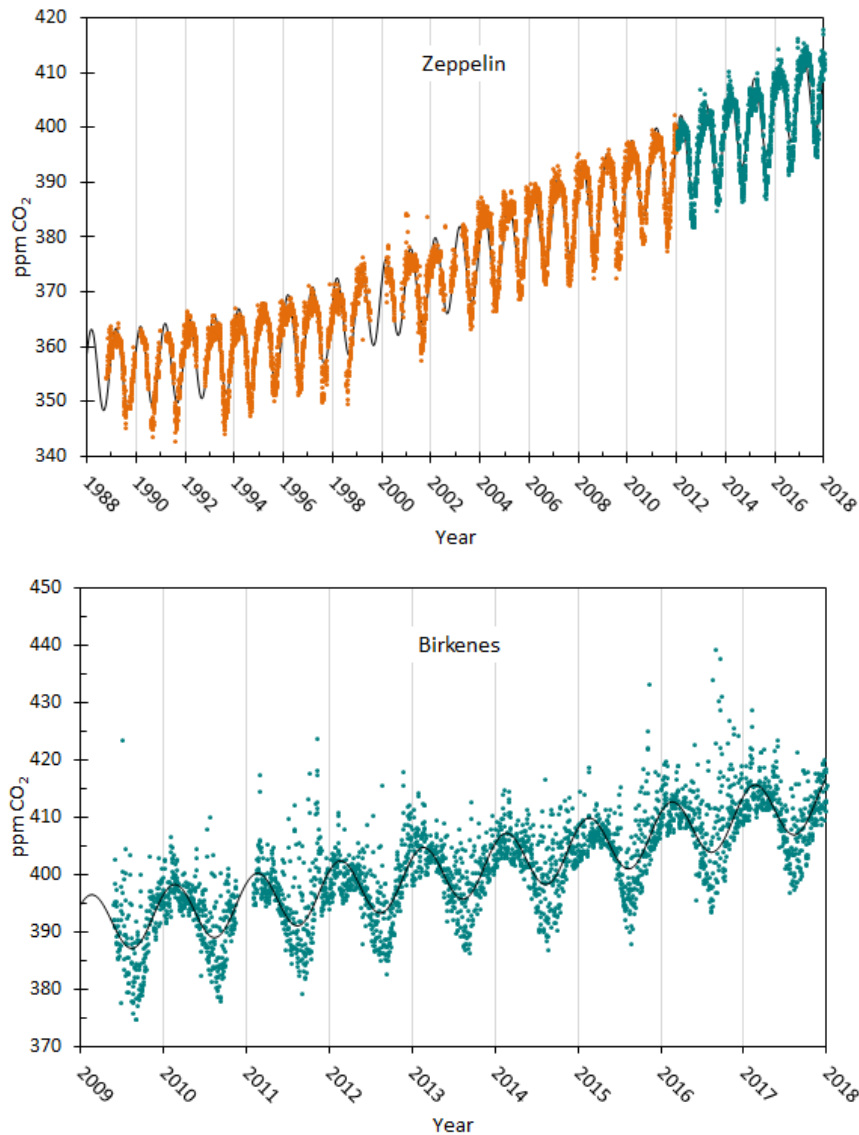


Figure 9: The atmospheric daily mean CO_2 concentrations measured at the Zeppelin Observatory for the period mid 1988-2017 are presented in the upper panel. Prior to 2012, ITM University of Stockholm provided all data, shown in orange. The green dots represent measurements from the Picarro instrument installed by NILU in 2012. CO_2 measurements at Birkenes are shown in the lower panel, the green dots are daily mean concentrations.

The results show continuous increase since the start of the observations at both sites. As can be seen there is larger CO_2 variability at Birkenes than Zeppelin. At Zeppelin the largest variability is during winter/spring, whereas Birkenes has large variability all year around. In summer there is also a clear diurnal variation with high values during the night and lower values during daytime (not shown). This is mainly due to changes between plant photosynthesis and respiration, but also the general larger meteorological variability and diurnal change in planetary boundary layer, particularly during summer, contributes to larger variations in the concentrations. In addition to the diurnal variations, there are also episodes with higher CO_2 levels at both sites due to long range transport of pollution. In general, there are high levels when the meteorological situation results in transport from Central Europe or United Kingdom at Birkenes, and from central Europe or Russia at Zeppelin.

The maximum daily mean CO₂ value in 2017 at Birkenes was 428.8 ppm, measured on 3rd February. During this period the wind direction and atmospheric circulation pattern indicate that air was transported from central Europe. At Zeppelin the highest daily mean value in 2017 was 418.1 ppm, measured 5th December. This day the air at Zeppelin had been transported from Eastern Europe and Western Siberia, areas with large oil and gas installations.

Figure 10 shows the development of the annual mean concentrations of CO₂ measured at Zeppelin for the period 1988-2017 (in orange) together with the values from Birkenes (in green) for the period 2009-2017. The global annual mean values as given by WMO (WMO, 2017) are in black. The yearly annual changes at Zeppelin and Birkenes are shown in the lower panel.

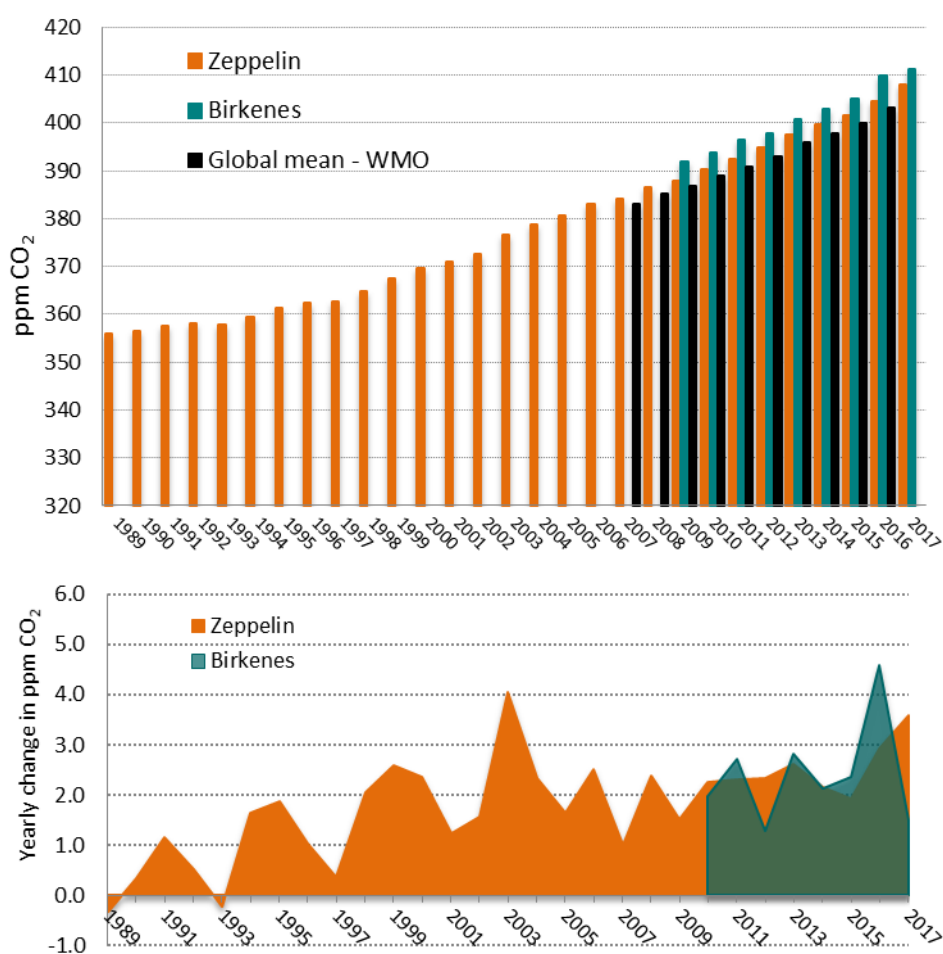


Figure 10: Upper panel: the annual mean concentrations of CO₂ measured at Zeppelin Observatory for the period 1989-2017 shown in orange. Prior to 2012, ITM University of Stockholm provides all data. The annual mean values from Birkenes are shown as green bars. The global mean values as given by WMO (2017) are included in black. The yearly annual changes are shown in the lower panel, orange for Zeppelin, green for Birkenes.

The annual mean CO₂ values at Birkenes and Zeppelin are higher than the global annual mean value as there are more anthropogenic sources and pollution at the Northern hemisphere. The

mixing of air between the two hemispheres takes time, ca 2-3 years. The annual CO₂ mean in 2017, shown in Table 3 and Figure 10 (upper panel), was 411.3 ppm and 408.0 ppm at Birkenes and Zeppelin, respectively. The annual CO₂ changes illustrated in Figure 10, lower panel, shows an increase of 3.6 ppm at Zeppelin from 2016 to 2017. This is the second highest increase registered since the measurements started in 1988. At Birkenes the CO₂ increase from 2016 to 2017 was 1.5 ppm, which is relatively moderate compared to the record high 2015-2016 growth of 4.6 ppm. The global mean annual CO₂ increase from 2016 to 2017 is not yet official, but preliminary analyses suggest that the increase is less pronounced than last year.

Trend analyses based on method described in Appendix II show that CO₂ at Birkenes and Zeppelin increased by 2.5 and 2.6 ppb/yr, respectively. This corresponds to an increase of 0.6-0.7%/yr. These trend analyses are based on data from Picarro instruments, only (which were installed by NILU in 2012). If we use the entire Zeppelin time series back to 1988, the trend is less pronounced (-0.5%/yr), indicating that the CO₂ increase has intensified over the last decade. Note that the CO₂ time series from the Picarrros are relatively short and the reported trends should be used with caution. In short time series the start and end year might have large impact on the trend results.

A paper in Nature Geoscience (Myhre et al, 2017) comments on the relation between CO₂ forcing and the CO₂ concentrations. Halfway to a doubling in the CO₂ concentration since 1750 is 417 ppm, which will be reached before 2025 with current CO₂ growth rates. However, with a global mean CO₂ abundance in 2016 at 403 ppm the halfway point to a doubling of CO₂ in terms of radiative forcing has already been reached. Hence, at CO₂ concentrations between 393 ppm and 417 ppm we are more than a halfway to a doubling of CO₂ in terms of radiative forcing, but not in concentration (see also section 1.4.1).

Key findings for CO₂: The CO₂ concentrations have increased all years subsequently, in accordance with accumulation of gas in the atmosphere and the global development and increase in anthropogenic emissions. The annual CO₂ means in 2017 reached new record levels: 408.0 ppm at Zeppelin and 411.3 ppm at Birkenes. The increases from 2016 to 2017 are 3.6 ppm and 1.5 ppm, respectively. In general, the CO₂ increase has been high last years and it is urgent to understand the reasons, including the impact of natural sources and sinks on the annual variations.

2.1.2 Methane (CH₄)

Methane (CH₄) is the second most important greenhouse gas from human activity after CO₂. IPCC reported a radiative forcing is 0.48 W m⁻² since 1750 and up to 2011 (Myhre et al., 2013b), but as high as 0.97 W m⁻² for the emission based radiative forcing (Figure 5, page 21) due to complex atmospheric effects. Etminan et al. (2016) presented revised forcing estimates for all main greenhouse gases (see section see section 1.4.1 at page 22), and for CH₄ this resulted in 25% stronger forcing than given in the IPCC Fifth Assessment Report, increasing from 0.48 W m⁻² to 0.61 W m⁻². This emphasises the importance of understanding the recent changes in CH₄.

In addition to being a dominant greenhouse gas, methane also plays central role in the atmospheric chemistry. The atmospheric lifetime of methane is approx. 12 years, when indirect effects are included, as explained in section 1.4.

The main sources of methane include boreal and tropical wetlands, rice paddies, emission from ruminant animals, biomass burning, and extraction and combustion of fossil fuels. Further, methane is the principal component of natural gas and e.g. leakage from pipelines; off-shore and on-shore installations are a known source of atmospheric methane. The distribution between natural and anthropogenic sources is approximately 40% natural sources, and 60% of the sources are direct result of anthropogenic emissions. Of natural sources there is a large unknown potential methane source under the ocean floor, so called methane hydrates and seeps. Further, a large unknown amount of carbon is captured in the permafrost layer in Siberia and North America and this might be released as methane if the permafrost layer thaws as a feedback to climate change.

The average CH₄ concentration in the atmosphere is determined by a balance between emission from the various sources and reaction and removal by free hydroxyl radicals (OH) to produce water and CO₂. A small fraction is also removed by surface deposition. Since the reaction with OH also represents a significant loss path for the oxidant OH, additional CH₄ emission will consume additional OH and thereby increasing the CH₄ lifetime, implying further increases in atmospheric CH₄ concentrations (Isaksen and Hov, 1987; Prather et al., 2001). The OH radical has a crucial role in the tropospheric chemistry by reactions with many emitted components and is responsible for the cleaning of the atmosphere (e.g. removal of CO, hydrocarbons, HFCs, and others). A stratospheric impact of CH₄ is due to the fact that CH₄ contributes to water vapour build up in this part of the atmosphere, influencing and reducing stratospheric ozone.

The concentration of CH₄ was, after a strong increase during the 20th century, relatively stable over the period 1998-2006 (Dalsøren et al, 2016). The global average change was close to zero for this period, also at Zeppelin. Recently (since 2005) an increase in the CH₄ levels is evident from our observations both at Zeppelin and Birkenes as well as observations at other sites, and in the global mean (see e.g. section 2.2.1.1.2 in Hartmann et al, 2013).

Figure 11 shows daily mean observations of CH₄ at Zeppelin since the start in 2001 (upper panel) and Birkenes since start in 2009 (lower panel). As can be seen from the figures there has been an increase in the concentrations of CH₄ at both sites the last years, and in general the concentrations are higher at Birkenes than at Zeppelin. The highest ambient CH₄ concentration ever detected at Zeppelin, 2016.3 ppb, was measured on the 5th December 2017, the same day as maximum CO₂ concentrations were recorded. The transport pattern this day indicates strong influence from Eastern Europe and Western Siberia. Fugitive emissions from Russian gas installations is normally a source of high CH₄ values. However, both CO and CO₂ levels were also very high this day, indicating an industrial pollution episode, and influence of Russian urban areas as well.

The maximum CH₄ daily mean value recorded at Birkenes in 2017 was 2051 ppb, measured 3rd February. Again, this was the same day as the maximum CO₂ level at Birkenes was recorded. Trajectory analyses this day indicate that the polluted air originated from central Europe.

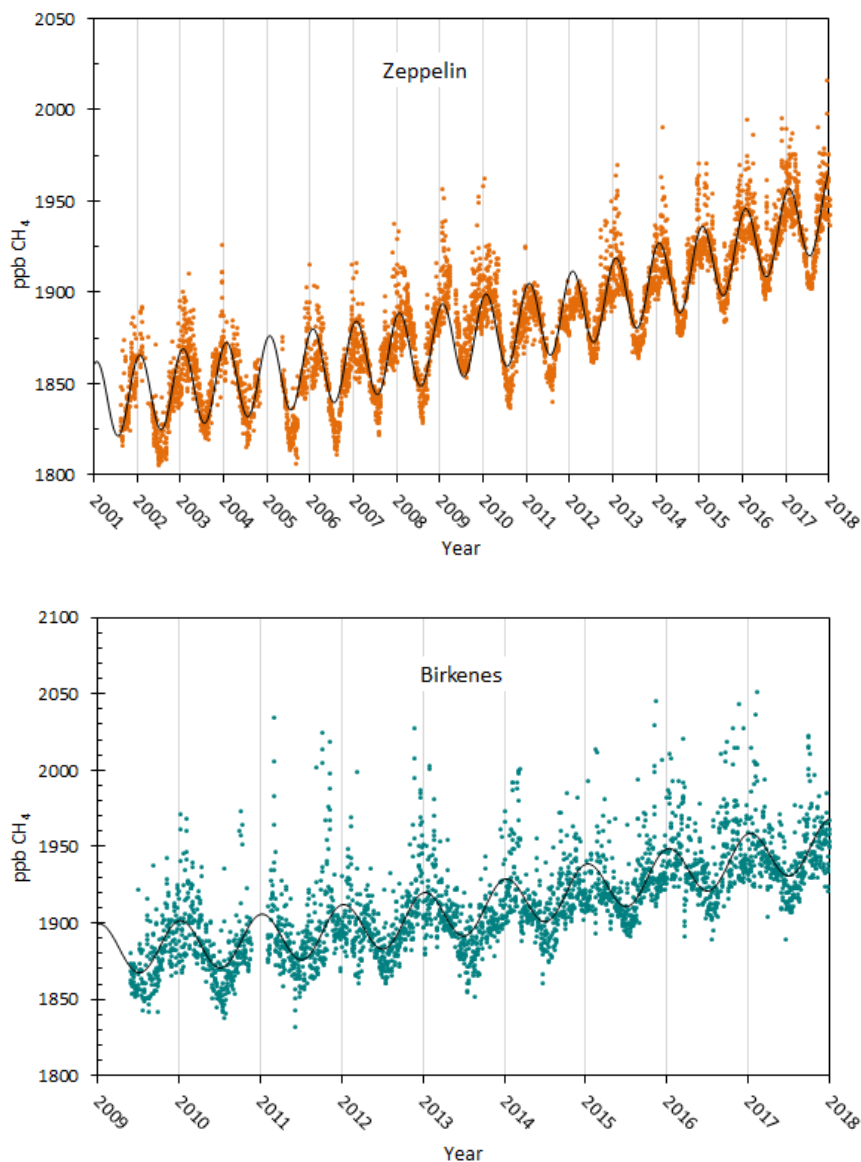


Figure 11: The upper panel shows observations of daily averaged methane mixing ratio for the period 2001-2017 at the Zeppelin Observatory. The black solid line is empirical fitted methane mixing ratio. The lower panel shows the daily mean observations for Birkenes (green dots).

For both Zeppelin and Birkenes, the seasonal variations are clearly visible, although stronger at Birkenes than Zeppelin. This is due longer distance to the sources at Zeppelin, and thus the sink through reaction with OH dominates the variation. The larger variations at Birkenes are explained by both the regional sources in Norway, as well as a stronger impact of pollution transported from central Europe or UK. For the daily mean in Figure 11, the measurements show very special characteristics in 2010 and 2011 at Zeppelin. There is remarkably lower variability in the daily mean in 2011 with fewer episodes than the typical situation in previous and subsequent years (e.g. summer/autumn 2012). This seem to be due to less impact of episodes from Northern Russia (Thompson *et al*, 2017).

At Zeppelin there are now 17 years of data, for which the trend has been calculated. To retrieve the annual trend in methane for the entire period the observations have been fitted by an empirical equation, shown as the black solid line in Figure 11. This corresponds to a

trend of 5.9 ppb/yr or 0.3%/yr. For Birkenes, shown in Figure 11 lower panel, the time series is shorter and the trend analysis is less accurate. However, indicative trend calculations demonstrate a CH₄ increase of 8.2 ppb/yr (0.4%/yr) for the period 2009-2017.

The methane increase over the last years is visualised in Figure 12, showing the CH₄ annual mean concentration at Zeppelin for the period 2001-2017 (orange) and Birkenes for the period 2009-2017 (green). The global mean values given by WMO from 2005-2016 (WMO, 2017) are included for comparison. The big difference between global mean, and the observations at Zeppelin and Birkenes is due to the fact that most emissions are in the Northern hemisphere, and global mixing and dilution takes some time.

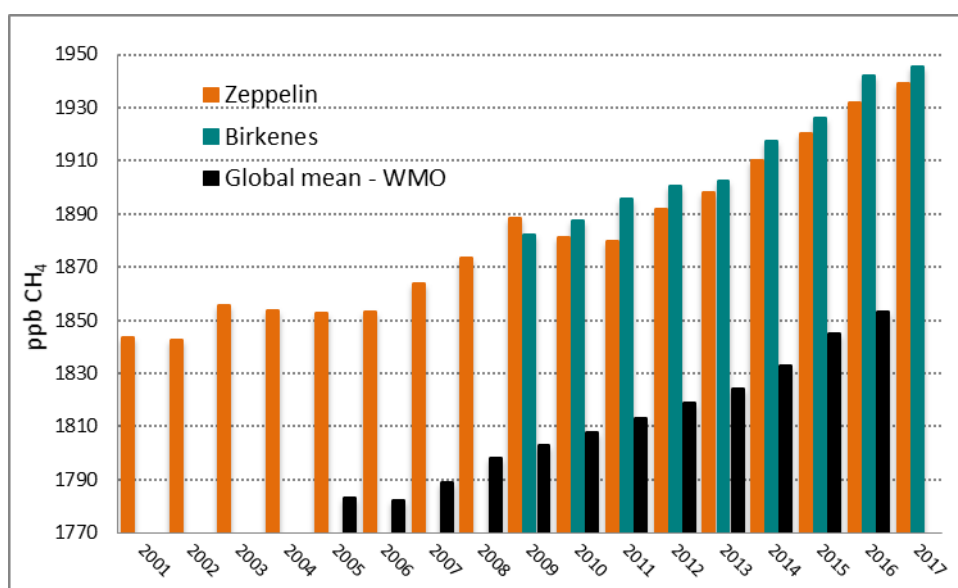


Figure 12: Development of the annual mean mixing ratio of methane in ppb measured at the Zeppelin Observatory (orange bars) for the period 2001-2017, Birkenes for the period 2010-2017 in green bars, compared to global mean provided by WMO as black bars (WMO, 2017).

The annual means are mainly based on measured methane values, however, model fitted values are used to fill in gaps if measurements are missing. Figure 12 demonstrates a gradual increase in the concentrations of methane at both Zeppelin and Birkenes the last decade. The annual mean concentrations in 2017 were 1939 and 1945 at Zeppelin and Birkenes, respectively. These values are 7 ppb and 3 ppb higher, respectively, than the annual mean values in 2016. Even if the increase is less dramatic than previous year, the enhanced methane concentrations are of concern. The increase since 2005 at Zeppelin is 86 ppb (approx. 4.6 %) which is high compared to the development of the methane mixing ratio for the period 1999-2005, both at Zeppelin and globally.

2.1.2.1 Observations of the atmospheric $\delta^{13}\text{C}_{\text{CH}_4}$ and discussion of potential change in CH_4 sources

The isotopic signature of CH_4 (expressed as $\delta^{13}\text{C}_{\text{CH}_4}$) varies by emission source (France et al., 2016, see Figure 13) and measurements of the isotopic signature provides very valuable information for the understanding of the development of CH_4 sources, complementing other measurements (e.g. CH_4 , CO and ethane). As can be seen from Figure 13 the isotopic signatures of CH_4 from e.g. biomass burning is different from natural gas. Leaks from gas installations, world-wide, both onshore and offshore might be an increasing source.

Hence, it is essential to find out if the increase since 2005 is due to high emissions from point sources, or if it is caused by newly initiated processes releasing methane to the atmosphere e.g. the thawing of the permafrost layer or processes in the ocean, both related to permafrost and others. As a part of a research project under Norwegian Research Council (2011-2013⁹) NILU initiated measurements of $\delta^{13}\text{C}_{\text{CH}_4}$ at Zeppelin in the year 2012 to provide more insight into Arctic sources of CH_4 , and potential changes. The continuation of these measurements were implemented into the Norwegian Monitoring programme in 2017, and the results are now included in this reports for the first time. Figure 14 shows the full time series of $\delta^{13}\text{C}_{\text{CH}_4}$ since May 2012 (red dots), together with ambient CH_4 concentrations (blue line) measured at the Zeppelin Observatory. The observations are mainly 5 times per week, performed in the morning. (For details about the measurements, see Appendix II).

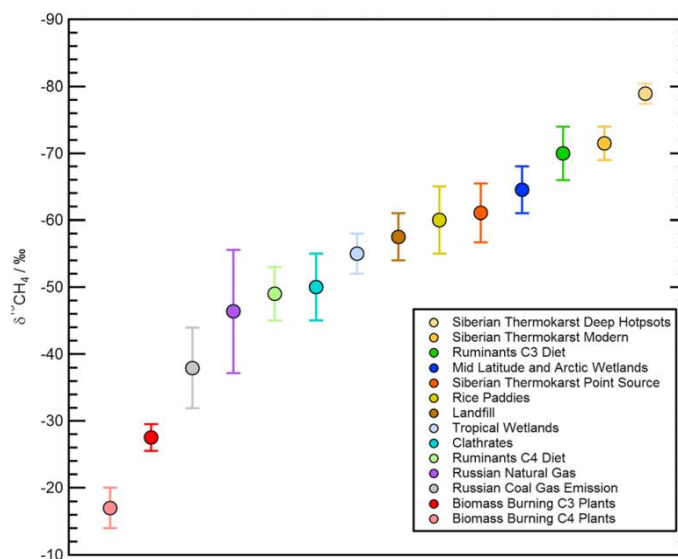


Figure 13: Isotopic ranges of $\delta^{13}\text{C}_{\text{CH}_4}$ for a variety of CH_4 sources as given in France et al, 2016.

⁹ <https://www.forskningsradet.no/prosjektbanken/#/project/NFR/207587>

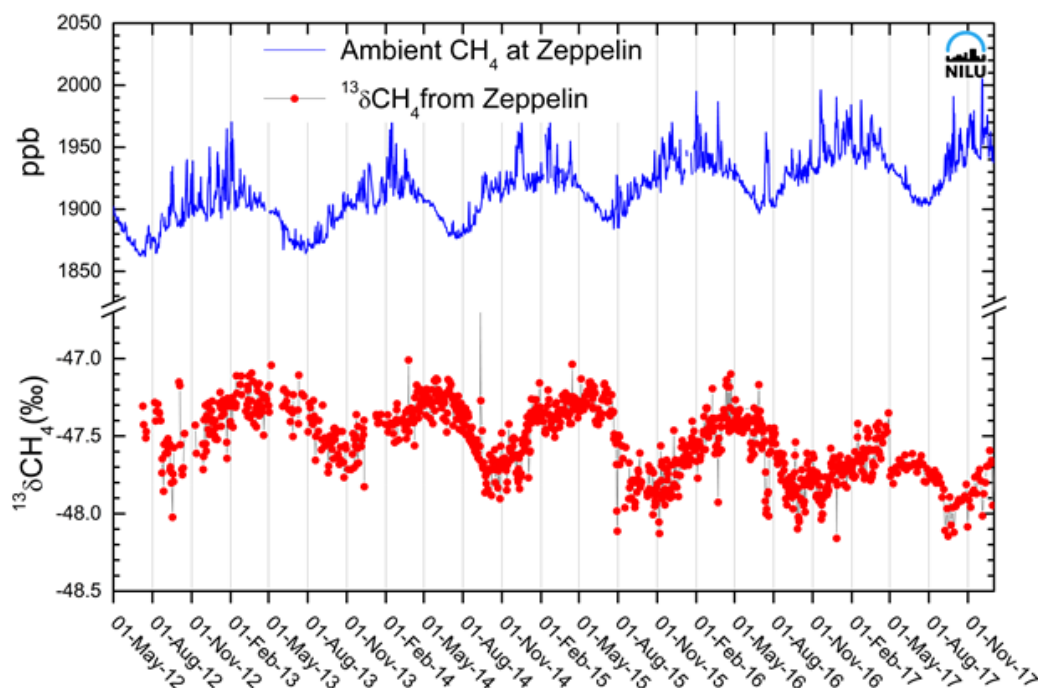


Figure 14: Long term measurements of methane (CH_4 , blue) and the ^{13}C isotopic signature of CH_4 ($\delta^{13}\text{C}-\text{CH}_4$, red) at the Zeppelin Observatory, Svalbard 78°N .

A clear reduction (shift towards more negative values) in $\delta^{13}\text{C}_{\text{CH}_4}$ is evident after 2012, parallel to the increase in the ambient concentration. Back in the 1980's, $\delta^{13}\text{C}_{\text{CH}_4}$ shifted to more positive values, indicative of gas leaks and coal emissions ($\delta^{13}\text{C}_{\text{CH}_4}$ of fossil fuels is slightly higher than the ambient average), but it seems that increases in CH_4 concentrations since 2005 have been accompanied by a negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$, as illustrated in Figure 14. These recent negative shifts in ambient $\delta^{13}\text{C}_{\text{CH}_4}$, and increases in the CH_4 mixing ratio imply changes in the balance of sources and sinks, i.e. since changes in total fossil fuel emissions cannot account for a negative shift they also cannot entirely account for increases in the mixing ratio. Hence the development in $\delta^{13}\text{C}_{\text{CH}_4}$ observed at Zeppelin suggests a role for biosphere and/or agriculture (wetland or ruminant) emissions, since these do have strongly negative $\delta^{13}\text{C}_{\text{CH}_4}$ compared to ambient values and fossil sources. Changes in the sink strength (reaction with hydroxyl radicals, OH) is also a possibility. It is worth noting that as the major sink for almost all atmospheric Volatile Organic Compounds (VOCs), (not only CH_4), changes in OH would have profound implications for the whole atmosphere. There are a few signs of that. Ethane and methane are emitted together from fossil oil and gas sources, and the global decrease in ethane supports the hypothesis that wetland changes is a large contributor to the change in CH_4 . Additionally, recent and ongoing scientific discussions point in the direction of increased emissions from wetlands located both in the tropical region and in the Arctic region. Thawing of permafrost, both in terrestrial regions and in marine region, might introduce new possible methane emission sources initiated by the temperature increase the last years in the Arctic region. Gas hydrates at the sea floor are widespread in thick sediments in this area between Spitsbergen and Greenland. If the sea bottom warms, this might initiate further emissions from this source. This was the core of the large polar research project *MOCA - Methane Emissions from the Arctic Ocean to the Atmosphere*:

*Present and Future Climate Effects*¹⁰, which started at NILU in October 2013, and was finalized spring 2017. A few results from this is included in the section 2.1.2.2 at page 36.

To summarise, isotopic signatures provide very valuable data for understanding the development of the source of CH₄, but careful interpretation of observational data and emission inventories is required before assigning observed shifts to a particular source. This further demonstrates the need for observational data on emission fluxes from the large natural reservoirs. There is most likely a combination of causes explaining the increase in methane the last years, and the dominating reason is not clear. A possible explanation is increased methane emissions from wetlands, both in the tropics as well as in the Arctic region, in addition to increases in emission from the fossil fuel industry in some areas, but this alone cannot explain the development in the isotopic signature of CH₄ at Zeppelin.

2.1.2.2 Methane from Arctic Ocean to the atmosphere?

A comprehensive research project “*MOCA - Methane Emissions from the Arctic Ocean to the Atmosphere: Present and Future Climate Effects*” ended in 2017 focusing on methane from Arctic Ocean to the atmosphere. This was a collaboration with CAGE (The Centre for Arctic Gas Hydrate, Environment and Climate, at UiT The Arctic University of Norway) and Cicero, but lead by NILU. The driving questions for MOCA were:

- I. What is the status and current release of methane from marine seep sites and methane hydrates in the Arctic Ocean, and specifically around Svalbard?
- II. How are these processes depending on trends in sea temperature and annual variations?
- III. Where are the most important areas in the Arctic Ocean which could constitute a possible source of atmospheric methane, now and in the future?
- IV. What is the present CH₄ emission from the seabed to the atmosphere?
- V. What is the most likely change in flux, the next 50 and 100 years under realistic climate scenarios? And what is the global effect of this?
- VI. What is the ocean temperature threshold for large changes in emission of methane from hydrates?

What are gas hydrates?

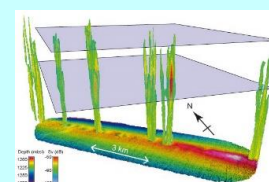
Large amounts of natural gas, mainly methane, are stored in the form of hydrates in continental margins worldwide, particularly, in the Arctic. Gas hydrate consists of ice-like crystalline solids of water molecules



encaging gas molecules, and is often referred to as ‘the ice that burns’.

What are seeps?

Cold seeps are locations where hydrocarbons are emitted from sub-seabed gas reservoirs into the ocean. This can be both from petroleum reservoirs and methane hydrates. The illustration is gas bubbles rising 800 m up from Vestnesa Ridge, offshore Svalbard (Smith et al. 2014).



<http://cage.uit.no>

¹⁰ <http://moca.nilu.no/>

In this project we used integrated approaches to answer these questions. Figure 15 shows the exploratory platforms involved. Comprehensive measurement campaigns involving ship, aircrafts and Zeppelin have been performed. Additionally we used models both atmospheric chemistry, methane hydrates modelling at sea floor, and climate models.

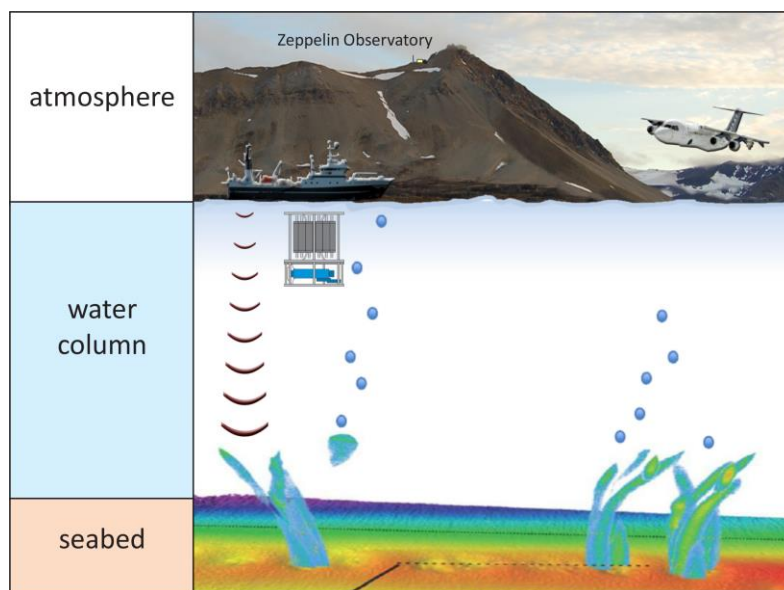


Figure 15: Field campaign and measurement platforms at the sea floor, in the water column, and in the atmosphere west of Svalbard in June-July 2014. Seeps on the seafloor, represented here by swath bathymetry, release gas bubbles that rise through the water column. The Research Vessel Helmer Hanssen detected gas bubbles and collected water samples at various depths, and provided online atmospheric CH₄, CO and CO₂ mixing ratios and discrete sampling of complementary trace gases and isotopic ratios. The Facility of Airborne Atmospheric Measurements (FAAM) aircraft measured numerous gases in the atmosphere, and an extended measurement programme was performed at the Zeppelin Observatory close to Ny-Ålesund. Data from Zeppelin was used for comparison to detect possible oceanic sources.

The main message from MOCA is that CH₄ gas released from the Arctic seabed can lead to an increased methane concentration in the ocean, but surprisingly, very little of the methane gas rising up through the sea appears to reach the atmosphere in the summer. This was an important message to bring to the debate on the state of the ocean and atmospheric system in the Arctic. It is important to emphasize that the Arctic has in recent years experienced major changes and average temperatures well above normal values, and this can change and also be different in other time of the year. Summer was in focus in this work. We used different state-of-the art models to assess and understand the variation in Arctic methane, to investigate potential oceanic sources in various regions, in particular methane hydrates and cold seeps. Results are presented in Myhre et al. (2016), Pisso et al. (2016), Dalsøren et al (2018), Thompson et al (2017), and some information can be found here: <http://forskning.no/havforskning-klima-arktisk/2016/05/metan-slipper-ikke-ut-av-polhavet-om-sommeren>. More papers are in the final stage (Platt et al, 2018), and will add important information to our understanding of CH₄ and ethane and the interaction between ocean and atmosphere both now and in the future.

Key findings for Methane: In 2017 the mixing ratios of methane increased to new record levels at both Zeppelin and Birkenes. At Zeppelin the annual mean value reached 1939 ppb, which is an increase of 7 ppb since 2016. At Birkenes the annual mean value in 2017 was 1945 ppb, 3 ppb higher than previous year. The methane changes at Zeppelin the last 10 years are

large compared to previous periods, thus it is important with continued research focus to improve the understanding of methane sources and sinks.

2.1.3 Nitrous Oxide (N₂O)

Nitrous Oxide (N₂O) is a greenhouse gas with both natural and anthropogenic sources. The sources include oceans, tropical forests, soil, biomass burning, cultivated soil and use of particular synthetic fertilizer, and various industrial processes. There are high uncertainties in the major soil, agricultural, combustion and oceanic sources of N₂O. Also frozen peat soils in Arctic tundra is reported as a potential source (Repo et al., 2009), but studies identify tropical and sub-tropical regions as the largest source regions (Thompson et al, 2013). N₂O is an important greenhouse gas with a radiative forcing of 0.17 W m⁻² since 1750, contributing around 6 % to the overall well-mixed greenhouse gas forcing over the industrial era. N₂O is also the major source of the ozone-depleting nitric oxide (NO) and nitrogen dioxide (NO₂) in the stratosphere, thus the component is also influencing the stratospheric ozone layer. The Assessment of the ozone depletion (WMO, 2014b) suggests that current emissions of N₂O are presently the most significant substance that depletes ozone.

In 2009, NILU installed a new instrument at Zeppelin measuring N₂O with high time resolution of 15 minutes. The instrument was in full operation in April 2010 and the results for 2010 - 2017 are presented in Figure 16. The annual mean N₂O value at Zeppelin was 330 ppb in 2017, an increase of 0.94 ppb from the 2016 annual mean value. N₂O has increased from around 270 ppb prior to industrialisation and up to an average global mean of 329.8 ppb in 2017 (Dlugokencky et al., 2018)

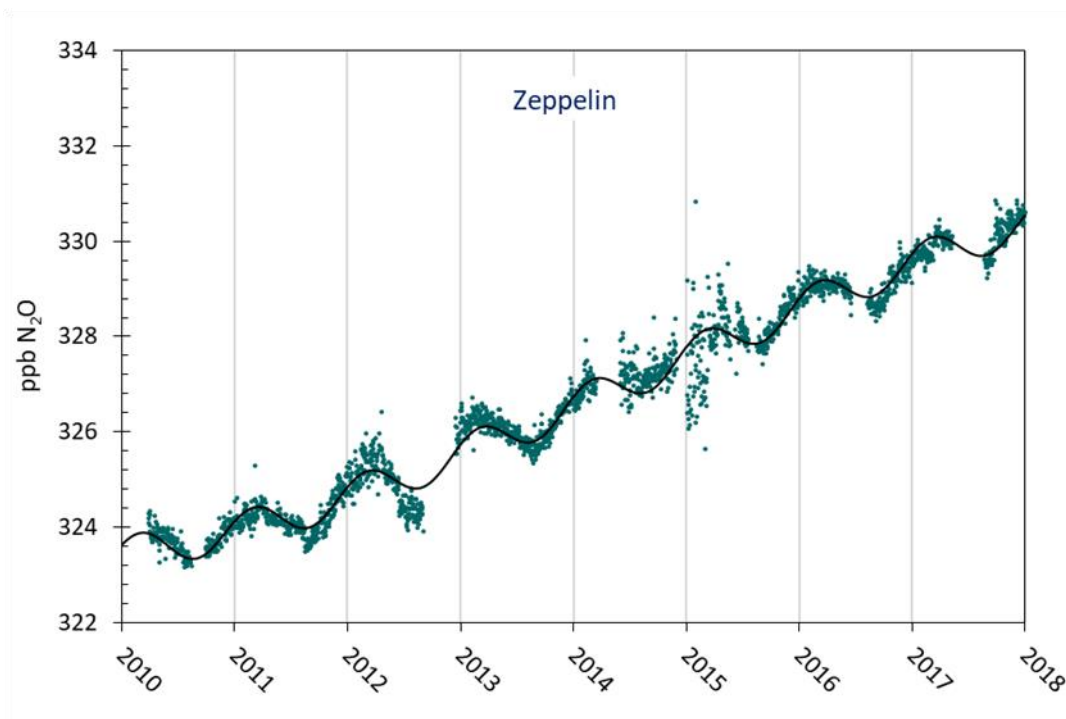


Figure 16: Measurements of N₂O at the Zeppelin Observatory for 2010-2017. The black line is empirical modelled N₂O mixing ratio.

As seen from Figure 16 there has been a gradual increase in N₂O at Zeppelin since the measurements started in 2010. For the period 2010-2017 the overall trend is 0.93 ppb/yr, an increase of 0.3%/yr. However, the time series is relatively short and has suffered from several interruptions due to instrumental problems. Thus, the time series is not ideal for reliable trend calculations.

Figure 17 shows annual average concentrations of N₂O measured at Zeppelin for the periods 2010-2017. The global annual means of N₂O (WMO, 2017) are included as a black bars for comparison. The concentrations at Zeppelin are slightly higher than the global means.

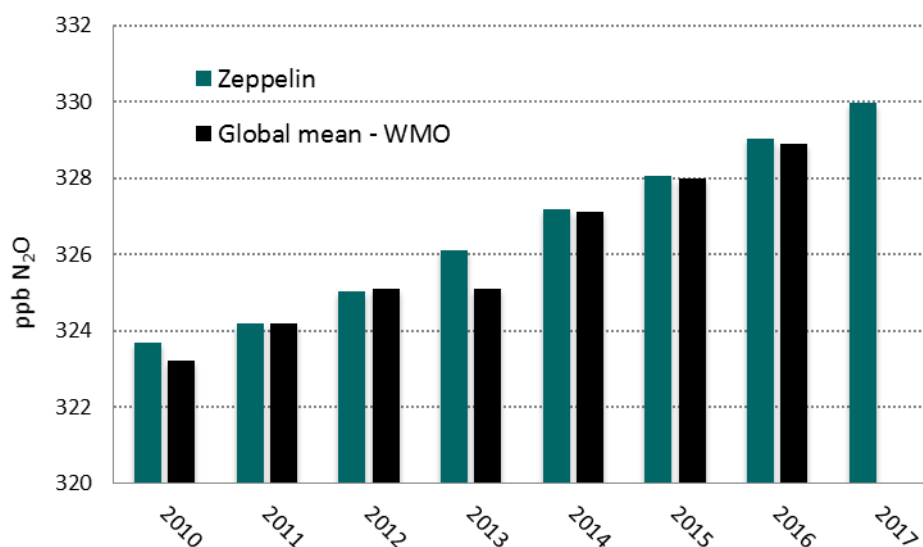


Figure 17: Annual mean concentration of N₂O at the Zeppelin Observatory for 2010-2017.

Key findings Nitrous Oxide: N₂O is a strong greenhouse gas and contributes ~6% to the total forcing of well-mixed greenhouse gases since 1750. N₂O is also the major source of the ozone-depleting NO and NO₂, thus the component is influencing the stratospheric ozone layer. The global mean level of N₂O has increased from around 270 ppb prior to industrialization and up to an average global mean of 329.8 ppb in 2017. At Zeppelin the annual mean N₂O level was 330.0 ppb in 2017.

2.1.4 Volatile organic compounds (VOC)

Volatile Organic Compounds (VOCs) are a large group of carbon-based compounds that have a high vapor pressure and easily evaporate at room temperature. VOCs oxidation contributes to the production of tropospheric ozone and influences photochemical processing, both impacting climate and air quality. Sources of VOCs (here ethane, propane, butane, pentane, benzene and toluene) include natural (mostly geological but also from wild fires) and anthropogenic (fossil fuels) ones. CH₄ and VOCs are co-emitted from oil and natural gas sources, for CH₄ to ethane the mass ratio varies from 7 to 14 (Helmig et al, 2016). The atmospheric ethane budget is not well understood and state-of-the-art atmospheric models

underestimate ethane-mixing ratios, implying that current emission inventories require additional sources to balance the global atmospheric ethane budget.

Helmig *et al.* (2016) showed, from long-term observations of ethane from a global network that concentrations are increasing (since circa 2010), and that there is a strong latitudinal gradient, with the highest abundances observed in the Arctic, and a steep decline towards the south. They concluded that emissions from North American oil and natural gas development are the primary cause of increasing concentrations in recent years. However, there might be other factors. A recent study by Nicewonger *et al.* (2016) used analysis of polar ice cores to estimate the pre-industrial emissions and concluded that natural ethane emissions from geologic seeps contributed significantly to the preindustrial ethane budget. Etiope and Ciccioli (2009) suggested that a substantial part of the missing ethane source can be attributed to gas seepage, but they did not include the Arctic in their study.

A new study in Nature Geoscience performed under the MOCA project (see section 2.1.2.2) unveils a need of revising previous ethane and propane emissions studies, as these emissions have been underestimated by more than 50%. Such revision could in turn improve our understanding of the forceful and related methane emissions, still largely enigmatic to atmospheric sciences (Dalsøren *et al.*, 2018).

There has been large progress recently in the access to quality assured measurements of VOC under ACTRIS, with high time resolution. In this new study we compare recent atmospheric measurement data from a number of sites with the simulations from an atmospheric chemistry transport model, and found that a substantial upward revision of the emissions is needed. We show that substantial geologic emissions are necessary to reproduce observations of preindustrial ethane concentrations. This provides an important constraint on both preindustrial and current natural emission budgets, reducing the relative contribution of fossil sources.

In 2010 a Medusa-GCMS instrument was installed at Zeppelin, which made it possible to perform online VOC measurements. Figure 18 shows the daily mean observations of the four non-methane hydrocarbons included in the programme in 2010: ethane, propane, butane and pentane.

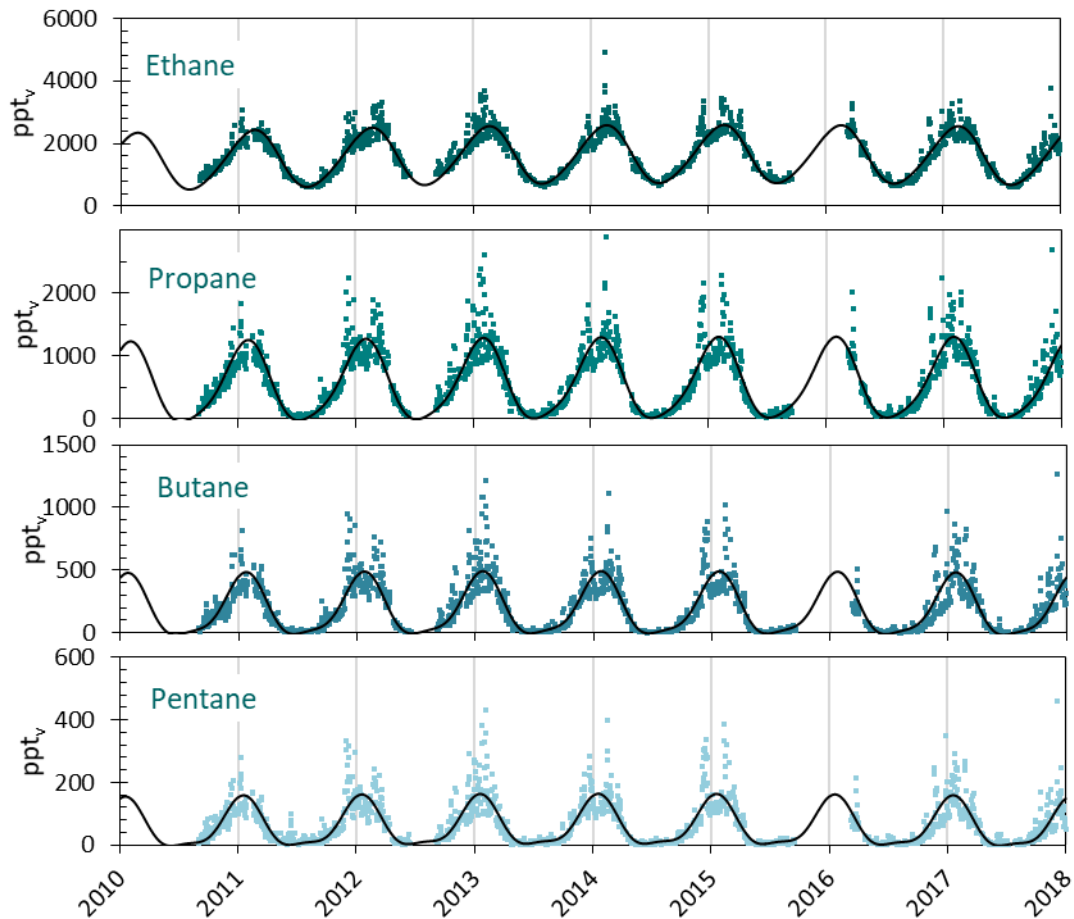


Figure 18: Observations of daily averaged mixing ratios of ethane, propane, butane, and pentane for the period September 2010 - 2017 at the Zeppelin Observatory.

Due to the short lifetimes, ranging from a few days for pentane to 2-3 months for ethane, the annual cycles are very strong and are regulated by OH reactions. The annual mean from 2011 to 2017 are shown in Figure 19.

As seen from the figure the annual mean concentrations vary from one year to another and for most compounds it is not possible to draw any conclusions about development and trend.

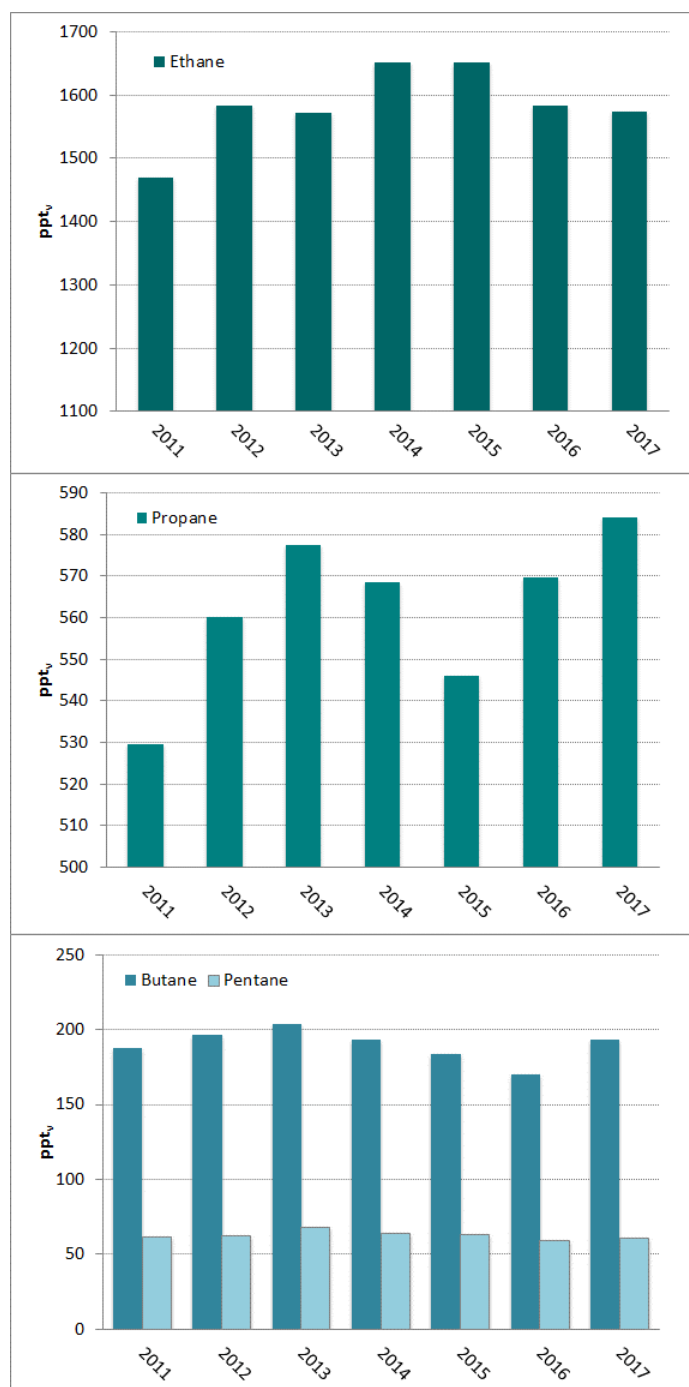


Figure 19: Development of the annual means of the measured non-methane hydrocarbons at the Zeppelin Observatory for the period 2011-2017. Upper panel in dark green: ethane, mid-panel propane, and lower panel butane and pentane. All concentrations are in ppt_v.

At Zeppelin two other cyclic heavy VOCs are measured, *benzene* and *toluene*, which belong to a group of VOCs found in petroleum hydrocarbons, such as gasoline. The compounds have attracted much attention since they are considered strong carcinogens (especially benzene), but they are also relevant for climate. The VOCs have relatively short atmospheric lifetimes and small direct impact on radiative forcing. However, anthropogenic secondary organic aerosols (SOA) are formed from photo oxidation of benzene and toluene (Ng et al., 2007),

which indirectly impacts the climate. The SOA formation from these VOCs is most effective under low- NO_x conditions and when ambient concentration of organic aerosols is high. Thus, benzene and toluene influence climate through their production of secondary organic aerosols and their involvement in photochemistry, i.e., production of O_3 in the presence of light.

Figure 20 shows the daily mean observations of benzene and toluene at Zeppelin for the period 2010-2017. After an upgrade of the Medusa-GCMS in September 2017, the toluene values became unrealistic low. Thus, the toluene values measured after 25. September 2017 are flagged as “absent”.

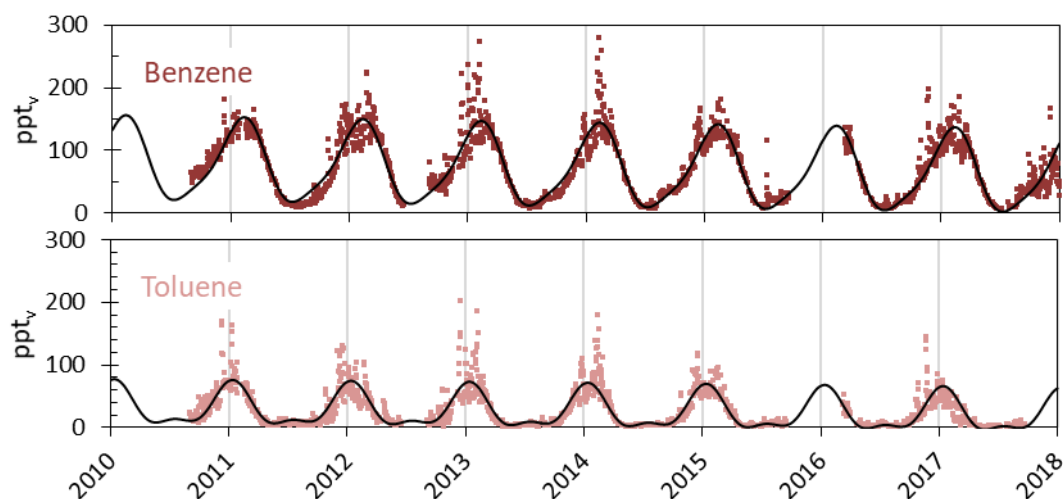


Figure 20: Observations of daily averaged mixing ratio of benzene (upper panel) and toluene (lower panel) for the period September 2010 - 2017 at the Zeppelin Observatory.

As can be seen from the figure there are strong annual variations, mainly explained by the reactions induced by sunlight. The annual means of benzene and toluene for the period 2011-2017 are presented in Figure 21. The figure shows a declining tendency for both compounds and the annual mean values in 2017 are lower than previous years. However, it should be noted that lack of toluene measurements in the fall/winter 2017 might have influenced the results, even if model data (black curve in Figure 20) are used to fill in gaps where measurements are absent.

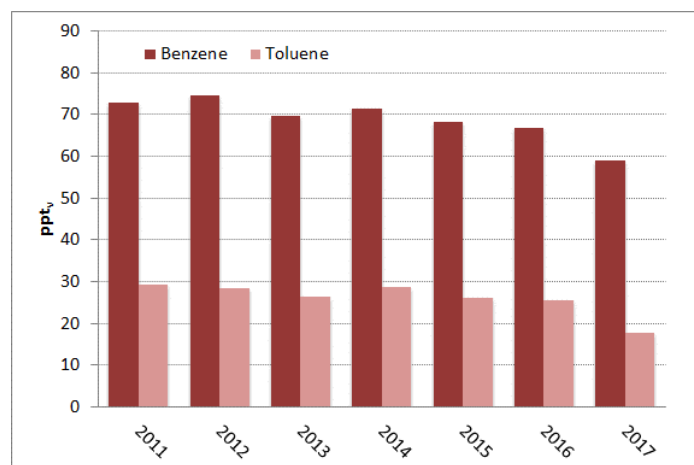


Figure 21: Development of the annual means of benzene (brown) and toluene (light red) for the period 2011 - 2017 at the Zeppelin Observatory. All concentrations are in ppt.

Key findings - VOCs: The VOCs are short lived greenhouse gases with low direct greenhouse gas effects, however, they have an indirect climate effect as they influence the levels of e.g. aerosols, ozone, and CO. Six different VOCs have been measured at Zeppelin since September 2010. The annual mean concentrations vary from one year to another and for most compounds it is not possible to draw any conclusions about developments and trends. Benzene and toluene are exceptions, where the annual means have decreased the last few years.

2.1.5 Carbon monoxide (CO)

Atmospheric carbon monoxide (CO) has both natural and anthropogenic sources. CO is produced when various organic gases are oxidized, such as methane, VOCs emitted from fossil fuel, and gases from biomass burning. Additionally, emissions from plants and ocean are important sources. CO is not considered as a direct greenhouse gas, as it does not absorb terrestrial thermal IR energy strongly enough. However, CO is able to modulate the level of methane and production of tropospheric ozone, which are both very important greenhouse gases. Hence, CO is considered as a climate gas although not a greenhouse gas. CO is closely linked to the cycles of methane and ozone and it also plays a key role in the control of OH radicals.

CO at Zeppelin is included in the current monitoring programme and the observed CO mixing ratios for the period September 2001-2017 are shown in Figure 22.

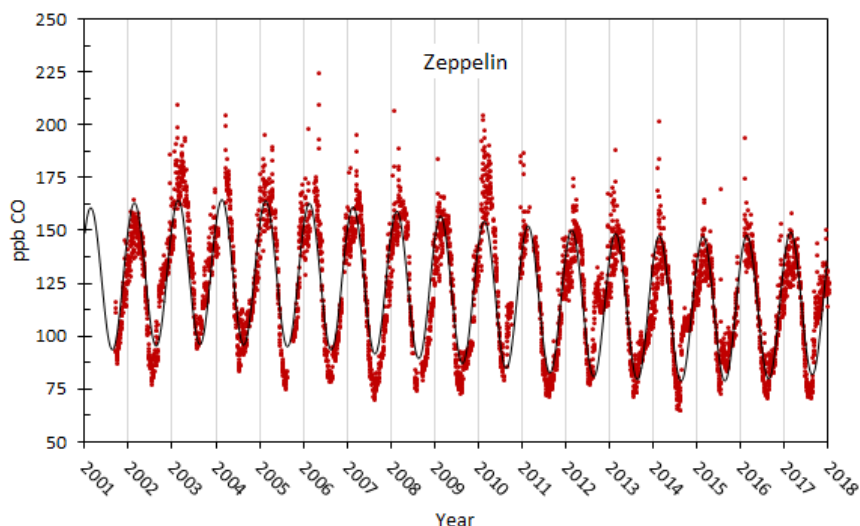


Figure 22: Observations of carbon monoxide (CO) from September 2001 to 2017 at the Zeppelin observatory. Red dots: daily averaged observed mixing ratios. The solid line is the empirical fitted mixing ratio.

The concentrations of CO show characteristic seasonal variations with a clear annual cycle with a late winter (February/March) maximum and a late summer (August) minimum. This seasonal cycle is driven by variations in OH concentration as a sink, emission by industries and biomass burning, and transportation on a large scale. As seen from the figure there are also peak values which are due to long-range transport of polluted air to Zeppelin and the Arctic. The highest mixing ratio of CO ever observed at Zeppelin is 225 ppb, measured on the 2nd of May 2006. This peak value was due to agricultural fires in Eastern Europe. In general, CO is an excellent tracer for transport of smoke from fires (biomass burning, agricultural- or forest fires).

The maximum CO value at Zeppelin in 2017 was 159 ppb, measured on 23rd of February. This day the air was transported from Russia. However, there were no episodes with extreme CO concentrations in 2017.

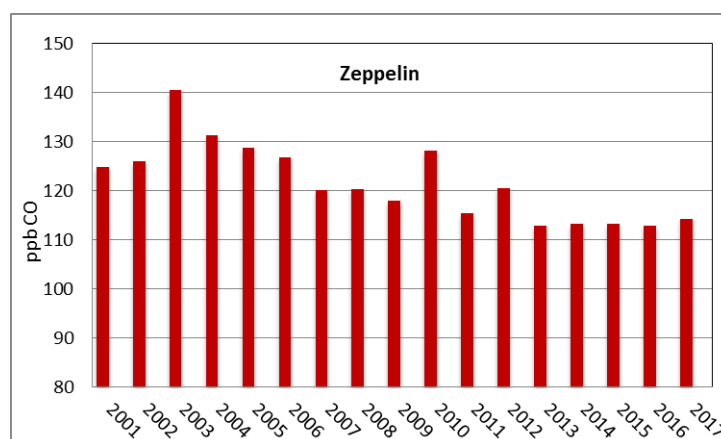


Figure 23: Development of the annual means of CO measured at the Zeppelin Observatory for the period 2001-2017. The concentrations are in ppb.

The development of the annual CO means at Zeppelin for the period 2001-2017 is presented in Figure 23. In general the CO concentration at Zeppelin shows a decrease during the period 2003 to 2009, and stable levels the last years with a small peak in 2010. For the period 2001-2017 a trend of -1.3 ppb/yr is calculated.

Key findings CO: CO is not a direct greenhouse gas, but it is still considered as a climate gas as it regulates the levels of methane and ozone. CO is also an excellent tracer for long-range transport of smoke from fires. CO has been measured at Zeppelin since 2001. For the period 2001-2017 a decreasing trend of -1.3 ppb/yr is recorded. In 2017 no strong CO transport episodes were registered at Zeppelin.

2.1.6 Chloromethane at the Zeppelin Observatory

Chloromethane (also called methyl chloride, CH_3Cl) is the most abundant chlorine containing organic gas in the atmosphere. It contributes approximately 16% to the total chlorine from the well-mixed gases in the troposphere (WMO, 2014b), and through this a strong contributor to ozone depletion. The main sources of chloromethane are natural, and the dominating sources include ocean, biomass burning, fungi, wetlands, rice paddies, and tropical forests. Due to the dominating natural sources, this compound is not regulated through the Montreal or the Kyoto protocols. To reach the stratosphere, the lifetime in general needs to be above the order of 2-4 years, but this is also dependant on the source strength and the regional distribution of the gas. Chloromethane has a relatively high mixing ratio, and contributes to the stratospheric chlorine burden.

The result of the measurements of this gas for the period 2001-2017 is shown in Figure 24. The lifetime of chloromethane is only one year, resulting in large seasonal fluctuations due to rapid changes in emission, as shown in Figure 24.

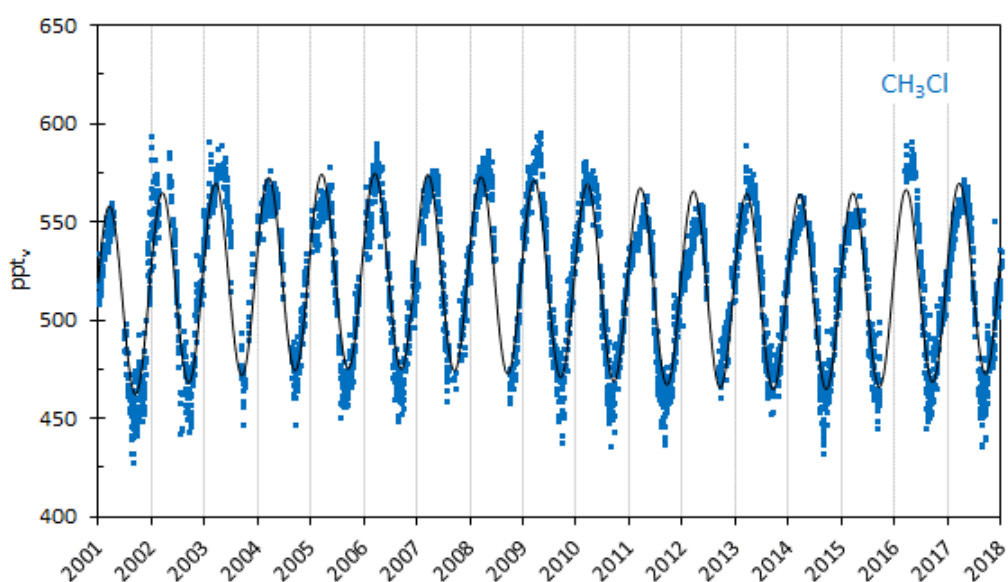


Figure 24: Observations of chloromethane, CH_3Cl , for the period 2001-2017 at the Zeppelin Observatory. Blue dots: daily averaged concentrations from the observations, solid line: empirical fitted mixing ratios.

The annual means of chloromethane for the period 2001-2017 are shown in Figure 25. Days with missing observations are filled with empirical fitted data. Only small changes have been observed since the measurements started in 2001. The trend for the period 2001-2017 is -0.17 ppt/yr (-0.5% for the entire period). From 2002 to 2009 the chloromethane concentrations were relatively stable, but since 2009 there has been larger variability and a decreasing tendency, except from 2016 where the annual mean value was 10 ppt higher than previous year. The reason for this is not clear, and sources resulting in the rapid observational changes the last years need to be investigated. The black bars in Figure 25 show that the global annual means in 2016 (Hall et al., 2017) and 2017 (Dlugokencky et al., 2018) were 30-40 ppt (6-7%) higher than the annual mean value at the Zeppelin Observatory. This is likely explained by strong emission sources in the tropics, resulting in increased CH_3Cl mixing ratios towards lower latitudes (Umezawa et al., 2014).

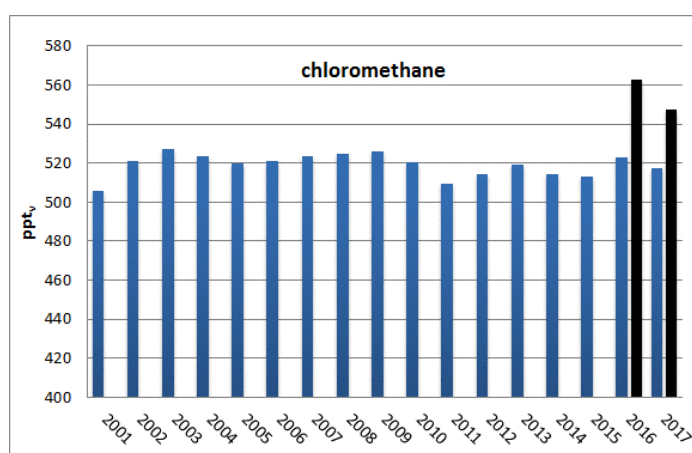


Figure 25: Development of Chloromethane annual means measured at the Zeppelin Observatory for the period 2001-2016. Global annual means for 2016 and 2017 are included as a black bars. All units are in ppt.

Key findings - Chloromethane: The atmospheric concentration of chloromethane is relatively stable. The gas has a dominating natural origin and is not regulated through the Kyoto or Montreal protocol. The CH_3Cl concentration at Zeppelin has decreased by ~0.5% since 2001.

2.1.7 Bromomethane - CH_3Br at the Zeppelin Observatory

Bromomethane (also called Methyl bromide, CH_3Br) is an important reservoir for atmospheric bromine, which reacts with ozone to deplete the ozone layer. CH_3Br has both natural and anthropogenic sources. The natural sources such as the ocean, plants, and soil, can also be a sink for this gas. The primary anthropogenic source of bromomethane has been from its use as a fumigant, e.g. for pest control and pesticide in the control of weeds. Other anthropogenic sources of CH_3Br include the combustion of leaded gasoline, biomass burning, and emissions from certain crop species (e.g. canola, rice, mustard and cabbage). Even though bromomethane is a natural substance, the additional contribution from anthropogenic sources contributes to the man-made depletion of the ozone layer. Total organic bromine from halons and bromomethane peaked in the mid-1990s, but the tropospheric concentration has

decreased by ~25% over the past 20 years. Also, the stratospheric abundance of bromide has started to decrease (WMO, 2014b).

The result of the daily averaged observations of bromomethane for the period 2001-2017 is shown in Figure 26. Bromomethane is a greenhouse gas which is twice as strong as CO₂ and has a lifetime of 0.8 years (Myhre et al, 2013b). The short lifetime explains the strong annual and seasonal variations of this compound.

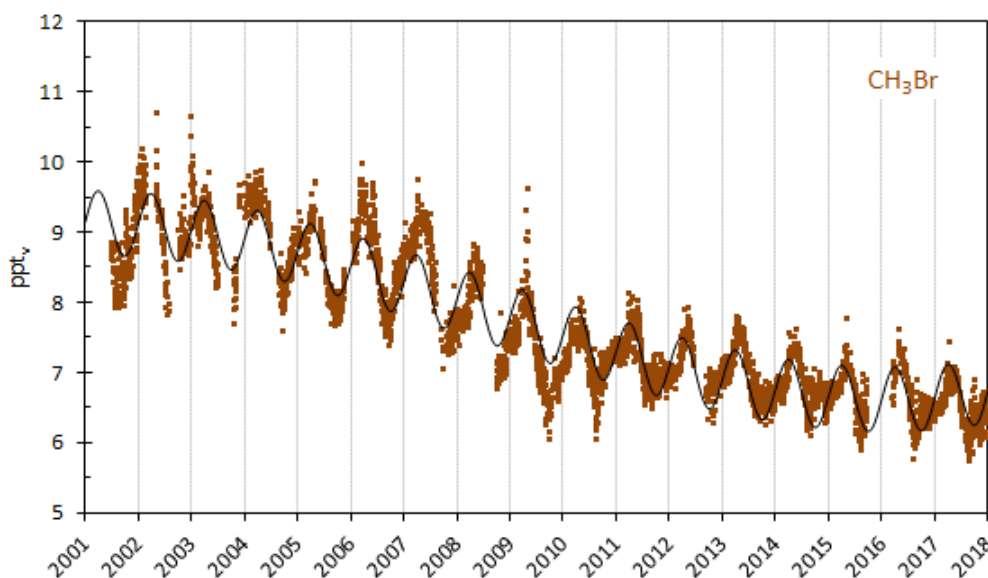


Figure 26: Observations of Bromomethane, CH₃Br, for the period 2001-2017 at the Zeppelin Observatory. Brown dots: daily averages mixing ratios from the observations, solid line empirical fitted mixing ratios.

The development of the annual means for the period 2001-2017 is presented in Figure 27. For this period there is a trend in the mixing ratio of -0.18 ppt/yr. The overall observed change since 2001 is -2.5 ppt, i.e. -28%. Similar to CH₃Cl, the annual mean CH₃Br concentration in 2016 was higher than previous year. However, in 2017 the concentration continued to decrease and the annual average value was 6.5 ppt. CH₃Br shares many natural sources and sinks with CH₃Cl, which probably explains the high 2016 concentrations for both species. In general, the decline in bromomethane is explained by considerable reduction in emissions; The uses of CH₃Br has decreased steadily as a result of the implementation of the Montreal Protocol, from over 50 000 tonnes/yr in the late 1990s to about 4 000 tonnes/yr in 2012 (WMO, 2014b).

The global mean mixing ratio published by BAMS in “State of the Climate” (Hall et al., 2017; Dlugokencky et al., 2018), presented by the black bars in Figure 27, was 6.6 ppt in 2017. This is close to the annual mean value observed at the Zeppelin Observatory.

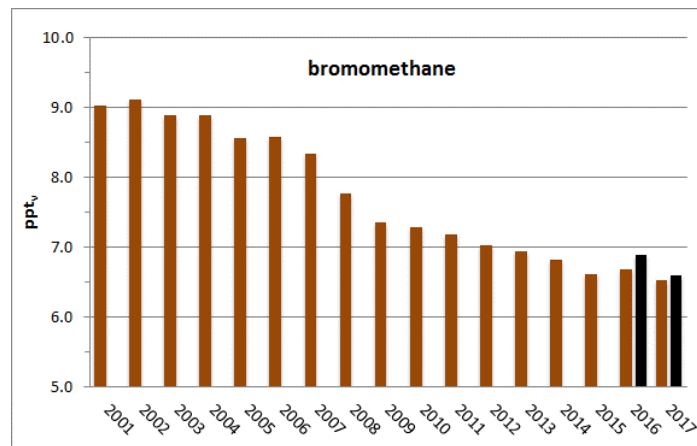


Figure 27: Development of the annual means of bromomethane measured at the Zeppelin Observatory for the period 2001-2017. The global annual mean for 2016 and 2017 are included as black bars.

Key findings - Bromomethane: The atmospheric concentration of CH₃Br at Zeppelin has decreased by ~28% since the observations started in 2001. The decrease is a result of the implementation of the Montreal Protocol. However, CH₃Br has many natural sources with uncontrolled fluctuations from one year to another.

2.2 Greenhouse gases with solely anthropogenic sources

All the gases presented in this chapter have solely anthropogenic sources. These man-made greenhouse gases include CFCs, HCFCs, HFCs, SF₆, NF₃, SO₂F₂ and halons, and most of these gases did not exist in the atmosphere before the 20th century. Although the gases have much lower concentration levels than most of the natural gases mentioned in the previous section, they are strong infrared absorbers, many of them with extremely long atmospheric lifetimes resulting in high global warming potentials (see Table 3). Together as a group, the gases contribute to around 12% to the overall global radiative forcing since 1750 (Myhre et al, 2013b). The annual mean concentrations for all years and all gases included in the monitoring programme are given in Appendix I (Table A 1, page 111), whereas trends, uncertainties and regression coefficients are found in Table A 2.

Some of these gases are ozone depleting, and consequently regulated through the Montreal protocol. Especially chlorine and bromine from CFCs, HCFCs and halons contribute to the depletion of the ozone layer, allowing increased UV radiation to reach the earth's surface, with potential impact not only on human health and the environment, but on agricultural crops as well. In 1987 the Montreal Protocol was signed in order to reduce the production and use of these ozone-depleting substances (ODSs) and the amount of ODSs in the troposphere reached a maximum around 1995. The amount of most ODSs in the troposphere is now declining slowly and the concentrations are expected to be back to pre-1980 levels around year 2050. In the stratosphere the peak ODS level was reached around year 2000. According to WMO (2014b) the amount of Equivalent Effective Stratospheric Chlorine (EESC) declined by about 10% in Polar region and 15% in Mid-latitude regions from 2000 to 2012.

There are two generations of substitutes for the CFCs. The first generation substitutes are included in the Montreal protocol as they also deplete the ozone layer. This comprises the components called HCFCs listed in Table 3. The second-generation substitutes, the HFCs, are included in the Kyoto protocol. In addition the agreement in Kigali in 2016, which is an amendment to the Montreal Protocol, also aims to reduce the emission of HFCs. The general situation now is that the CFCs have started to decline, while their substitutes are increasing, and some of them are increasing rapidly.

2.2.1 Chlorofluorocarbons (CFCs) at Zeppelin Observatory

In total four chlorofluorocarbons (CFCs) are measured and analysed at the Zeppelin observatory: CFC-11, CFC-12, CFC-113, and CFC-115. These are the main ozone depleting gases. The anthropogenic emissions started around 1930s, and all these compounds were restricted in the first Montreal protocol from 1987. The main sources of these compounds were foam blowing, aerosol propellant, temperature control (refrigerators), solvent, and electronics industry. The highest production of the CFCs was around 1985 and maximum emissions were around 1987. The lifetimes of the compounds are long (see Table 3) and combined with strong infrared absorption properties the GWPs are high.

Recent studies have shown that the rate of decline of atmospheric CFC-11 concentrations has slowed down by about 50% after 2012 (Montzka et al., 2018). This suggests new production

and unreported CFC-11 emissions, probably in Asia, strongly inconsistent with the Montreal Protocol agreement. This demonstrates the importance of maintaining good monitoring networks, both to detect possible changes related to ODSs, but also to detect possible effects of climate change on the ozone layer.

Figure 28 shows the daily averaged observed mixing ratios of the four CFCs. Before 2010/2011 the instrumentation for measurements of CFCs at Zeppelin was not in accordance with recommendations and criteria of AGAGE, and consequently there are relatively high uncertainties in the observations of these compounds, see also Appendix I. As a result, the trends are connected with relatively large uncertainties. From September 2010, new and improved instrumentation was installed at Zeppelin, providing more accurate observations of CFCs. The higher precisions are clearly visualised in Figure 28, but due to several severe instrumental problems in 2013/14 and 2015/16, there are some periods without CFC measurements¹¹.

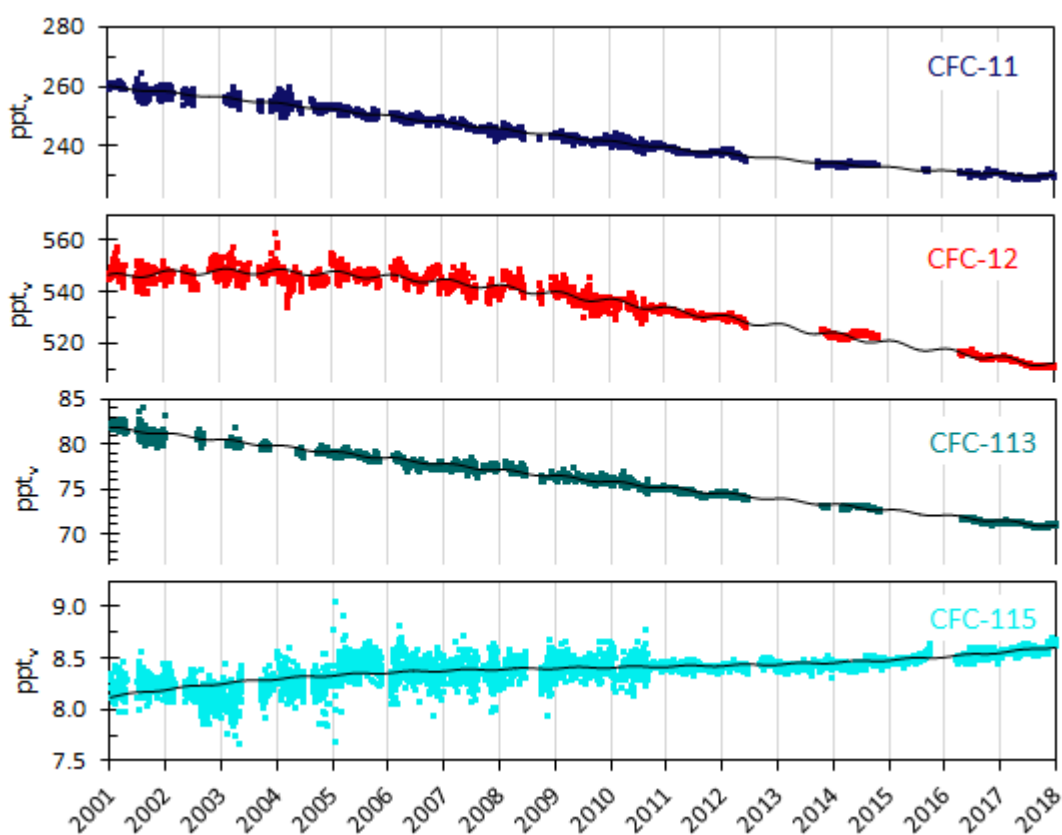


Figure 28: Daily averaged mixing ratios of the monitored CFCs at the Zeppelin observatory for the period 2001-2017: CFC-11 (dark blue), CFC-12 (red), CFC-113 (green) and CFC-115 (light blue). The solid lines are empirical fitted mixing ratios.

The trends per year for CFC-11, CFC-12 and CFC-113, given in Table 3, are all negative. The decreases are -1.9, -2.4, and -0.6 ppt/yr for CFC-11, CFC-12 and CFC-113, respectively. For the compound CFC-115 the trend is still slightly positive, +0.02 ppt/yr, and there has also

¹¹ The current instrumentation is not in accordance with recommendations and criteria of AGAGE for measurements of the CFCs and there are larger uncertainties in the observations of this compound, see also Appendix I. This is also why these compounds are particularly sensitive to instrumental problems.

been a small increase the last three years, after a stable period. It should be noted that the ambient concentration of CFC-115 is relatively low compared to CFC-11 and CFC-12, but it has a very long lifetime of 1020 years, which limits our ability to reduce the abundance in the atmosphere. In total, the development of the CFC levels at the global background site Zeppelin is promising and in accordance with the compliance of the Montreal protocol.

The 2001-2017 annual means for all the observed CFCs at Zeppelin are shown in Figure 29. Also, the global annual means of 2016 and 2017 as reported in “State of the Climate”, BAMS (Hall et al., 2017; Dlugokencky et al., 2018) are included as black bars for comparison. As can be seen, the observed concentrations at Zeppelin are close to the global mean for these compounds, as the lifetimes are long and there are hardly any present-day emissions.

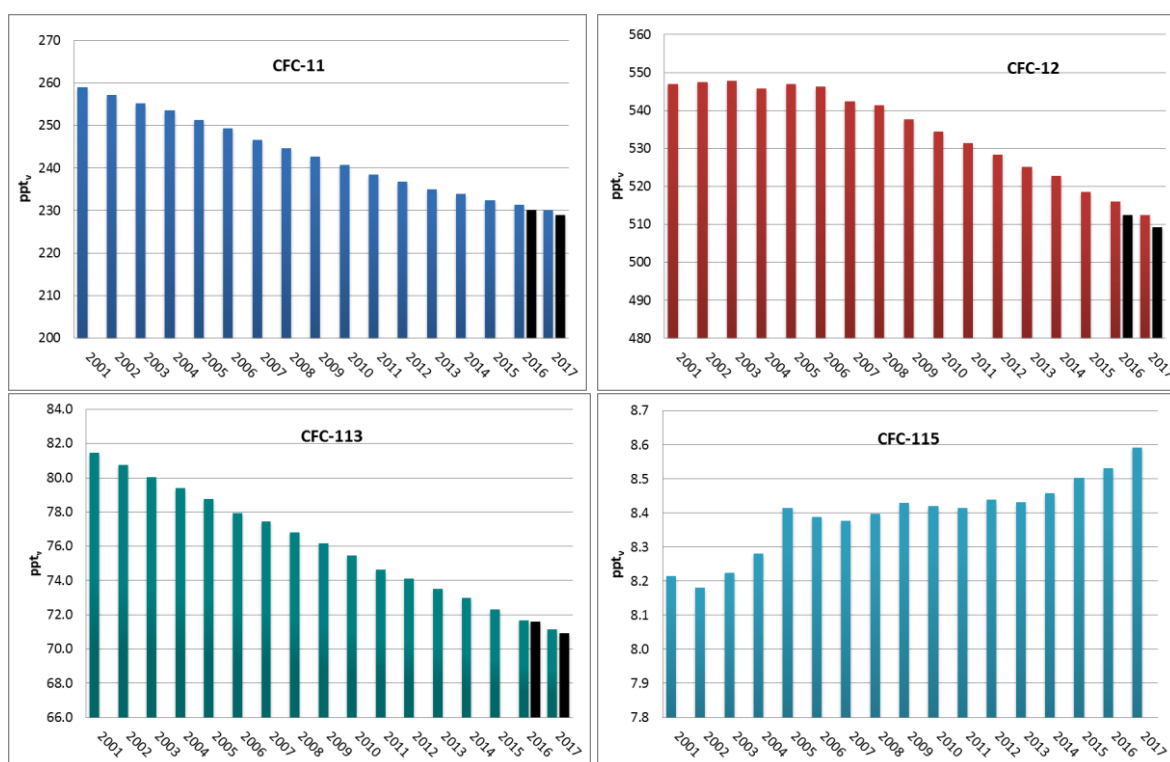


Figure 29: Development of CFC annual means at the Zeppelin Observatory for the period 2001-2017. Upper left panel: CFC-11, upper right panel: CFC-12, lower left panel: CFC-113, lower right panel: CFC-115. See Appendix I for data quality and uncertainty. The global annual means for 2016 and 2017 are included as black bars. All units are in ppt, when available.

According to WMO (2014b) the global mean mixing ratio of CFC-11 is decreasing with approximately 2.1 ppt/yr. This is in accordance with our results at Zeppelin. CFC-12 (the red diagram) has a very high GWP, 10 200, and it is also the most abundant CFC. This makes CFC-12 a very potent greenhouse gas. The global averaged atmospheric mixing ratio of CFC-12 decreased at a rate of 0.5%/yr over the period 2004-2008 (WMO, 2011). The same rate of decline was reported in WMO, 2014b. This fits well with our observations at Zeppelin. CFC-12 had a maximum around 2003-2005, but there has been a clear reduction over the last years: 35 ppt since 2005.

Key findings - CFCs: The development of the CFC levels at Zeppelin is promising and in good accordance with the compliance of the Montreal protocol. The concentrations of CFC-11,

CFC-12 and CFC-113 are all declining, and the mixing ratios of these gases are reduced with approx. 11.7%, 6.9% and 12.7% since 2001, respectively. However, recent studies have shown that the rate of decline in CFC-11 has slowed down by ~50% after 2012, probably related to unreported emissions in Asia. This is approximately parallel development at Zeppelin, and globally. CFC-115 has increased by 4.0% since 2001 and there has also been a small increase the last three years, after a stable period from 2004 to 2014.

2.2.2 Hydrochlorofluorocarbons (HCFCs) at Zeppelin Observatory

Hydrochlorofluorocarbons, HCFCs, represent the first generation of replacement gases for CFCs. The lifetimes are rather long, see Table 3, but although not as stable and persistent in the atmosphere as CFCs, they can still end up in the stratosphere where they can destroy the ozone layer. Consequently, the gases are regulated through the Montreal protocol. The Norwegian monitoring programme includes three HCFC species: HCFC-22, HCFC-141b and HCFC-142b. These compounds are mainly used as refrigerants, foam blowing agents and solvents. The use of the gases worldwide is now frozen, but they are yet not completely phased out. All the gases potentially have a strong warming effect, depending on their concentrations and absorption properties, i.e. their GWPs (see Table 3). HCFC-142b has the highest GWP, with a warming potential 1980 times stronger than CO₂, per kg gas emitted.

The daily averaged observations of the three HCFCs are shown in Figure 30 for the period 2001-2017.

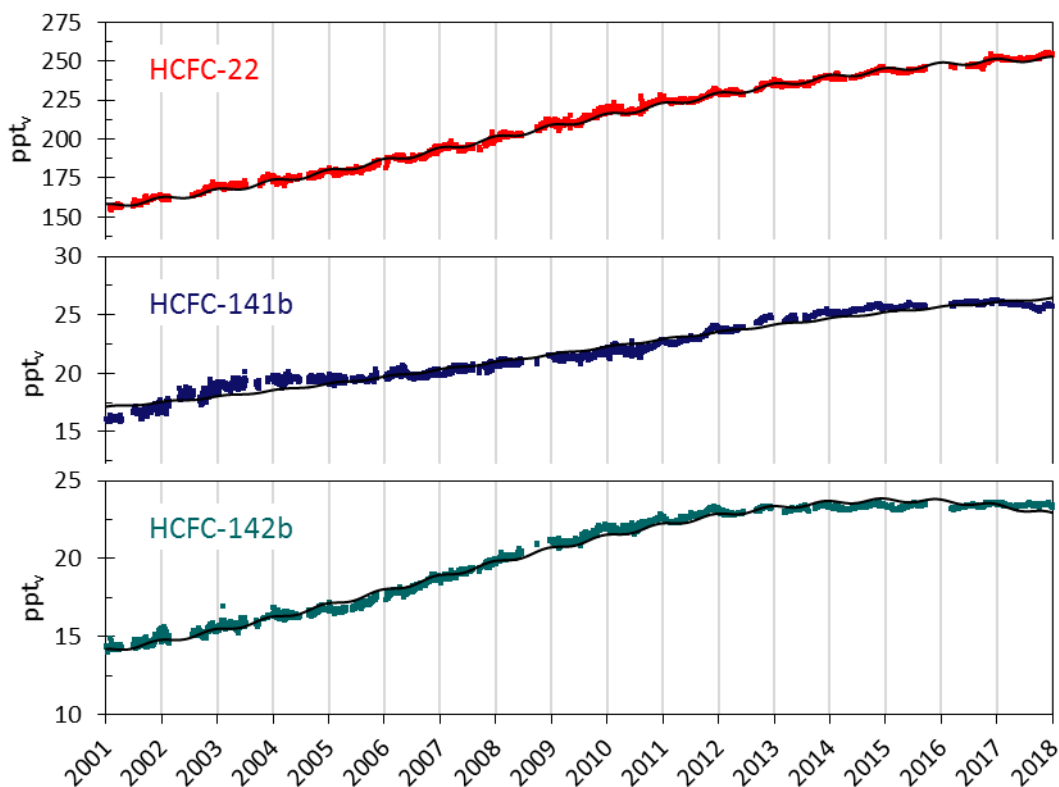


Figure 30: Daily averaged mixing ratios of the monitored HCFCs for the period 2001-2017 at the Zeppelin observatory: HCFC-22 (red), HCFC-141b (dark blue), and HCFC-142b (green). The solid lines are empirical fitted mixing ratios. All units are in ppt.

HCFC-22, HCFC-141b and HCFC-142b are all increasing over the period 2001-2017. HCFC-22 is the most abundant HCFC species and has been increasing at a rate of 6.2 ppt/yr over the period 2001-2017. The concentration of the two other compounds, HCFC-141b and HCFC-142b, are a factor of ten lower than HCFC-22, and the increase in absolute annual means are also a factor of ten lower; HCFC-141b and HCFC-142b both increase by 0.6 ppt/yr over the period 2001-2017. However, the development is positive for HCFC-141b and HCFC-142b, which both decreased slightly from 2016 to 2017. This is best illustrated in Figure 29, which shows the annual means for the full period. For all HCFCs there is a considerable increase from 2001 to 2010. With lifetimes in the order of 10-20 years, it is important to continue monitoring the development of the HCFCs for many years to come, as they have a significant influence on the ozone layer and are also strong greenhouse gases. The global annual means in 2016 and 2017 given in Hall et al. (2017) and Dlugokencky et al. (2018) are included in Figure 29 as black bars. The observed concentrations at Zeppelin are 4-6% higher than the global means.

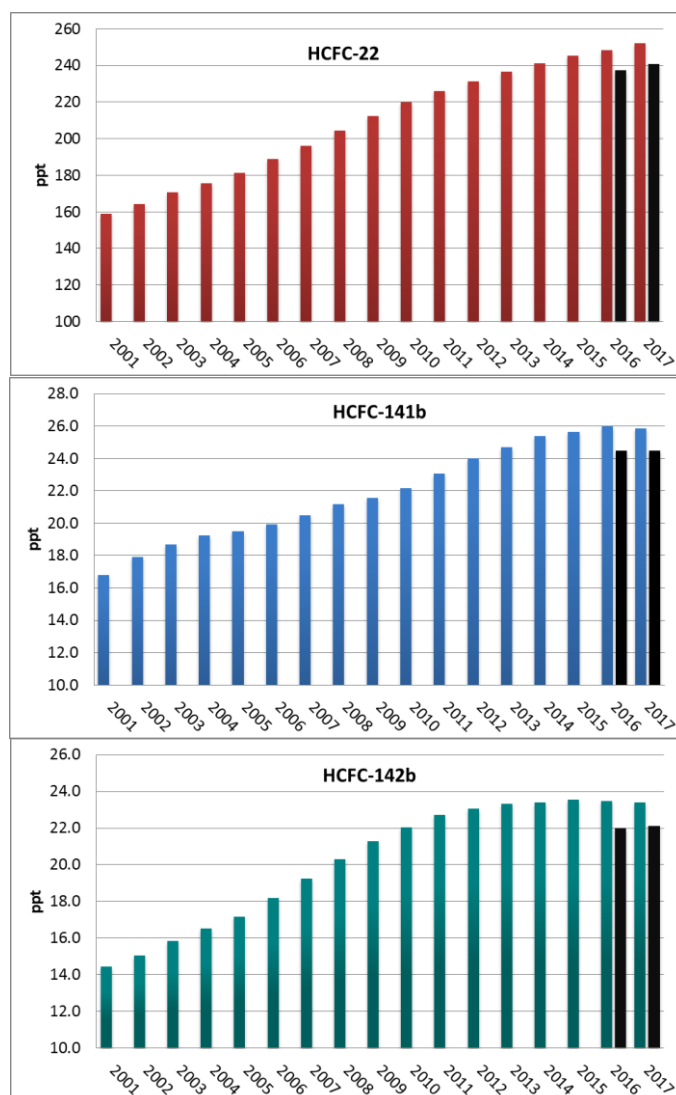


Figure 31: Development of the annual means of observed HCFCs at the Zeppelin Observatory for the period 2001-2017. HCFC-22 (red), HCFC-141b (blue), and HCFC-142b (green). The global annual means in 2016 and 2017 are included as black bars. All units are in ppt.

Key findings - HCFCs: The CFC substitutes HCFC-22, HCFC-141b and HCFC-142b have increased significantly since the measurements started at Zeppelin in 2001. HCFC-22 has a growth rate of 6.2 ppt/yr, whereas HCFC-141b has an average growth rate of 0.6 ppt/yr. From 2001 to 2017 HCFC-22, HCFC-141b and HCFC-142b increased by 58%, 54% and 62%, respectively. HCFC-22 is still increasing, but the situation is more positive for HCFC-141b and HCFC-142b, which have stabilized the last years.

2.2.3 Hydrofluorocarbons (HFCs) at Zeppelin Observatory

The substances called HFCs are the so-called second generation replacements of CFCs, which means that they are considered as better alternatives to the CFCs with respect to the ozone layer than HCFCs, as they do not contain chlorine or bromine. However, many of these compounds are strong greenhouse gases. 1 kg of HFC-23 is as much as 12 400 times more powerful greenhouse gas than CO₂ (see Table 3). The phase-down of HFCs under the Montreal Protocol has been under negotiation since 2009 and the successful agreement in Kigali, October 2016, represented an important progress (see also section 1.2). Presently, the contribution to global warming posed by HFCs is very limited. However, most of the compounds are increasing rapidly. The compounds are strong infrared absorbers with high GWP (see Table 3, page 24), hence it is crucial to reduce future emissions.

For the period 2001-2017 three compounds have been measured at the Zeppelin Observatory: HFC-125, HFC-134a, and HFC-152a. HFC-125 is mainly used as a refrigerant and fire suppression agent. HFC-134a is used as a temperature control for domestic refrigeration and automobile air conditioners, whereas HFC-152a is used as a refrigerant and propellant for aerosol sprays and in gas duster products. Since 1990, when HFC-134a was almost undetectable in the atmosphere, the concentration of this gas has risen massively, and HFC-134a is currently the HFC with highest atmospheric concentration.

In 2015 five new HFCs were included in the Norwegian monitoring programme: HFC-23, HFC-365mfc, HFC-227ea, HFC-236fa, and HFC-245fa. In 2016 three additional HFCs have been introduced to the programme: HFC-32, HFC-143a, and HFC-4310mee. All these species have been measured at Zeppelin since 2010, but they have not been analysed or reported to an international data base until 2016. The development of HFC-23 should be followed with extra care, since this gas has a relatively high concentration and an extremely high GWP. HFC-23 is a by-product of the production of HCFC-22 and is also used in the semiconductor industry, but it is also a useful refrigerant and fire suppressant.

Generally, the new HFCs are used for refrigeration and air conditioning, foam blowing, and fire extinguishing. Both HFC-245fa and HFC-365mfc are substitutes for HCFC-141b in foam blowing applications. HFC-236fa is also a foaming agent, in addition to a fire suppression agent and a refrigerant. HFC-227ea is mainly used to suppress fire in data equipment and telecommunication facilities, and in protection of flammable liquids and gases. HFC-227ea is also used as an aerosol propellant in pharmaceutical dose inhalers for e.g. asthma medication.

The three new HFCs introduced to the monitoring programme in 2016, are mainly used for refrigeration (HFC-32 and HFC-143a). In addition, HFC-143a is applied as propellant in canned air products for cleaning electronic equipment. HFC-4310mee is mainly used as a cleaning solvent in the electronics industry.

The seasonal cycle in HFC mixing ratios are closely linked to the variation in the incoming solar radiation and thus the lifetimes. HFC-152a has the shortest lifetime (1.5 year), and as seen in Figure 32 HFC-152a has the most distinct seasonal cycle. The gas is mainly destroyed in the lowest part of the atmosphere by photolysis and reactions with OH.

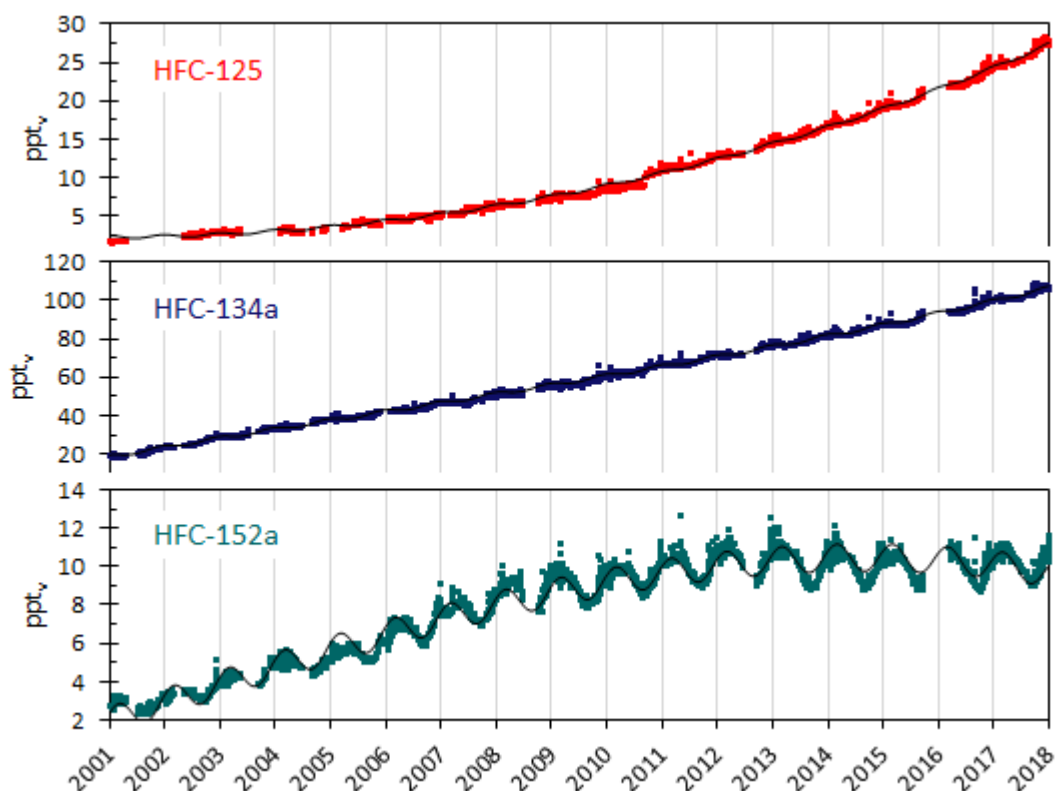


Figure 32: Daily averaged concentrations of the monitored HFCs for the period 2001-2017 at the Zeppelin observatory: HFC-125 (red), HFC-134a (dark blue), and HFC-152a (green). The solid lines are empirical fitted mixing ratios.

For the period 2001-2017 all HFCs shown in Figure 32 have increased significantly. HFC-134a has an increasing trend of 5.0 ppt/yr, which leaves this compound as the one with second highest change per year of the all the halocarbons measured at Zeppelin, next after HCFC-22. The mixing ratios of HFC-125, HFC-134a and HFC-152a have increased by as much as 1075%, 395% and 279% since 2001, respectively. For HFC-125 we can even see an accelerating trend. HFC-152a, however, is the only HFC where the rapid increase has levelled off and thereafter started to decline, due to shorter life time and rapid response to emission changes. This is clearly illustrated in Figure 32 and Figure 34.

The eight new HFCs included in the programme in 2015 and 2016 are shown in Figure 33. The time series are too short for reliable trend calculations, but the concentrations of all the

compounds have increased since 2010. The compounds have increased by 4-16%/yr, except from HFC-32 which has increased as much as 36%/yr.

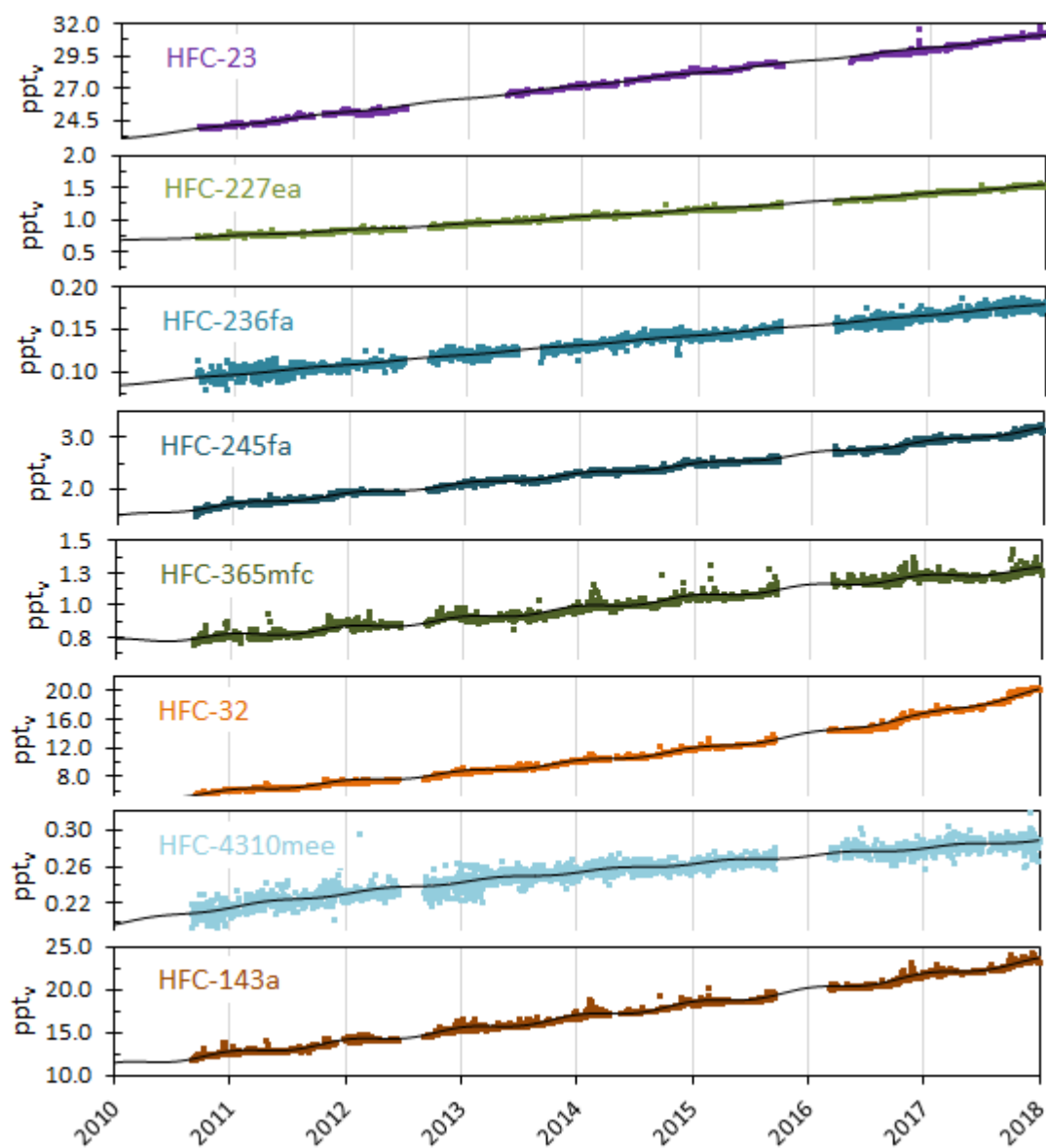


Figure 33: Daily averaged concentrations of monitored HFCs at the Zeppelin observatory for the period 2010-2017: HFC-23 (violet), HFC-227ea (light green), HFC-236fa (blue), HFC-245fa (dark blue), HFC-365mfc (dark green), HFC-32 (orange), HFC-4310mee (light blue), and HFC-143a (brown). The solid lines are empirical fitted mixing ratios.

The development of annual means of all reported HFCs are shown in Figure 34. The global annual means of 2016 and 2017 as given in Hall et al. (2017) and Dlugokencky et al. (2018) are included as black bars for comparison. As for HCFCs the concentrations at Zeppelin are slightly higher than the global means. Also, the increasing tendency for most HFCs is clear, even if the concentrations are still very low, particularly for the new HFC-365mfc, HFC-245fa, HFC-236fa, HFC-227ea, and HFC-4310mee, with 3 ppt as maximum for HFC-245fa.

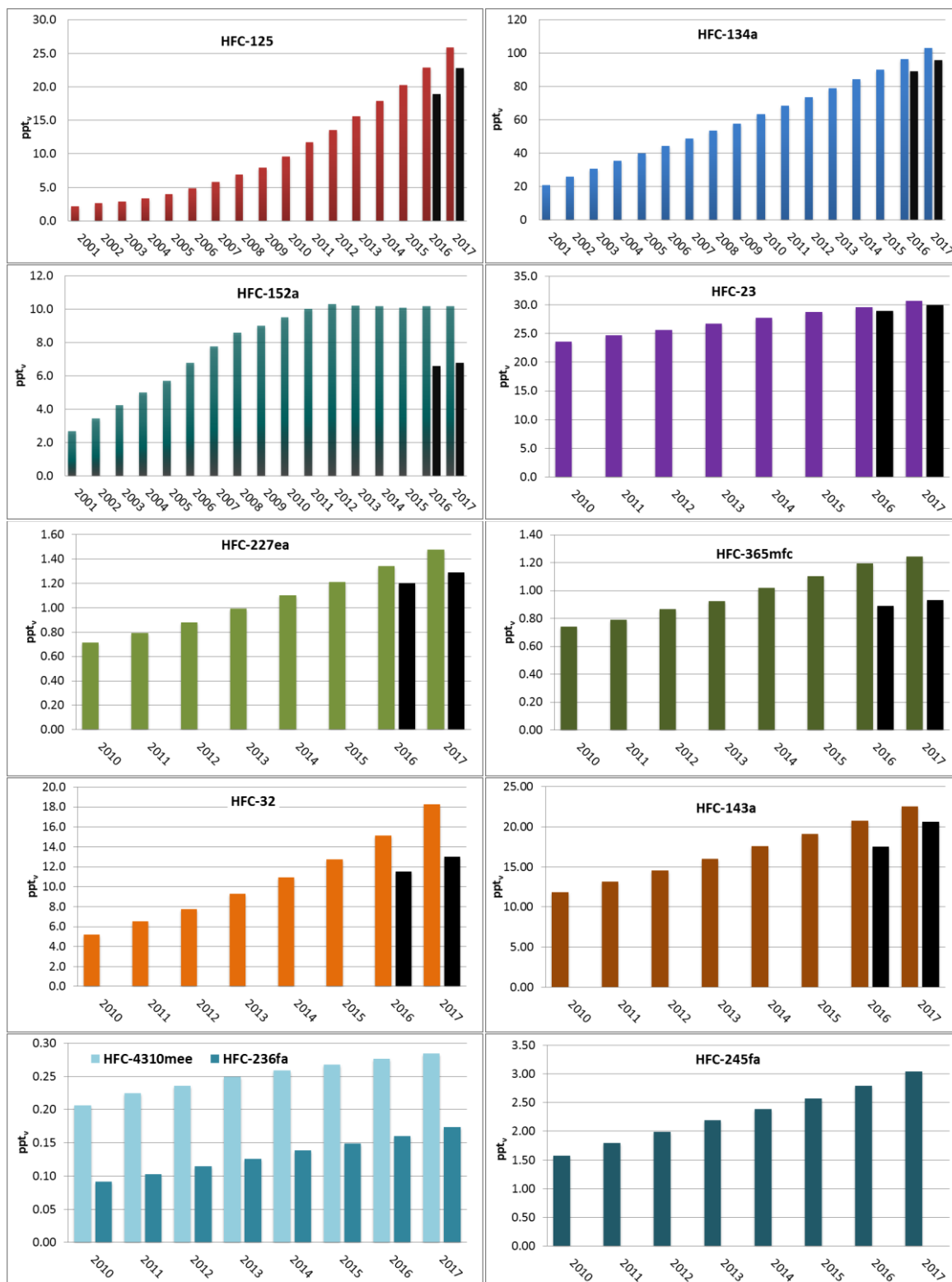


Figure 34: Development of the annual means of observed HFCs at the Zeppelin Observatory. For the period 2001-2017: HFC-125 (red), HFC-134a (blue), and HFC-152a (dark green). For the period 2010-1017: HFC-23 (violet), HFC-227ea (light green), HFC-365mfc (dark green), HFC-32 (orange), HFC-143 (brown), and light to dark blue: HFC-

4310mee, HFC-236fa, and HFC-245fa. The global annual means in 2016 and 2017 are included as black bars, when available.

Key findings - HFCs: The hydrofluorocarbons (HFCs) have been introduced as replacements for the ozone depleting CFCs and HCFCs. They pose no harm to the ozone layer since they do not contain chlorine, but still they are strong greenhouse gases. The mixing ratios of HFC-125, HFC-134a, and HFC-152a have been measured at the Zeppelin observatory since 2001, and for the period 2001-2017 the concentrations have increased by 1075%, 395% and 279% since 2001, respectively. Eight new HFCs were introduced to the monitoring programme in 2015 and 2016: HFC-23, HFC-365mfc, HFC-227ea, HFC-236fa, HFC-245fa, HFC-32, HFC-143a, and HFC-4310mee. The mixing ratios of these gases are analyzed back to 2010, showing an increase of 4-16%/yr, except for HFC-32 which increase by as much as 36%/yr. HFC-152a is the only HFC which has levelled off and started to decline,- for all the other HFCs the steep growth rate continued in 2017. The contribution from the HFCs to the global warming is still relatively small, but given the rapid atmospheric increase it is crucial to follow the development of these gases. The phase-down of HFCs under the Montreal Protocol, agreed in Kigali in 2016, is important in curbing the growth in these gases

2.2.4 Halons measured at Zeppelin Observatory

Halons are greenhouse gases containing bromine, thus regulations of halons are also important to protect the ozone layer. Actually, bromine is even more effective in destroying ozone than chlorine. The halons are regulated through the Montreal protocol and the concentration of most substances are decreasing. The main source of halons were fire extinguishers.

Previously, two halons were measured at the Zeppelin observatory: H-1301 and H-1211. In 2016 H-2402 was also included to the monitoring programme. H-2402 was used primarily in the former USSR and was the main halon fire suppressant in that region.

The ambient concentrations of the three halons are fairly low, all below 4 ppt. Figure 35 shows the daily average concentrations of the monitored halons at Zeppelin. The halon trend analyses, listed in Table 3 and visualized in Figure 35, show an increase for H-1301 during the period 2001-2017 and a relaxation for H-1211. The concentration of H-1211 is ~19% lower today than when the measurements started in 2001, whereas H-1301 has increased by ~13%. The new compound, H-2402, shows a relaxation of ~11% from 2010 to 2017. The development of the annual means for all three compounds are shown in Figure 36.

The annual means have not changed dramatically over the measured period, which is explained by low emissions and relatively long lifetimes (16 years for H-1211, 65 years for H-1301, and 20 years for H-2402). However, clear declines are evident for Halon-1211 and H-2402, which have the shortest lifetimes. According to the last Ozone Assessment report (WMO, 2014b) the total stratospheric bromine concentration decreased by $-0.6 \pm 0.1\%/yr$ between peak levels observed in 2000-2001 and 2012.

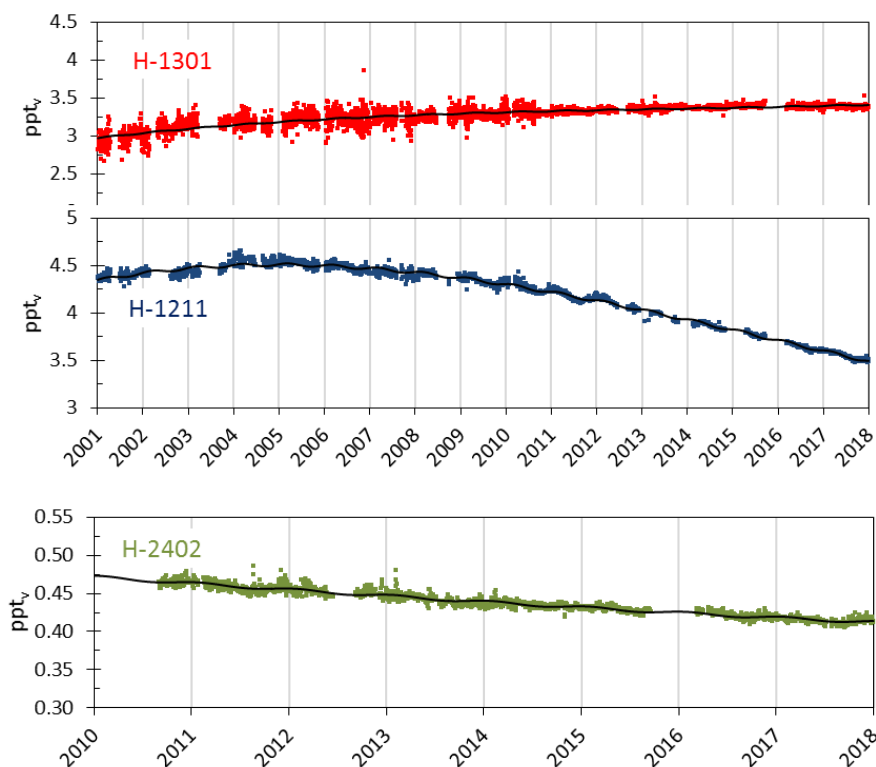


Figure 35: Daily averaged concentrations of the monitored halons at the Zeppelin Observatory. For the period 2001-2017: H-1301 (red) and H-1211 (blue). For the period 2010-2017: H-2402 (green). The solid lines are empirical fitted mixing ratios.

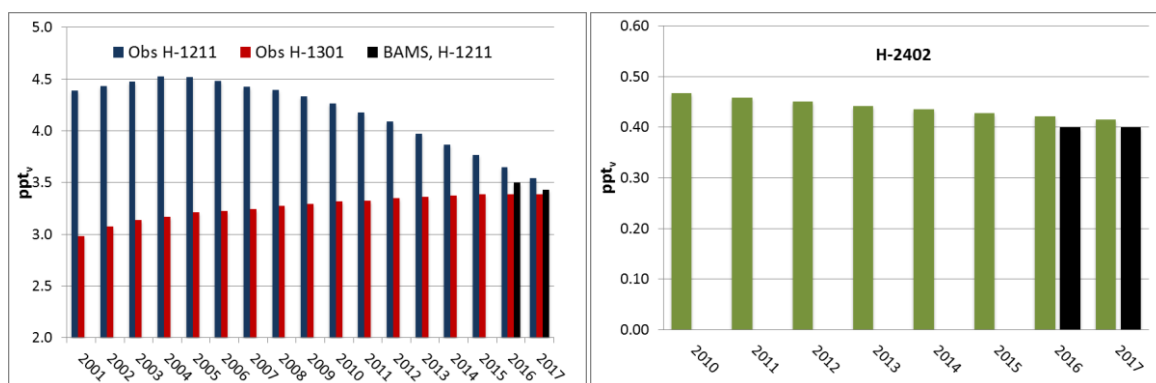


Figure 36: Development of the annual means of the observed halons at the Zeppelin Observatory. Red: H-1301, blue: H-1211, and green: H-2402. The global annual means in 2016 and 2017 are included as black bars. All units are in ppt, when available.

Key findings - Halons: Halons are bromine containing halocarbons that contribute both to the depletion of the ozone layer, and to global warming. Three halons are measured at Zeppelin: H-1211, H-1301, and H-2402. For H-1211 a maximum was observed in 2004, followed by a gradual decline. The concentration of H-1211 is ~19% lower today than when the measurements started in 2001. H-1301, however, has increased by ~13% during the same period, whereas H-2402 has decreased by ~11% from 2010 to 2017. According to the last Ozone Assessment (WMO, 2014b) the stratospheric bromine concentration has now started to decrease.

2.2.5 Other chlorinated hydrocarbons at Zeppelin Observatory

This section describes the following components measured at the Zeppelin Observatory: Trichloroethane (also called methyl chloroform, CH_3CCl_3), dichloromethane (CH_2Cl_2), trichloromethane (also called chloroform, CHCl_3), trichloroethene (TCE, CHClCCl_2), and tetrachloroethene (PCE, CCl_2CCl_2). The daily average concentrations are shown in Figure 38. Additionally, a new compound was added in 2015; Carbon tetrachloride (CCl_4). The main sources of all these substances are solvents. Trichloroethane (chloroform) has also a natural sources, with offshore seawater as the largest single source.

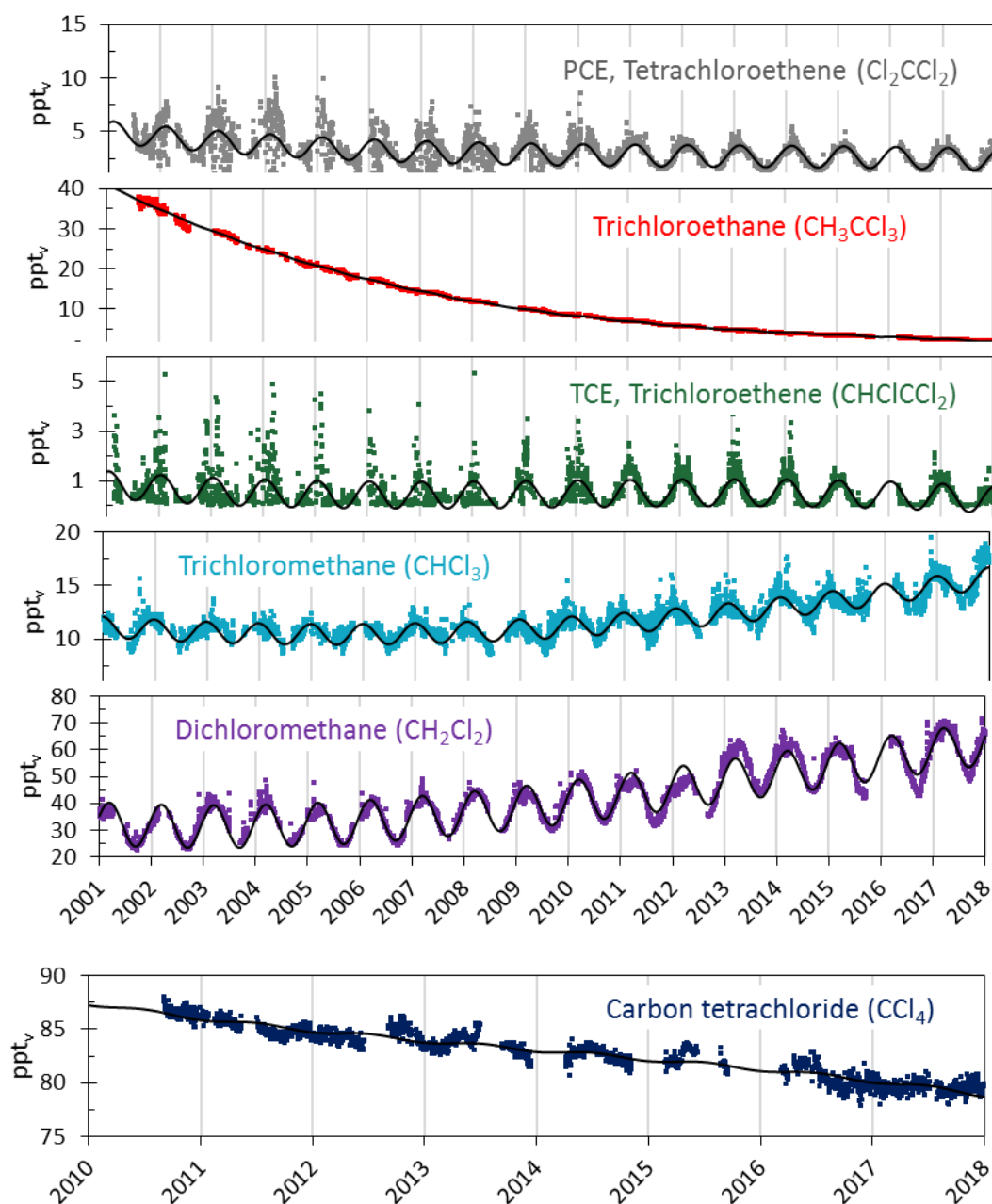


Figure 37: Daily averaged concentrations of chlorinated hydrocarbons at the Zeppelin observatory: For the period 2001-2017: Tetrachloroethene (grey), trichloroethane (red), trichloroethene (green), trichloromethane (light blue), and dichloromethane (violet). For the period 2010-2017: carbon tetrachloride (dark blue). Solid lines are empirical fitted mixing ratios.

The global fraction of trichloroethane (CH_2Cl_2), which is controlled under the Montreal Protocol, has been declining steadily since the peak values in the early 1990s. In 2012 the global mean mole fraction of 5.4 ppt was only 4% of its maximum value (WMO, 2011; 2014b). The measurements at Zeppelin show that the concentration has further decreased to 2.3 ppt in 2017, a reduction of 94% since the measurements started in 2001. Today trichloroethane contributes negligible to the atmospheric chlorine burden.

It is worth noting the strong recent increase in dichloromethane (violet), and trichloromethane (light blue) in Figure 37 and Figure 38. Dichloromethane has a lifetime of less than 6 months and responds rapidly to emissions changes, where about 90% has industrial origin. Its main applications are from paint strippers, degreasers and solvents; in foam production and blowing applications; and as an agricultural fumigant (WMO, 2011). The natural sources of dichloromethane, which account for ~10% of the total emission, is mainly from biomass burning and marine sources. At Zeppelin the increase since 2001 is about 96%. The concentration is currently 61.4 ppt, an increase of 4.6 ppb since 2016.

Even if dichloromethane has a relatively short lifetime, modelling studies imply that chlorine from this compound can reach the lower stratosphere and potentially destroy stratospheric ozone (Hossaini et al., 2017). Thus, sustained growth in dichloromethane might offset some of the gains achieved by the Montreal Protocol, further delaying recovery of Earth's ozone layer.

Trichloromethane (light blue) has a lifetime of ~1 year, thus the response to emission changes is also relatively rapid. The annual mean value of trichloromethane has increased with 46% since 2005 at Zeppelin, this is also observed at other sites (e.g. Mauna Loa at Hawaii and Barrow in Alaska). From known emissions of this compound this increase is not expected. The reason for this increase is not yet clear, and it might be explained by natural sources.

The atmospheric concentrations of trichloroethene (TCE; green) and tetrachloroethene (PCE; grey) are low, and the annual variabilities are quite high, especially before 2011. This makes it difficult to draw conclusions about trends and development of these species. In 2017 the annual average concentrations of PCE and TCE were 2.48 ppt and 0.32 ppt, respectively.

The concentration of carbon tetrachloride (CCl_4) has been measured at Zeppelin since 2010. This compound was once a popular solvent in organic chemistry, but because of its adverse health effects it is rarely used today. Today CCl_4 is sometimes used as a solvent for infrared spectroscopy. For the period 2010-2017 the annual mean values have decreased by 1.2%/yr, i.e. from 86.8 ppt in 2010 to 79.5 ppt in 2017.

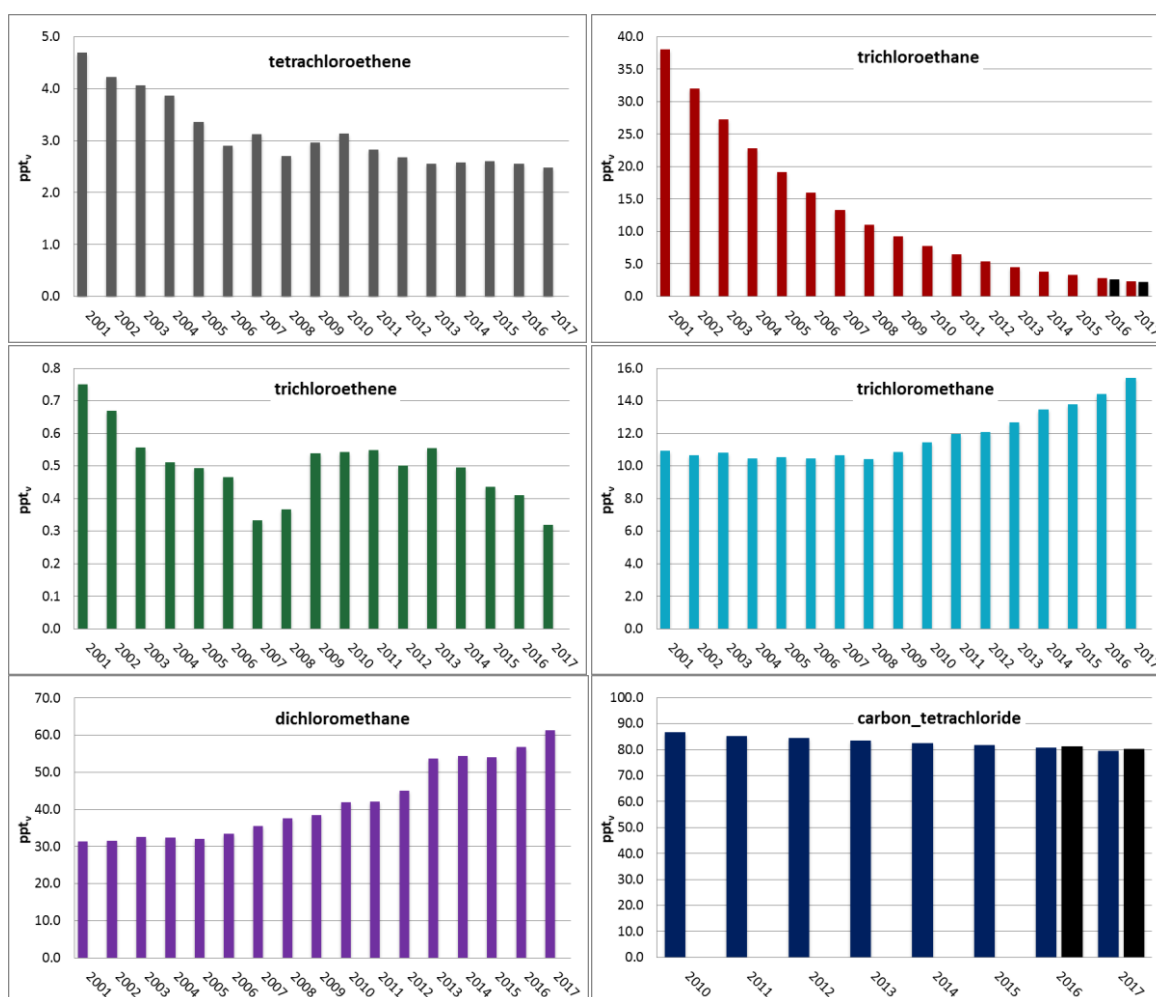


Figure 38: Annual means of the chlorinated hydrocarbons. Upper panel: tetrachloroethene (grey) and trichloroethane (red). Mid panel: trichloroethene (green) and trichloromethane (light blue). Lower panel: dichloromethane (violet) and carbon tetrachloride (dark blue). The global annual means for CH_3CCl_3 and CCl_4 in 2016 and 2017 are included as black bars, when available. All units are in ppt.

Key findings - Other Chlorinated greenhouse gases. In addition to chloromethane and bromomethane described in section 2.1.6 and 2.1.7, the following six chlorinated gases are measured at the Zeppelin Observatory: Trichloroethane (CH_3CCl_3), dichloromethane (CH_2Cl_2), trichloromethane (CHCl_3), trichloroethene (CHClCCl_2), and tetrachloroethene (CCl_2CCl_2). The concentration of most of these gases are decreasing. Exceptions are trichloromethane and dichloromethane, which have increased by 41% and 96% since 2001, respectively. The increase and development of dichloromethane needs a broader consideration, as this is a potential source of stratospheric chlorine and can delay the recovery of Earth's ozone layer.

2.2.6 Perfluorinated compounds at Zeppelin Observatory

Perfluorinated compounds belong to a group of long-lived greenhouse gases, and their contribution to the Earth's radiative forcing has increased over the past several decades. The impact of these highly fluorinated compounds on climate change is a concern because of their exceptionally long atmospheric lifetimes, as well as their strong absorption in the infrared "window" region (Baasandorj et al., 2012).

Up to 2015, the Norwegian national monitoring programme only included measurements of one perfluorinated compound, SF₆. However, other perfluorinated compounds are also very powerful greenhouse gases with atmospheric lifetimes up to 50 000 years (See Table 3) and with increasing concentrations in the atmosphere. NILU has from 2010 extended the monitoring of perfluorinated compounds at Zeppelin as we have new and improved instrumentation. Several of these compounds, so-called perfluorocarbons (PFCs), were included in the current monitoring programme from 2015, with analysis back in time to September 2010. Two additional compounds were included in the 2016 monitoring programme: Nitrogen trifluoride (NF₃) and Sulphuryl fluoride (SO₂F₂). For the latter compound data can be analysed back to 2010. The instrument at Zeppelin was re-built and optimised in 2015 and 2016, which made it possible to measure NF₃. Thus, NF₃ data are only available from 2016.

2.2.6.1 Sulphurhexafluoride (SF₆), Sulphuryl fluoride (SO₂F₂), Nitrogen trifluoride (NF₃)

Sulphurhexafluoride, SF₆, is an extremely strong greenhouse gas emitted to the atmosphere mainly from the production of magnesium and electronics industry. Measurements of this component has been a part of the programme since 2001. The atmospheric lifetime of this compound is 3 200 years, and the global warming potential is 23 500, which means that the emission of 1 kg of SF₆ has a warming potential which is 23 500 times stronger than 1 kg emitted CO₂ (Myhre et al, 2013b).

Sulphuryl fluoride, SO₂F₂, has a lifetime of 36 years and a GWP of 4 090. SO₂F₂ is normally used as a pesticide fumigant for dried fruits, nuts, and other agricultural commodities that must be kept pest-free during storage. It is one of the most common replacements for bromomethane, an ozone-depleting substance whose use is being phased out.

Nitrogen trifluoride, NF₃, has a lifetime of 500 years and a GWP as high as 16 100, meaning that it also is an extremely strong greenhouse gas. NF₃ is used in the manufacturing of new generation solar panels, flat-screen televisions, touch-sensitive screens, and electronic processors. The use of NF₃ has widely increased in the past because of the rising demand in flat-screen televisions and microelectronics.

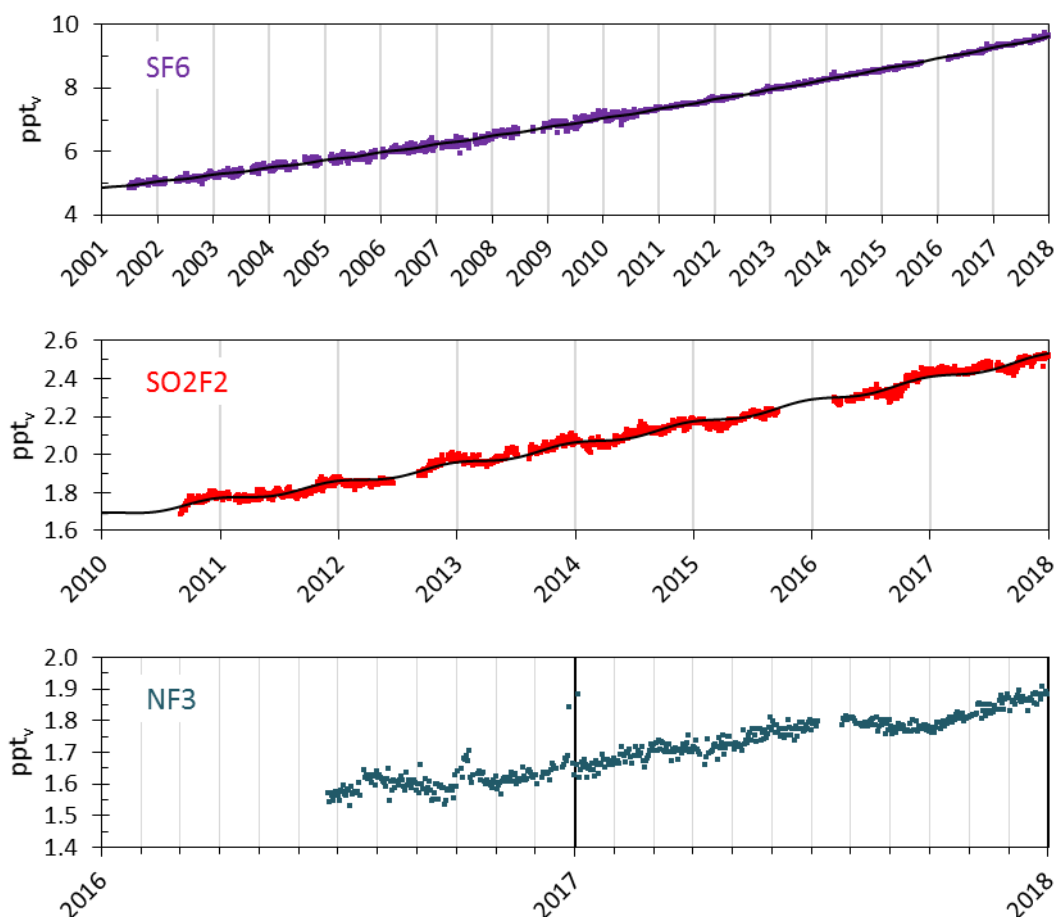


Figure 39: Daily averaged concentrations at the Zeppelin Observatory. Upper panel: SF_6 for the period 2001-2017. Middle panel: SO_2F_2 for the period 2010-2017. Lower panel: NF_3 from Jun-Dec 2017. Solid lines are statistical fitted mixing ratios.

The daily averaged concentrations of SF_6 , SO_2F_2 and NF_3 are presented in Figure 39. SF_6 is increasing with a rate of 0.28 ppt/yr and has increased as much as 91% since the start of our measurements in 2001. The instrumentation before 2010 was not optimal for measurements of SF_6 , thus there are higher uncertainties for this compound's mixing ratios than for most of the other compounds reported from 2001 to 2010 (see Appendix I).

The concentration of SO_2F_2 has also increased significantly the last years (see Figure 39). The total increase for the period 2010-2017 is 0.74 ppt (43%). No trend is derived for NF_3 , as the measurements started in 2016. Daily average observations of NF_3 are visualized in Figure 39, lower panel, and the figure clearly demonstrates a gradual increase since the measurements started in June 2016. The annual average NF_3 concentration in 2017 was 1.71 ppt.

Figure 40 shows annual average concentrations of SF_6 (left panel) and SO_2F_2 (right panel) measured at Zeppelin for the periods 2001-2017 and 2010-2017, respectively. The global annual means of SF_6 in 2016 and 2017 (Hall et al., 2017; Dlugokencky et al., 2018) are included as a black bars for comparison. Again, the concentrations at Zeppelin are slightly higher than the global means.

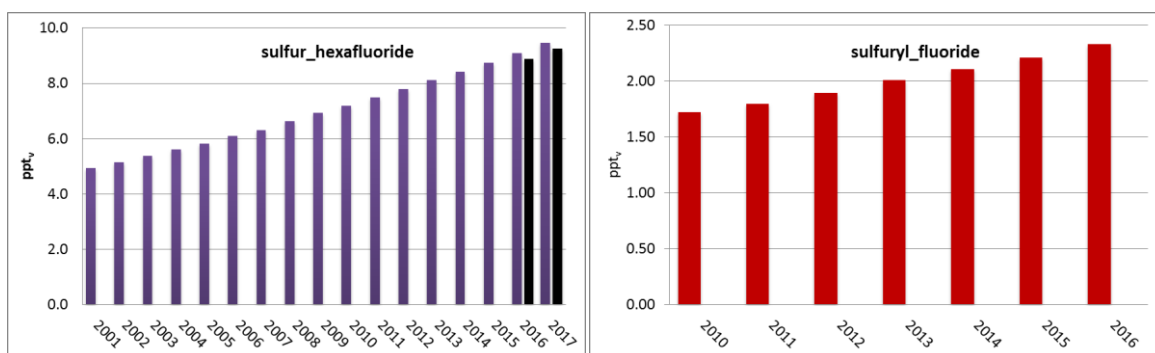


Figure 40: Annual means of SF₆ for the period 2001-2017 (left) and SO₂F₂ for the period 2010-2017 (right) at the Zeppelin observatory. The global annual mean for SF₆ in 2016 and 2017 are included as a black bar.

2.2.6.2 Perfluorocarbons or PFC's: PFC-14, PFC-116, PFC-218, PFC-318

Perfluorocarbons or PFCs are compounds that contain only carbon and fluorine. Four of these compounds are currently measured and reported at Zeppelin: PFC-14, PFC-116, PFC-218, and PFC-318. PFC-14, also called tetrafluoromethane or CF₄, is the most persistent PFC greenhouse gas with an atmospheric lifetime of 50 000 years and a greenhouse warming potential of 6 630. It is used as a low temperature refrigerant, in electronics microfabrication, and in neutron detectors. Another potent greenhouse gas is hexafluoroethane, PFC-116, which has an atmospheric lifetime of 10 000 years and a GWP of 11 100. The gas is used as an etchant in e.g. semiconductor manufacturing, and aluminium and the semiconductor manufacturing industries are the major emitters of PFC-116. Fraser et al. (2013) showed that the release of PFC-14 at Hydro's aluminium plant in Australia was 10 times greater than the release of PFC-116. Octafluoropropane, PFC-218, which has an atmospheric lifetime of 2 600 years and a GWP of 8 900, is also used in the electronics industry as a plasma etching material. In medicine, PFC-218 microbubbles reflect sound waves well and is used to improve the ultrasound signal backscatter. Octafluorocyclobutane, PFC-318, with an atmospheric life time of 3 200 years and a GWP of 9 540, is the third most abundant PFC in the atmosphere (Oram et al, 2012). Although a number of potential sources of PFC-318 have been reported, including the electronics and semi-conductor industries, there remains a large discrepancy in the atmospheric budget.

The daily averaged concentrations of the PFCs measured at Zeppelin are shown in Figure 41. For the period 2010-2017 PFC-116, PFC-218 and PFC-318 increased by 2.2%/yr, 2.6%/yr and 4.4%/yr, respectively. For PFC-14 no data exist until October 2014, but the measurements in 2015, 2016, and 2017 indicate that the concentration of PFC-14 increase by ~1%/yr.

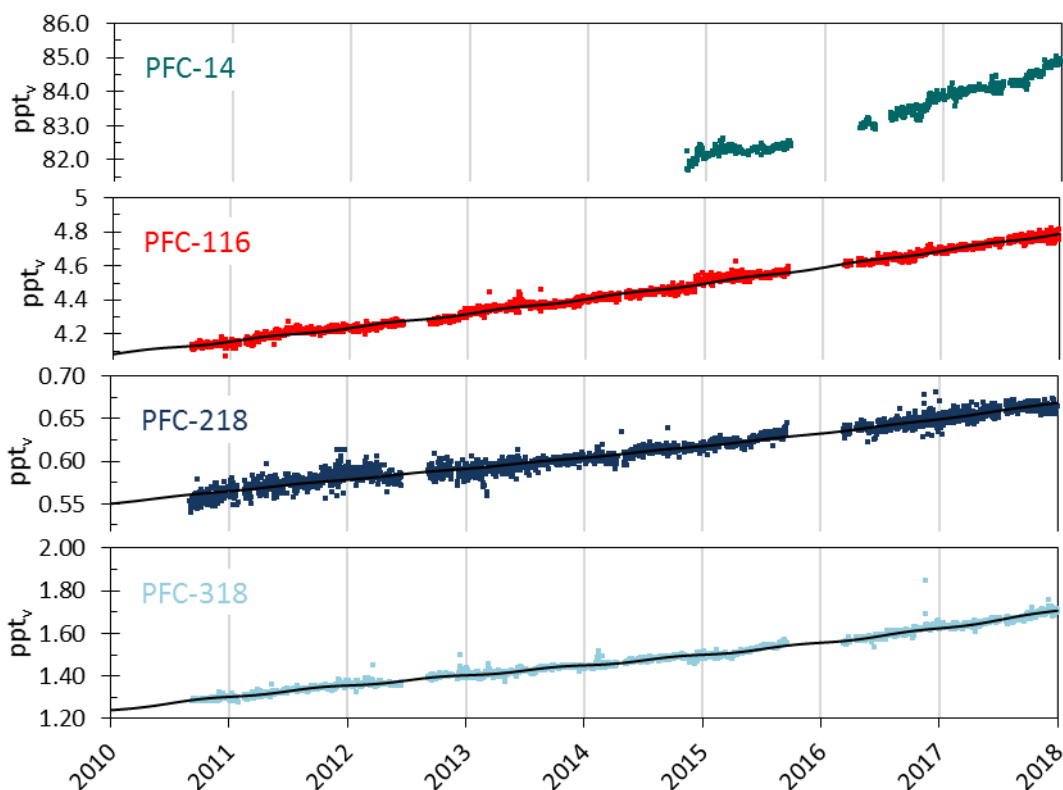


Figure 41: Daily averaged concentrations of perfluorocarbons at the Zeppelin observatory for the period 2010-2017: PFC-14 (green), PFC-116 (red), PFC-218 (dark blue), and PFC-316 (light blue). PFC-14 is only ranging back to autumn 2014. The solid lines are statistical fitted mixing ratios.

The development of the annual means of the PFCs are shown in Figure 42. For PFC-116 the global annual mean in 2016 and 2017 (Hall et al., 2017; Dlugokencky et al., 2018) are shown as a black bars for comparison. The concentrations of most PFCs are relatively low. However, PFC-14 is an exception. With an annual mean concentration of 84.3 ppt in 2017, a lifetime of 50 000 years and GWP of 6 630, this is an important greenhouse gas that should be followed carefully.

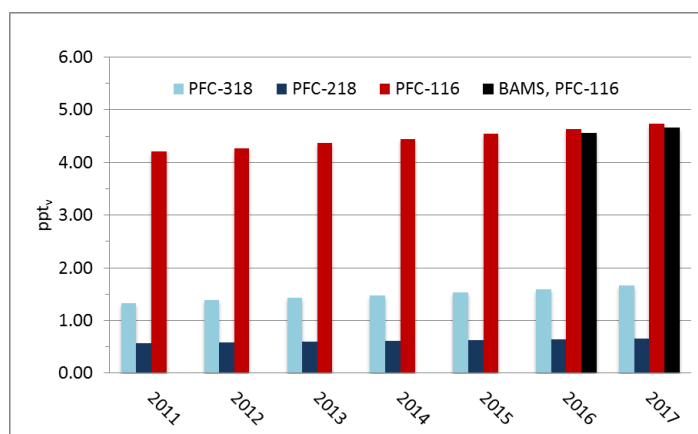


Figure 42: Annual mean concentrations of perfluorocarbons for the period 2010-2017 at the Zeppelin observatory: PFC-116 (red), PFC-218 (dark blue), and PFC-318 (light blue). The global annual mean for PFC-116 in 2016 and 2017 are included as a black bars.

Key findings - Perfluorinated compounds. This group of compounds include SF₆, SO₂F₂, NF₃, PFC-14, PFC-116, PFC-218, and PFC-318. Generally, these compounds are extremely potent greenhouse gases, but their concentrations are still low. An exception is PFC-14, which had a mixing ratio of 84.3 ppt in 2017, an increase of 1.0 ppt since 2016. SF₆ should also be followed closely, as this compound has an atmospheric life time of 3 200 years and an extremely high GWP of 23 500. This compound has increased by 91% since 2001.

2.3 Summary of measurements of climate gases

The Norwegian greenhouse gas monitoring programme is set up to meet national and international needs for greenhouse gas measurement data, both for the scientific community, national environmental authorities and global policy making. The programme includes measurements and trend calculations of 46 climate gases. The key findings from the monitoring programme shows that concentration in the atmosphere of the main greenhouse gases with high anthropogenic emissions has been increasing over the period of investigation, except for CFCs and a few halogenated gases. These gases have strong ozone depleting effect and are regulated through the successful Montreal protocol. The positive effect of this regulation on the recovery of the ozone layer is well documented. It is also a benefit for the climate.

More specific the programme shows for the main greenhouse gases CO₂, CH₄, and N₂O:

- **CO₂** concentrations have increased all years subsequently, in accordance with accumulation of gas in the atmosphere and the global development and increase in anthropogenic emissions. The new annual mean record levels in 2017 are 408.0 ppm at Zeppelin and 411.3 ppm at Birkenes. The increase from 2016 to 2017 is 3.6 ppm and 1.5 ppm, respectively.
- **CH₄**: In 2017 the mixing ratios of methane increased to a new record level both at Zeppelin and Birkenes. At Zeppelin the annual mean value reached 1939 ppb with an increase of 7 ppb since 2016. The changes at Zeppelin the last 10 years are large compared to the evolution of the methane levels in the period 1998-2005, when the change was close to zero both at Zeppelin and globally, after a strong increase during the mid 20th Century. For the isotopic signature of CH₄, a clear trend in $\delta^{13}\text{C}_{\text{CH}_4}$ is evident after 2012. The observed negative shifts in ambient $\delta^{13}\text{C}_{\text{CH}_4}$, and increases in the CH₄ ambient mixing ratio imply changes in the balance of sources and sinks. The development in $\delta^{13}\text{C}_{\text{CH}_4}$ suggest increase in the biosphere and/or agriculture (wetland or ruminant) emissions, since these do have strongly negative $\delta^{13}\text{C}_{\text{CH}_4}$ compared to ambient values and fossil sources. Another explanation is a change in the sink processes.
- **N₂O**: The global mean level of N₂O has increased from around 270 ppb prior to industrialisation and up to an average global mean of 329.8 ppb in 2017. This is slightly lower than at Zeppelin, where the annual mean was 330.0 ppb in 2017. The

difference is not significant, hence the global mean concentration and the concentration at Zeppelin are more or less the same.

The man-made greenhouse gases CFCs, HCFCs, HFCs, SF₆, NF₃, SO₂F₂, and halons hardly existed in the atmosphere before the 20th century. The gases have relatively low concentrations, but most of them have extremely long atmospheric lifetimes and high global warming potentials. In total, the gases contribute to around 12% to the overall global radiative forcing. Some of these gases are ozone depleting, especially CFCs, HCFCs and halons, and consequently regulated through the Montreal protocol.

- The development of the **CFC** levels at Zeppelin is very promising and in accordance with the compliance of the Montreal protocol. The concentrations of CFC-11, CFC-12 and CFC-113 have declined significantly since the measurements started at Zeppelin in 2001. This is a benefit both for the ozone layer and the climate. However, there has been a concern that the rate of decline in CFC-11 has slowed down after 2012, probably due to unreported CFC emissions in Asia.
- The CFC substitutes **HCFC-22**, **HCFC-141b** and **HCFC-142b** have increased significantly the last decades. From 2001 to 2017 HCFC-22, HCFC-141b and HCFC-142b increased by 58%, 54% and 62%, respectively. HCFC-22 is still increasing, but the situation is more positive for HCFC-141b and HCFC-1412b, which have stabilized the last years.
- The hydrofluorocarbons (**HFCs**) are newer replacements for CFCs and HCFCs. They are not ozone depleting, but instead they are strong greenhouse gases. Currently 11 HFCs are measured and reported at Zeppelin. All compounds have increased significantly since the measurements started, and the steep growth rate continued in 2017, except for HFC-152a. HFC-32 has the strongest increase with a trend of 36%/yr. The phase-down of HFCs under the Montreal Protocol, agreed in Kigali in 2016, is important for controlling future HFC emissions.
- **Halons** are bromine containing halocarbons and contribute both to the depletion of the ozone layer and to global warming. Three halons are measured at Zeppelin: H-1211, H-1301, and H-2402. H-1301 has increased by ~13% from 2001 to 2017, whereas H-1211 and H-2402 have declined by ~19% and ~11% during the same period, respectively.
- Six **chlorinated gases** with solely anthropogenic origin are measured at the Zeppelin Observatory: trichloroethane (CH₃CCl₃), dichloromethane (CH₂Cl₂), trichloromethane (CHCl₃), trichloroethene (CHClCCl₂), and tetrachloroethene (CCl₂CCl₂). Trichloromethane and dichloromethane have increased by 41% and 96% since 2001, respectively. The other four compounds are decreasing.
- The group of **fluorinated** compounds includes SF₆, SO₂F₂, NF₃, PFC-14, PFC-116, PFC-218, and PFC-318. Generally, these compounds are extremely potent greenhouse gases, but their concentrations are still low. An exception is PFC-14, which had a mixing ratio of 84.3 ppt in 2017, an increase of 1.0 ppt since 2016. SF₆ should also be followed closely, as this compound has an atmospheric lifetime of 3 200 years and an extremely high GWP of 23 500. This compound has increased by 91% since 2001.

Volatile Organic Compounds (VOCs) are a large group of carbon-based compounds that have a high vapor pressure and easily evaporate at room temperature. VOCs oxidation contributes to the production of tropospheric ozone and influences photochemical processing, both impacting climate and air quality. Their chemistry is essential for cleaning the atmosphere of a range of pollutants and greenhouse gases by initiating the oxidation of reduced compounds.

In addition to volatile organic compounds (VOCs) carbon monoxide (CO) is measured at the Zeppelin observatory.

- **VOCs:** Six VOCs are measured at Zeppelin: ethane, propane, butane, pentane, benzene and toluene. The annual mean concentrations vary from one year to another and for most compounds it is not possible to draw any conclusions about development and trend. Benzene and toluene are exceptions, where the annual means have decreased the last few years.
- **CO:** In general the CO concentrations measured at Zeppelin show a decrease during the period 2003 to 2009, and stable levels the last years with a small peak in 2010. CO is an excellent tracer for transport of smoke from fires (biomass burning, agricultural- or forest fires). 2017 was not particularly influenced by such fire episodes.

3 Aerosols and climate

Atmospheric aerosol influences climate by scattering incoming solar radiation back into space, or absorb the radiation. This so called direct aerosol climate forcing results mostly in cooling, but can be moderated if the aerosol itself absorbs solar radiation, e.g. if it consists partly of light absorbing carbon or light absorbing minerals. In this case, the aerosol warms the surrounding atmosphere. Atmospheric aerosol influences climate as outlined in section 1.4. Atmospheric aerosol particles also affect the reflectivity and lifetime of clouds, which is termed the indirect aerosol climate effect. The effect can be cooling as well as warming for climate, but in most cases, the cloud reflectivity and lifetime are increased, leading again to a cooling effect (see Figure 3). Figure 43 gives an overview of the main natural and anthropogenic sources of atmospheric aerosols, also described in detail the annual report of Aas et al. 2018 on long range transport of air pollution.

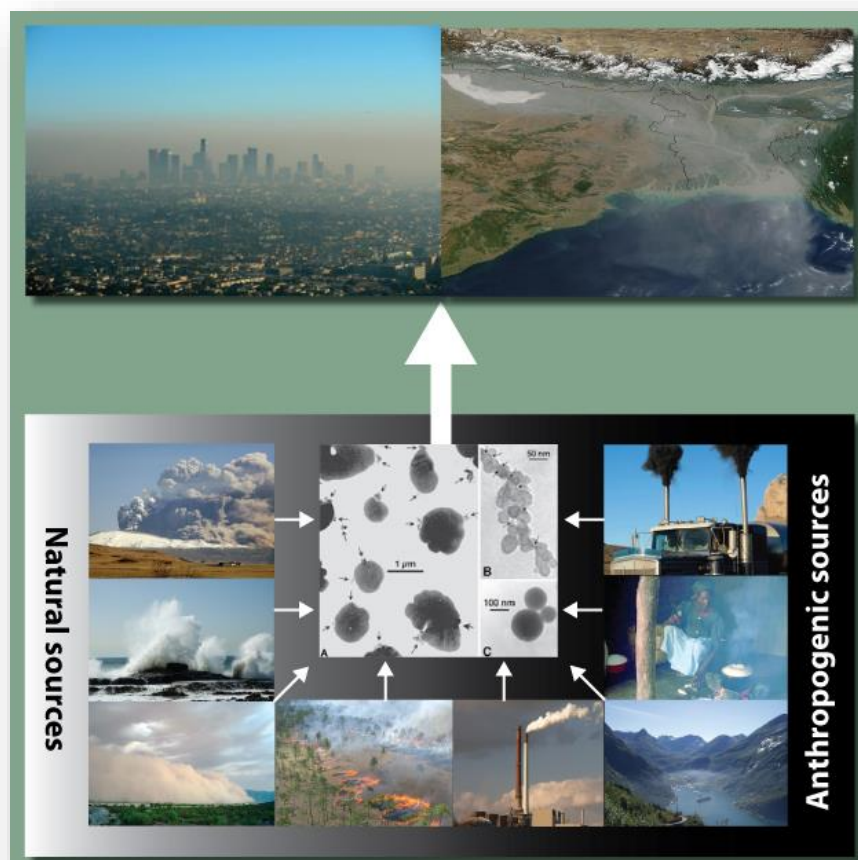


Figure 43: Illustration of the main natural and anthropogenic sources of atmospheric aerosols taken from Myhre et al (2013a). Top: local and large scale air pollution. Sources include (bottom, counter clockwise) volcanic eruptions (producing volcanic ash and sulphate), sea spray (sea salt and sulphate aerosols), desert storms (mineral dust), savannah biomass burning (BC and OC), coal power plants (fossil fuel BC and OC, sulphate, nitrate), ships (BC, OC, sulphates, nitrate), cooking* (domestic BC and OC), road transport (sulphate, BC, VOCs yielding OC). Additionally, Biogenic VOC to SOA and primary biological (e.g. pollen) from vegetation is crucial. Center: Electron microscope images of (A) sulphates, (B) soot, (C) fly ash, a product of coal combustion (Posfai et al., 1999).

Uncertainties in assessing aerosol climate forcing hamper the attribution of changes in the climate system. IPCC AR5 (IPCC, 2013) has high confidence in stating that total atmospheric aerosol negative radiative forcing in the past, has offset a significant fraction of greenhouse gas radiative warming, although the magnitude is connected with uncertainty. Due to the decline of aerosol concentrations as reported in Tørseth et al (2012), Collaud Coen et al., (2013), and Asmi et al., (2013), and summarized in Hartmann et al. 2013) the total anthropogenic radiative forcing (from both greenhouse gases and aerosols) will be even larger, i.e. more warming, in the future, due to less negative forcing from cooling aerosols (sulphates etc.)

This monitoring programme includes measurements of aerosol properties relevant for quantifying the direct and indirect aerosol climate effect from 3 observatories NILU operates: 1) Zeppelin Observatory (in collaboration with the Norwegian Polar Institute and Stockholm University); 2) Birkenes Atmospheric Observatory, Aust-Agder, Southern Norway; 3) Trollhaugen Atmospheric Observatory, Antarctica (observatory operated by NILU, main station operated by Norwegian Polar Institute). Recent developments at these stations include:

1. **Zeppelin Observatory:** Within the Swedish - Norwegian co-operation operating the atmospheric observatory on Zeppelin mountain, Stockholm University maintains a set of instruments measuring the fine-range (diameter < 1 μm) particle number size distribution, as well as the aerosol particle scattering and absorption coefficients. Through funding from the Norwegian Ministry of environment for improving the monitoring at the Zeppelin observatory, a number of additional measurements have been installed. I.e. NILU is now operating a latest generation aethalometer at Zeppelin since June 2015. The Magee AE33 instrument is a filter absorption photometer providing the spectral aerosol particle absorption coefficient at 7 wavelengths from the UV to the infra-red. The instrument is designed to be less prone to systematic uncertainties than the previous instrument generation, and complements existing observations of that type at Zeppelin. The new instrument is due to replace a previous generation instrument operated at Zeppelin by the Greek Demokritos Research Institute (Athens). Both instruments are operated in collaboration between Greece and Norway. Moreover, NILU installed a Mobility Particle Size Spectrometer (MPSS) at Zeppelin that now measures the fine-range particle number size distribution in near-real-time, with data publicly available in NILU's EBAS database within 1 hour of measurement. During autumn 2016, this MPSS was extended to also measure the particle size distribution of the fine-range non-volatile aerosol as a proxy of the absorbing aerosol fraction. The extension has been taken into operation first at the end of 2017 due to the need for extended testing.
2. **Birkenes Observatory:** In 2017, operation of the extended aerosol instrument set at Birkenes resumed in compliance with the quality standards of EMEP, WMO GAW and the European research infrastructure ACTRIS. A further improvement of the Birkenes aerosol observation programme was implemented in November 2017. The station has been equipped with a new generation filter absorption photometer measuring the particle absorption coefficient with an extended spectral range (from ultraviolet to infrared) as compared to before (from blue to red). The intention behind this upgrade is to better distinguish sources of absorbing aerosol between fossil and biomass combustion, which are supposed to have different spectral absorption signatures in

the extended wavelength range. The new instrument will also serve as a backup for the older filter absorption photometer. The redundancy will reduce the risk for gaps in the time series of the aerosol absorption coefficient since this parameter is related to emissions from wood combustion, e.g. by domestic heating.

3. **Trollhaugen Observatory:** Activities at Trollhaugen Atmospheric Observatory continued to follow up on a 2014 publication (Fiebig et al., 2014). The article investigates the annual cycle of the baseline aerosol at Troll as observed in the particle number size distribution and aerosol scattering coefficient data collected at Troll. It is shown that the baseline aerosol annual cycles in both parameters correspond. A comparison with data collected at the Antarctic stations South Pole and Dome C, yields that the baseline air annual cycle observed at Troll is common to the whole Central Antarctic plateau. Following the Troll baseline air masses backwards with the Lagrangian transport model FLEXPART, the article demonstrates that these air masses descend over Antarctica after being transported in the free troposphere and lower stratosphere from mid-latitudes (there uplifted in fronts) or from the inter tropical convergence zone (uplifted by convection). The article shows further that the aerosol particles contained in Antarctic baseline air are formed in situ by photochemical oxidation of precursor substances. A current project (AtmosCAire) following up on the discussed findings is funded by the Norwegian Antarctic Research Expeditions (NARE) programme. Among others, the project investigates the Antarctic background aerosol further by collecting an ultra-long exposure filter sample for chemical analysis despite the low concentrations, and use the cluster analysis method developed with Birkenes to identify the source regions and aerosol types found at Troll. This work is intended to further improve our knowledge about the aerosol and aerosol processes in pristine regions of the globe, which are often used as proxy for pre-industrial aerosol. Uncertainty about the climate effect of pre-industrial aerosol is still one of the main sources of uncertainty in climate predictions (Carslaw et al., 2013).

Table 4: Aerosol observations at Zeppelin, Birkenes and Troll Observatory following the ACTRIS recommendations. Parameters in green are funded by the Norwegian Environment Agency and included in this report.

	Zeppelin/Ny-Ålesund	Birkenes	Trollhaugen
Particle Number Size Distribution (fundamental to all aerosol processes)	fine mode ($0.01 \mu\text{m} < D_p < 0.8 \mu\text{m}$), NILU and in collaboration with Stockholm University	fine and coarse mode ($0.01 \mu\text{m} < D_p < 10 \mu\text{m}$)	fine mode ($0.01 \mu\text{m} < D_p < 0.8 \mu\text{m}$)
Aerosol Scattering Coefficient (addressing direct climate effect)	spectral at 450, 550, 700 nm, in collaboration with Stockholm University	spectral at 450, 550, 700 nm	spectral at 450, 550, 700 nm
Aerosol Absorption Coefficient (addressing direct climate effect)	single wavelength at 525 nm, (Stockholm University); single wavelength at 670 nm (Stockholm University); 7-wavelength (Demokritos Athens); 7-wavelength (NILU)	spectral at 470, 522, 660 nm wavelength.	spectral at 470, 522, 660 nm wavelength.
Aerosol Optical Depth (addressing direct climate effect)	spectral at 368, 412, 500, 862 nm in collaboration with WORCC	spectral at 340, 380, 440, 500, 675, 870, 1020, 1640 nm, in collaboration with Univ. Valladolid	spectral at 368, 412, 500, 862 nm
Aerosol Chemical Composition (addressing direct + indirect climate effect)	inorganic ions (ion chromatography), heavy metals (inductively-coupled-plasma mass-spectrometry)	main components (daily resolution, offline filter-based, ion chromatography), heavy metals (inductively-coupled-plasma mass-spectrometry)	inorganic ions and POPs (ion chromatography), discontinued from 2011 due to local contamination.
Aerosol Chemical Speciation (direct + indirect climate effect, source attribution, transport)	Particle main chemical species (hourly resolution, online mass spectrometry)	Particle main chemical species (hourly resolution, online mass spectrometry)	---
Particle Mass Concentration	---	PM _{2.5} , PM ₁₀	PM ₁₀
Cloud Condensation Nuclei (addressing indirect climate effect)	size integrated number concentration at variable supersaturation in collaboration with Korean Polar Research Institute	Size integrated number concentration at variable supersaturation	---

An overview of all aerosol parameters currently measured at the 3 observatories can be found in Table 4. Parameters included in this report, are written in green.

3.1 Observed optical properties of aerosols

Aerosol absorption and scattering are decisive for the cooling or warming effect of aerosol in climate. All types of aerosols scatter solar radiation, but the higher fraction of aerosol absorption, the more warming are the aerosols. The absorption is depending on the composition; black carbon (e.g. soot) and minerals absorb radiation.

3.1.1 Optical aerosol properties measured at the Birkenes Observatory

The comprehensive set of instruments observing in situ optical aerosol properties at Birkenes, i.e. those describing the direct effect of aerosol on climate, has now been in operation since 2010. The parameters covered include the scattering coefficient σ_{sp} and the absorption coefficient σ_{ap} at various wavelengths. Figure 44 summarises the essence of these observations for the years 2010 - 2017 in time series of the observations themselves and relevant directly derived parameters. All properties are measured for particles with aerodynamic diameter $D_{p,aero} < 10 \mu\text{m}$ and at relative humidity below 40%, thus avoiding water uptake by particles. This protocol follows the recommendations provided by of the European research infrastructure ACTRIS, also harmonised with the recommendations of the WMO GAW aerosol network. This is crucial to have comparable data at Zeppelin, Trollhaugen and Birkenes, and with other site on European and global scale. For more details concerning measurement principles and quality assurance routines, please see Appendix II.

Panel a) of Figure 44 displays the time series of the spectral scattering coefficient σ_{sp} at 450, 550, and 700 nm wavelength. Thin lines represent daily average values for the respective wavelength, whereas the heavy green line represents the running 8-week median for easier visibility of seasonal averages (green wavelength at 550 nm only for clarity). The σ_{sp} time series exhibits significant variability on the time scale of days, illustrating that particle load in an air mass varies significantly with air mass type and thus air mass origin, i.e. with the synoptic weather situation on a time scale of 1 - 3 days. When focussing on the graph of the running median, a slight seasonal variation can be detected, with values higher in summer than winter.

For optical aerosol properties, information is contained both in the absolute level and in the values at different wavelengths relative to each other. In order to make this spectral information accessible, the Ångström coefficient has been defined, that can be calculated for optical aerosol properties. Higher values of the Ångström coefficient correlate with higher concentration ratios of particles in the fine size range ($D_p < 1 \mu\text{m}$) as compared to the coarse size range ($D_p > 1 \mu\text{m}$). Moreover, the relative size of particles determining an optical aerosol property decreases with wavelength, i.e. smaller wavelengths “look at” smaller particles in relative terms, larger wavelengths “look at” larger particles. Already with these simple qualitative rules, many features exhibited by spectral aerosol optical property data in general and Ångström coefficient data in particular can be interpreted meaningfully.

Panel b) of Figure 44 shows the time series of the Ångström coefficient \hat{a}_{sp} , calculated from $\sigma_{sp}(\lambda)$, again as daily averages (thin line) and running 8-week median (heavy line). As with σ_{sp} , the strongest variability is associated to a time scale of 1-3 days, indicative of changes associated with air mass type, origin, and synoptic weather situation. Looking at the running median however, the seasonal cycle is more pronounced for \hat{a}_{sp} as for σ_{sp} , with \hat{a}_{sp} values around 1.1 in winter and 1.8 in summer. This indicates a stronger contribution of smaller particles, i.e. particles with D_p smaller than about 120 nm, to σ_{sp} in summer than in winter.

This is consistent with number concentrations of particles in this size range exhibiting a similar seasonal cycle, as will be discussed below. The time series of σ_{sp} and \hat{a}_{sp} don't show any trend, which is consistent with the findings for other European continental background stations at Jungfraujoch (Switzerland, mountain top), Hohenpeissenberg (Southern Germany, elevated boundary layer), and Pallas (Northern Finland, boreal background) (Collaud Coen et al., 2013). Also the range of σ_{sp} values encountered, 3 - 50 Mm^{-1} with an annual average of about 12.5 Mm^{-1} , is consistent with findings at comparable stations (Delene & Ogren, 2002).

The observations of the particle absorption coefficient σ_{ap} at Birkenes were upgraded in 2012 and in November 2017. All 3 concerned instruments use the same physical principle, tracking the change of attenuation of light that passes through a filter while the filter is loaded with sampled particles. The 2012 instrument measures σ_{ap} at 3 wavelengths (470, 522, 660 nm) with considerable less electronic noise and significantly better long-term stability than the original instrument from 2010 with one wavelength (522 nm). The 2017 instrument extends the spectral range to 7 wavelengths (370, 470, 525, 590, 660, 880, 950 nm). Panel c) of Figure 44 displays the σ_{ap} time series of the 2012, 3-wavelength filter absorption photometer, Panel d) the data of the 2010 one wavelength instrument. For the 2010 instrument, the wavelength has been recalculated to the same green wavelength as observed by the nephelometer (550 nm) assuming an Ångström coefficient of 1. Both panels use thin lines for daily averages, heavy lines for the running 8-week medians.

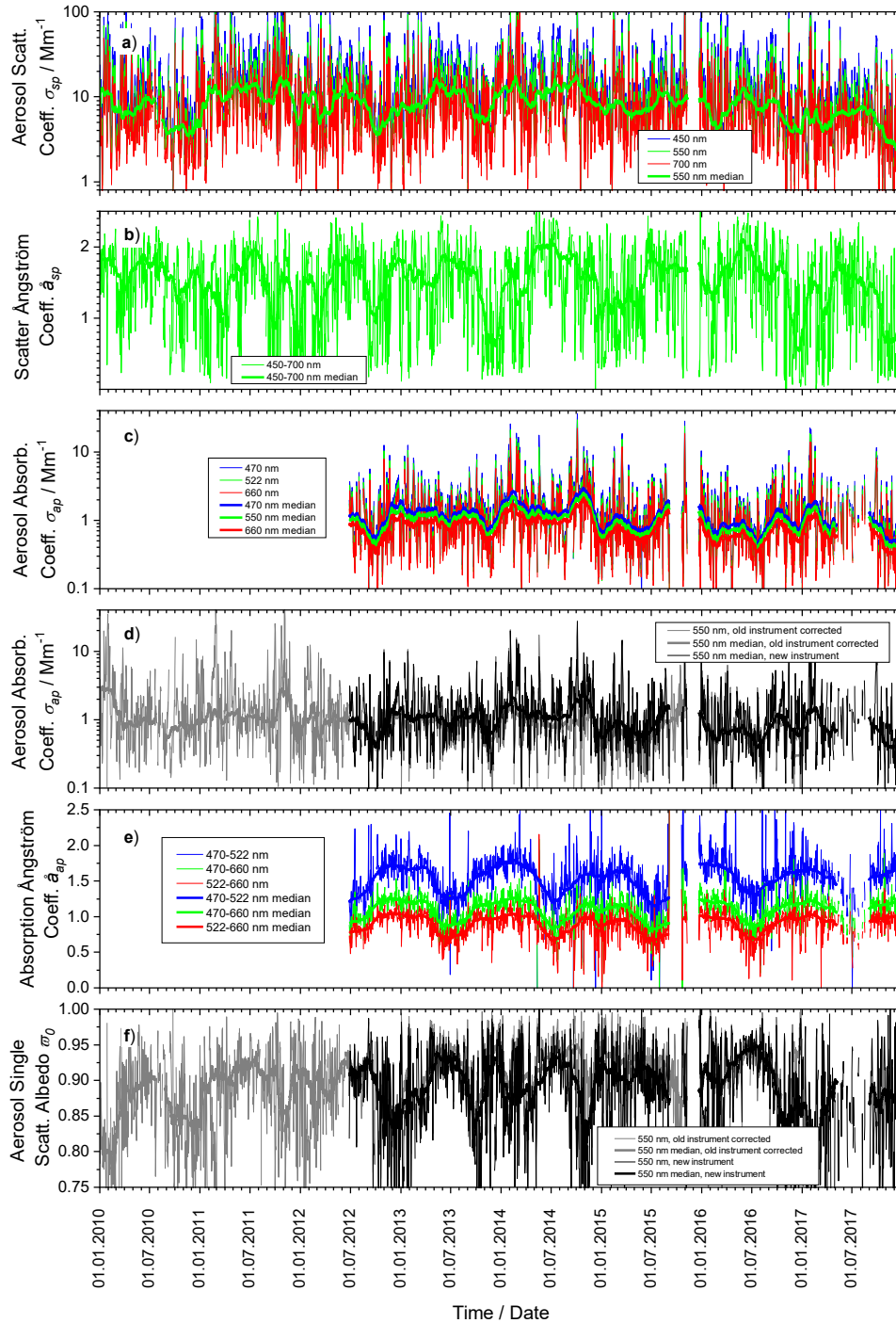


Figure 44: Time series of aerosol particle optical property daily means measured for 2010 - 2017 at Birkenes. Panel a) shows the aerosol scattering coefficient σ_{sp} at 450, 550, and 700 nm wavelength measured by integrating nephelometer. Panel c) the aerosol absorption coefficient σ_{ap} at 470, 522, and 660 nm wavelength measured by the newer filter absorption photometer, Panel d) the aerosol absorption coefficient σ_{ap} at 550 nm of the older filter absorption photometer, shifted from the instrument wavelength at 525 nm for consistent comparison assuming an absorption Ångström coefficient of -1. Panels b) and e) show the derived properties scattering and absorption Ångström coefficient \hat{a}_{sp} and \hat{a}_{ap} , respectively, while Panel f) depicts the single scattering albedo ω_0 . All plots also depict the running 8-week medians of the respective properties as heavy lines to visualize seasonal variations. To demonstrate consistency between old and new filter absorption photometer, respective comparison lines are displayed.

Panel d) also displays the σ_{ap} running median time series of the 2012 filter absorption photometer (also recalculated to 550 nm wavelength) together with the data of the old instrument as an indicator for the goodness of overlap between the 2 instruments measuring the same property. Even though the older filter absorption photometer underwent quality assurance by off-site intercomparison within ACTRIS in 2013, a drift of the reading of the older compared to the newer instrument can be observed between 2013 - 2015. These stability issues of the older filter absorption photometer were discovered already during the 2013 intercomparison. Consequently, also the newer filter absorption photometer was subjected to an ACTRIS intercomparison in 2015. The intercomparison discovered no issues with the newer instrument and confirmed stability of calibration. As a result, the whole σ_{ap} time series obtained with the old filter absorption photometer has been corrected with the 2012 instrument as reference, yielding a consistent aerosol absorption time series for Birkenes for the years 2010 - 2017. In 2017, the 2012 3-wavelength absorption photometer experienced problems related to an unstable measurement of the sample flow, leading to low data coverage in summer 2017. Due to the importance of the aerosol absorption measurement in relation to emissions from biomass combustion for domestic heating, it was decided already in 2016 to install the new 7-wavelength version in parallel, and keep both instruments running as mutual backup. However, the new instrument couldn't be installed before late 2017. Luckily, the heating season, for which the aerosol absorption measurements are most critical, is covered also in 2017.

Apart from the variation with synoptic transport and air mass origin, σ_{ap} does not seem to exhibit significant seasonal variation. The values of σ_{ap} are in the range of 0.3 - 4 Mm^{-1} with annual means around 1 Mm^{-1} .

In order to cover the largest possible time period, panel f) of Figure 44 shows the time series of the aerosol particle single scattering albedo ϖ_0 based on the time series of the older filter absorption photometer (grey lines), again daily averages (thin line) and 8-week running median (heavy line), together with the respective time series of the new instrument (black lines). The single scattering albedo ϖ_0 quantifies the fraction of light scattered by the particles rather than absorbed. It thus quantifies how absorbing the average aerosol particle is, with ϖ_0 values decreasing with increasing absorption of the average particle. For a purely scattering aerosol, ϖ_0 is 1, and decreases with increasing fraction of light absorbing components in the aerosol particle phase. The most prominent feature in the ϖ_0 time series is the pronounced annual cycle, varying between 0.86 - 0.96, with lower ϖ_0 values and higher particle absorption in winter. The annual cycle in ϖ_0 has been discussed in previous reports of the Birkenes aerosol dataset (Myhre et al, 2016), and has been connected to the combustion of biomass in wood stoves for domestic heating in the winter season.

Previous reports mentioned an increasing tendency in ϖ_0 (decreasing tendency in average particle absorption) until 2014 at Birkenes, with levelling out thereafter. A tendency towards less aerosol absorption has also been observed for stations in the continental U.S. and Alaska (Barrow), but only few stations in continental Europe (Hohenpeissenberg, Germany) (Collaud et al., 2013; Coen et al., 2013). After re-evaluating and drift correcting the Birkenes aerosol absorption coefficient time series following the ACTRIS inter-comparison in 2015, this tendency of increasing ϖ_0 cannot be confirmed. Both the time series and tabulated seasonal and annual averages of ϖ_0 (section 3.3) show rather stable values from year to year for corresponding seasons. This finding underlines the importance of careful quality management

for long-term time series of climate relevant parameters. Even small trends of a few percent in these parameters usually have impact on climate, requiring a constant high accuracy in the measurement of the same order. Connection to frameworks like ACTRIS and GAW and associated traceability is key to achieving this accuracy.

Panel e) of Figure 44 is based on data of the new filter absorption photometer, measuring σ_{ap} at 3 wavelengths, as opposed to 1 wavelength with the older instrument. Panel e) depicts the time series of the absorption Ångström coefficient \hat{a}_{ap} for all 3 wavelength pairs provided by the new instrument. For \hat{a}_{ap} , the information on relative particle size concerns not the overall aerosol particle phase, but the fraction of absorbing particles. Thus, considering \hat{a}_{ap} data allows to investigate changes in source and transport of the absorbing particle fraction in the aerosol. When looking at the \hat{a}_{ap} data in panel e) of Figure 44, an annual cycle is apparent that is opposite of the annual cycles seen in scattering Ångström coefficient \hat{a}_{sp} and single scattering albedo ω_0 . Both \hat{a}_{sp} and ω_0 increase in summer as compared to winter, \hat{a}_{sp} because of a summer increase in small particles due to particle formation from biogenic precursors, ω_0 because of fewer combustion emissions in summer than in winter. For \hat{a}_{ap} , values increase in winter as compared to summer, indicating, in relative terms, higher abundance of smaller absorbing particles in winter than in summer. This observation is consistent with assuming emissions from domestic heating by wood stoves to contribute to the Birkenes winter aerosol, which is the explanation for the decreased winter values of ω_0 . The size of the absorbing aerosol particles increases with aerosol age. Consequently, a smaller size of the absorbing particles indicates a younger combustion aerosol and a closer combustion source, which is consistent with a scattered distribution of houses using wood stoves for heating, i.e. typical for Southern Norway. The previous instrument upgrade of aerosol absorption measurements in Birkenes has thus provided another indication for the contribution of wood stove emissions to the Birkenes winter aerosol. Further information can be expected from the additional upgrade extending the spectral range of particle absorption observations in November 2017, which will give results for 2018.

Both σ_{sp} and σ_{ap} show a drop towards the end of 2017. This drop is consistent with the observations of other aerosol properties at Birkenes, and is likely related to an early start of the ground snow cover in winter 2017/18. Despite low coverage in aerosol absorption observations in summer 2017, there doesn't seem to be a significant change in the relative light absorption capacity of the aerosol particle phase during the heating season for 2017.

Key findings aerosol optical properties Birkenes: The higher aerosol absorption, the more warming is the aerosol. The tendency from earlier years towards a decreasing trend in absorption of the average aerosol particle at Birkenes is not confirmed. Careful quality management revealed that this slight tendency observed in earlier years was due to calibration drift of the associated instrument. Trends in aerosol optical properties cannot be observed at Birkenes, which corresponds to findings at other European stations. The aerosol scattering shows a stronger contribution of smaller particles in summer than in winter. The reason for these seasonal differences is that summer aerosol is influenced by biogenic emissions which tends to give larger amounts of newly formed small particles.

3.1.2 Optical aerosol properties measured the Zeppelin Observatory

The in situ optical properties of the particles at Zeppelin Observatory covered in this report are more limited as compared to Birkenes and Trollhaugen. NILU operates only a filter

absorption photometer, an AE33 aethalometer, at Zeppelin measuring the absorption coefficient $\sigma_{ap}(\lambda)$ between 370-950 nm wavelength. A nephelometer providing the spectral scattering coefficient $\sigma_{sp}(\lambda)$ is also deployed at Zeppelin, but operated by Stockholm University, and data is therefore not available for this report. This excludes also calculation and discussion of the particle single scattering albedo.

Figure 45 summarises the 2015 - 2017 time series, i.e. 2.5 years, of spectral aerosol particle absorption coefficient $\sigma_{ap}(\lambda)$ data collected at Zeppelin Observatory since deployment of the aethalometer in June 2015. The top panel displays daily averaged $\sigma_{ap}(\lambda)$ data for all 7 wavelengths (thin lines), as well as running 8-week medians (bold lines), here only for 3 wavelengths to improve readability of the graph. The middle panel shows graphs of the absorption Ångström coefficient \hat{a}_{ap} for 3 wavelength pairs representing short wave end, long wave end, and full range of the measured wavelength spectrum. The \hat{a}_{ap} data are plotted as 8-week median only since daily averages are too noisy due to the low particle load commonly observed at Zeppelin.

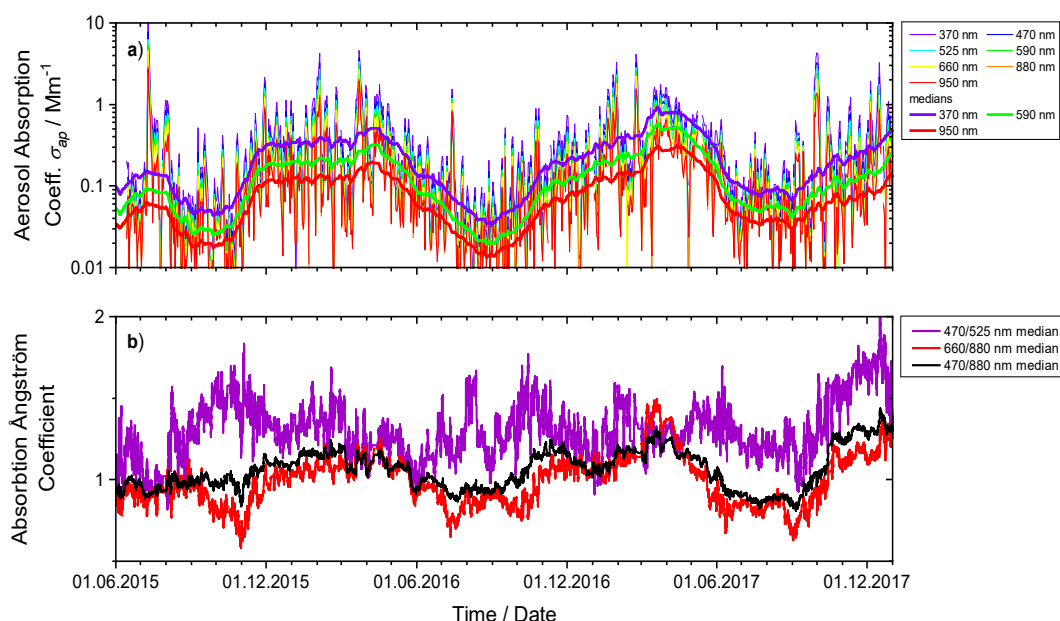


Figure 45: 2015 - 2017 time series graphs of data collected by the filter absorption photometer at Zeppelin Observatory since deployment in June 2015. Top: daily means of absorption coefficient at all 7 measured wavelengths (thin lines), and 8-week running medians for top, middle and bottom of observed spectral range (heavy lines). Bottom: 8-month running medians of absorption Ångström coefficient, top, bottom, and whole observed spectral range.

Even though the time series of the particle absorption data at Zeppelin spans only 2.5 years at this point, a few interesting features can be detected. The time series of σ_{ap} (top panel of Figure 45) exhibits the same annual cycle as described already in the data of the other aethalometer instrument at Zeppelin collected in earlier years (Eleftheriadis et al., 2009), with lower values in summer and higher ones in winter. These variations have been attributed to changes in combustion aerosol sources caused by emissions from domestic heating in the relevant source regions, e.g. Northern and Central Russia (Law & Stohl, 2007). This pattern is moderated by incidents of emissions from large forest fires reaching the Arctic, which can

increase aerosol particle absorption in episodes also in summer (e.g. Stohl et al., 2007). Absolute values of σ_{ap} vary in the range of 0.02 - 2 Mm⁻¹ at Arctic Zeppelin station, with median values roughly a factor of 5 lower than at boreal Birkenes, but a factor of 4 larger than at Antarctic Trollhaugen station.

Also interesting in the data provided by the new filter absorption photometer is the aerosol absorption Ångström coefficient \hat{a}_{ap} . The \hat{a}_{ap} values exhibit a distinct spectral dependence on wavelength, with \hat{a}_{ap} for shorter wavelengths deviating to higher values as compared to \hat{a}_{ap} at larger wavelengths for periods of lower aerosol absorption, i.e. in summer. Such a behaviour can be caused by absorbing aerosol components that exhibit a spectral dependence in absorption, e.g. brown carbon or dust. For black carbon in contrast, \hat{a}_{ap} at shorter and larger wavelength agree at values around 1. An explanation of these phenomena needs to await further measurements at Zeppelin. Also funded by the grant of the Norwegian Ministry of Climate and Environment, an instrument observing the size distribution of the non-volatile particle fraction as a proxy of the absorbing particle fraction has been installed at Zeppelin by NILU in late 2017.

Key findings aerosol optical properties Zeppelin: The aerosol absorption measurements at Zeppelin station are conducted in collaboration with the Greek Demokritos-Athens research institute, and continue a time series that has been started 2001. The decreasing trend in aerosol absorption observed since 2001 by Eleftheriadis et al. (2009) fits with the corresponding trend observed at other surface in situ stations in the Arctic (Coen et al., 2013).

3.1.3 Optical aerosol properties measured at the Trollhaugen Observatory

This section covers the aerosol optical properties collected at the station after the relocation in January 2014 from the Troll main base to Trollhaugen located above and upwind of the previous location. At the location of the old station, up to 80% of the collected data were contaminated by diffuse (unavoidable) emissions from the main buildings, making the scientific value and interpretation of the data from the old location difficult.

At Trollhaugen station, the same comprehensive set of instruments for observing optical aerosol properties as used in Birkenes is deployed. These cover the spectral scattering coefficient $\sigma_{sp}(\lambda)$, the spectral absorption coefficient $\sigma_{ap}(\lambda)$, as well as the derived properties scattering and absorption Ångström coefficient \hat{a}_{sp} and \hat{a}_{ap} , parameterising the wavelength dependence of σ_{sp} and σ_{ap} , as well as the single scattering albedo ω_0 . Figure 46 summarises the corresponding data collected at Trollhaugen for the years 2014 - 2017.

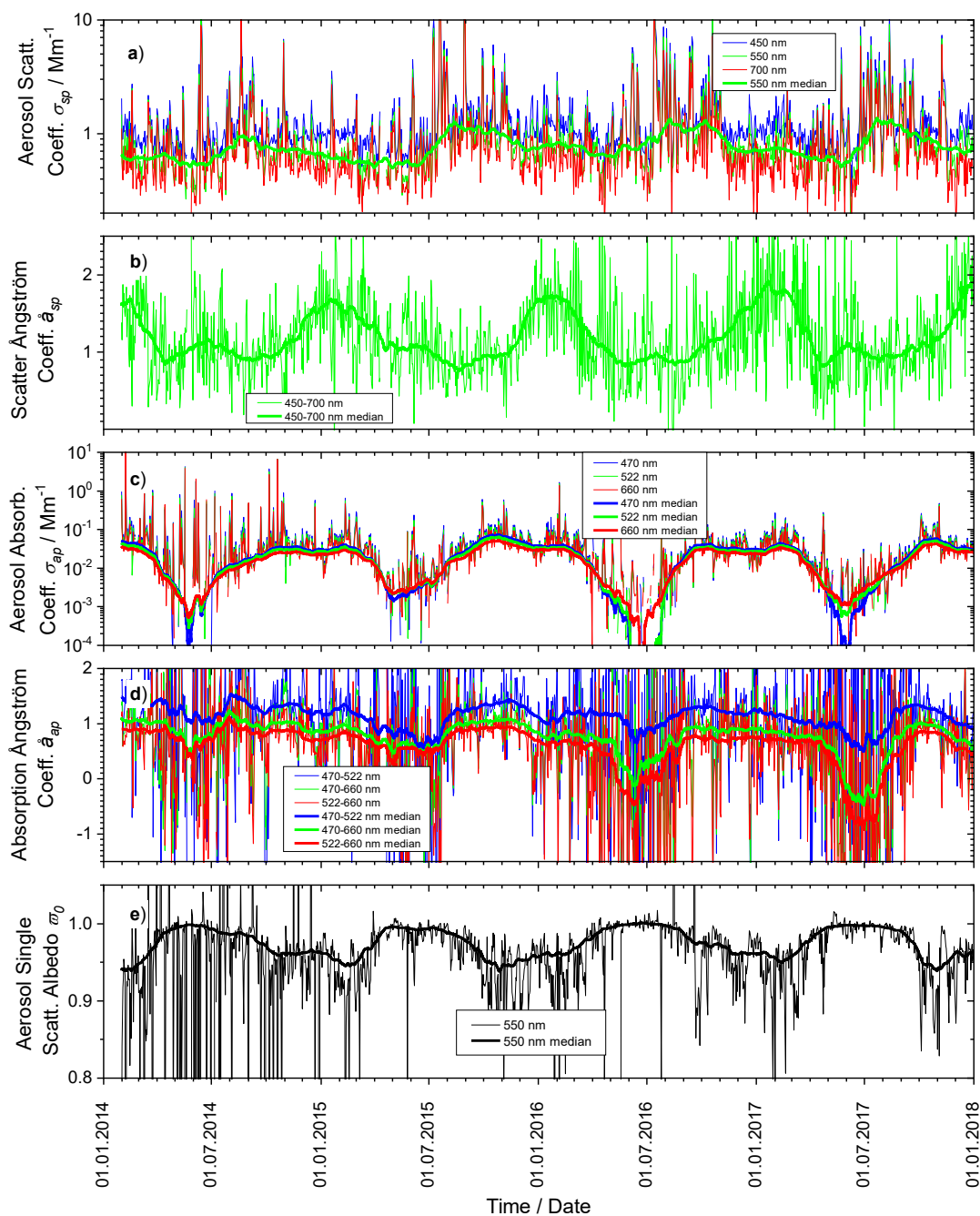


Figure 46: Time series of aerosol particle optical property daily means measured for 2014 - 2017 at Trollhaugen station. Panel a) shows the aerosol scattering coefficient σ_{sp} at 450, 550, and 700 nm wavelength measured by integrating nephelometer. Panel c) the aerosol absorption coefficient σ_{ap} at 470, 522, and 660 nm wavelength measured by filter absorption photometer. Panels b) and d) show the derived properties scattering and absorption Ångström coefficient \hat{a}_{sp} and \hat{a}_{ap} , respectively, while Panel e) depicts the single scattering albedo ω_0 . All plots also depict the running 8-week medians of the respective properties as heavy lines to visualize seasonal variations.

Starting with the time series of $\sigma_{sp}(\lambda)$ in panel a) of Figure 46, a slight annual cycle can be observed, with superimposed peaks throughout the year. The peaks are associated with intrusions of marine air during storm episodes, while the annual cycle is caused by an annual cycle of the particle number size distribution (PNSD) in Antarctic background air, with higher particle loads and larger particles in summer. The same annual cycle in the PNSD also

explains the annual cycle in the scattering Ångström coefficient \hat{a}_{sp} plotted in panel b). The scattering Ångström coefficient increases as the PNSD receives a relative peak at smaller particle sizes, which is the case in Antarctic background summer air.

An annual cycle can also be detected in the $\sigma_{ap}(\lambda)$ time series depicted in panel c) of Figure 46, even though the filter absorption photometer operates constantly around the detection limit. The minimum in the cycle occurs in Southern hemisphere winter when the Antarctic vortex decreases transport from lower latitudes through the lower and mid-troposphere. In the absorption Ångström coefficient \hat{a}_{ap} time series shown in panel d), the annual cycle is rather weakly pronounced, with the lowest values also occurring in winter.

The time series of the single scattering albedo ω_0 (panel e) of Figure 46) shows the highest values close to 1, i.e. almost no particle absorption at all, in Antarctic winter, coincident and consistent with the minimum in particle absorption. Lower ω_0 values, i.e. higher average particle absorption, occurs towards summer when the Antarctic continent is subject to stronger transport from mid-latitude sources through the lower part of the troposphere.

Optical aerosol properties are tightly connected to the physical aerosol properties, which is why they are ideally interpreted together in context. The annual cycle in the optical aerosol properties observed at Trollhaugen is caused by a corresponding annual cycle in the physical properties, which will be discussed in section 3.2.3.

Key findings aerosol optical properties Trollhaugen: The length of the time series of reliable data on optical aerosol properties at Trollhaugen is too short for drawing conclusions on trends. The annual cycle of the aerosol optical properties at Trollhaugen has been studied in detail, and is associated to a natural, hemispheric-scale atmospheric pattern. The same annual cycle with higher particle loads and larger particles in summer is observed also at other stations on the central Antarctic continent.

3.2 Measurements of aerosol number and size

3.2.1 Physical aerosol properties measured at the Birkenes Observatory

Figure 47 shows the time series of the particle number size distribution (PNSD) measured at Birkenes in 2017, separated into 4 different panels by season. In this plot type, the x-axis shows the time of the observation, whereas the y-axis holds the particle diameter D_p on a logarithmic scale. The logarithmic colour scale holds the particle concentration, normalised to the logarithmic size interval, $dN / d \log D_p$. The use of logarithmic axis is common when displaying PNSD information since both, particle diameter and particle concentration, tend to span several orders of magnitude while containing relevant information over the whole scale. In this report, the PNSD reported for Birkenes covers the whole size range between 0.01 - 10 μm by combining the information of 2 instruments, one each focussing on the fine ($D_p < 1 \mu\text{m}$) and coarse ($D_p > 1 \mu\text{m}$) size ranges, into a common PNSD product (see Appendix II for details). For the common PNSD product, both instruments need to agree in their overlap size range. At observation times when this criterion isn't met, the resulting PNSD is rejected, which can lead to gaps in the PNSD time series. This occurs predominantly in winter when the aerosol particles are absorbing, which affects the instrument covering the coarse size range. Existing, operating procedures and quality standards defined by the European research infrastructure ACTRIS have been used (Wiedensohler et al., 2012).

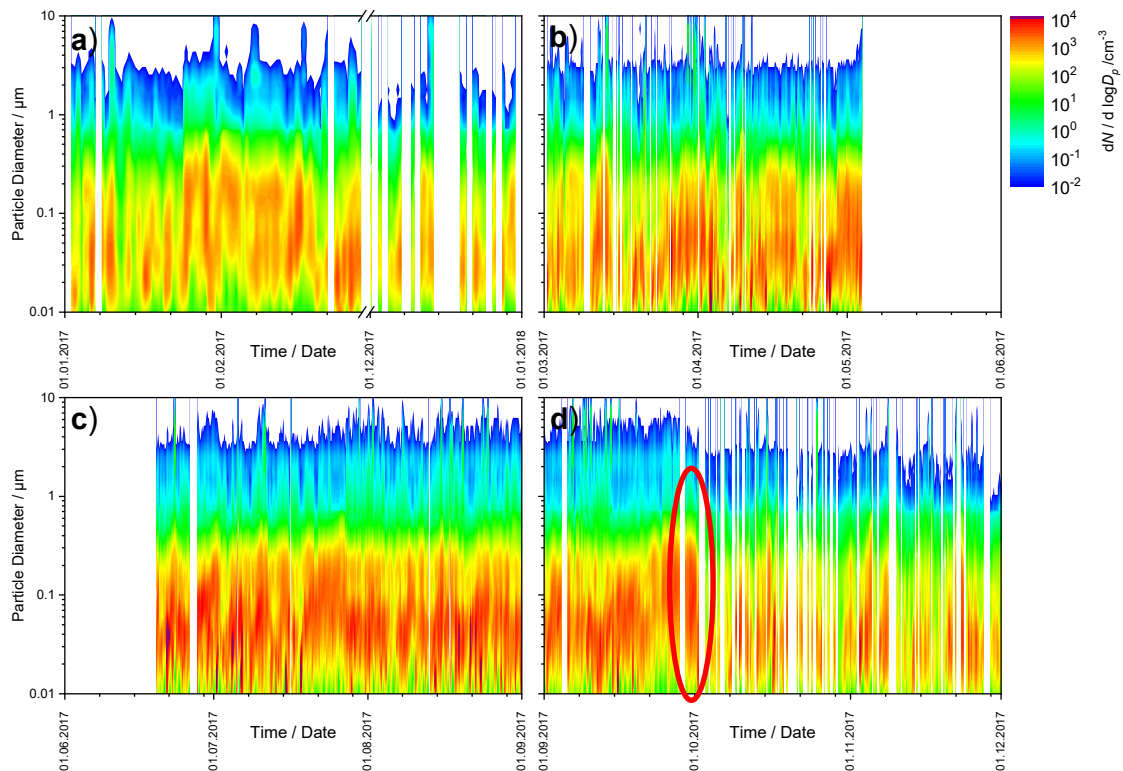


Figure 47: 2017 time series of particle number size distribution at Birkenes, panel a) winter, panel b) spring, panel c) summer, panel d) autumn. An episode associated to long range transport of biomass burning aerosols is starting on 27 September and lasting until 1 October is marked by a red oval shape.

There doesn't exist any unique connection between the PNSD and air mass type, but the PNSD still is normally fairly characteristic for the air mass, and can serve, together with the single scattering albedo ω_0 , and the scattering Ångström coefficient \hat{a}_{sp} , as valuable indication of air

mass origin, which at Birkenes shifts with the synoptic weather situation. Consequently, the information content of a PNSD time series plot is too high to be discussed in detail in this overview-type annual report. The PNSD and ω_0 observations reconfirm findings from earlier years on the dominant air mass types at Birkenes, which consist of: 1) clean Arctic background aerosol; 2) Central and Eastern European aerosol; 3) biogenic aerosol, i.e. vegetation emitted precursor gases condensing to the particle phase by photooxidation; 4) wood combustion aerosol from domestic heating.

Of particular interest in 2017 is an episode occurring around 27 September that year. Visible in panel d) of Figure 47, the episode is characterised by a PNSD dominated by a single accumulation mode peaking around $0.1 \mu\text{m}$ particle diameter. The episode in the PNSD data coincides with a peak in particle absorption of up to 11 Mm^{-1} , corresponding to a single scattering albedo down to 0.8. Aerosols exhibiting such a PNSD have been subject to intense auto-processing, either by being closed off from surrounding air, and/or by maintaining high particle concentrations under transport. In coincidence with elevated particle absorption, such a PNSD is indicative of an aerosol originating from wildfires. In the present case, this notion could be confirmed by use of trajectory analysis and satellite fire products, which connects the Birkenes episode to fires in the South Russian steppe.

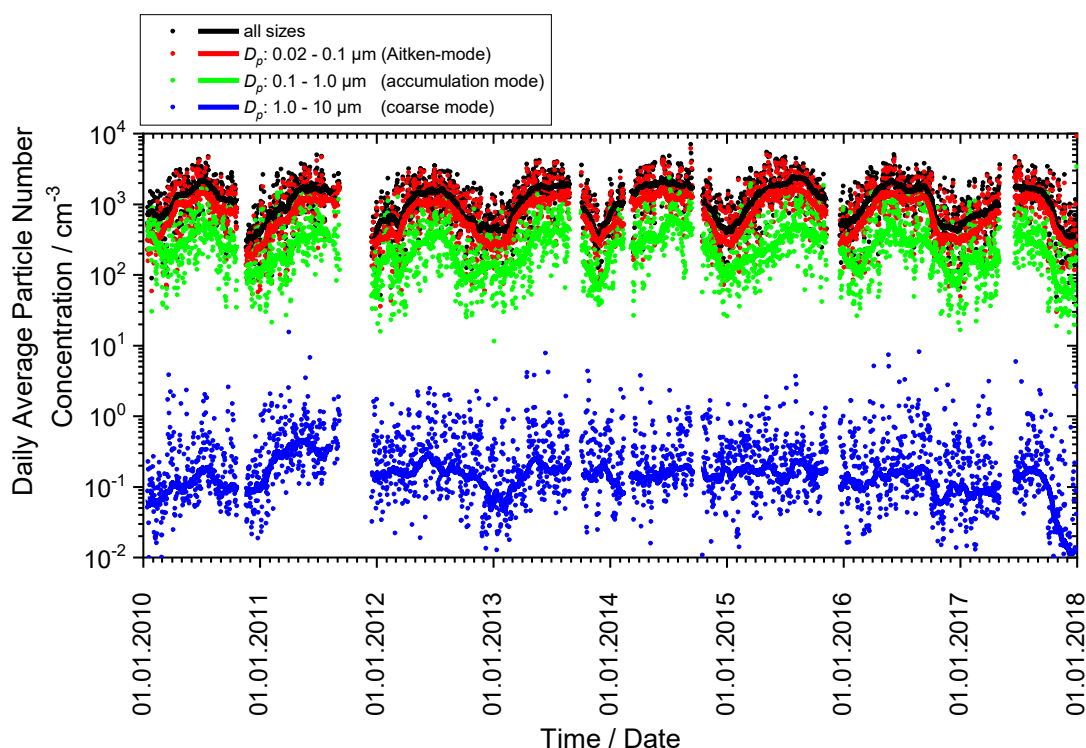


Figure 48: 2010-2017 time series of particle number concentration integrated over selected size ranges representing the different physical processes governing the atmospheric aerosol. The dotted graphs represent daily averages of the respective size range, the lines the 8-week running median.

In order to condense the information in the PNSD time series, Figure 48 shows the time series of selected PNSD integrals, i.e. the concentration of particles falling into selected size intervals. The size intervals are chosen to represent characteristic processes governing the atmospheric aerosol (see Appendix II for more details): 1) the Aitken-mode size range, $0.02 -$

0.1 μm ; 2) the accumulation mode size range, 0.1 - 1 μm ; 3) the coarse mode size range, 1 - 10 μm . The time series in Figure 48 represent daily averages over these PNSD integrals for the whole period since the Birkenes station upgrade in 2010, as well as the corresponding running 8-week medians to highlight seasonal variations. The respective size range integral particle concentrations are denoted N_{ait} for the Aitken mode, N_{acc} for the accumulation mode, and N_{coa} for the coarse mode.

As to be expected, the particle concentration in the Birkenes aerosol in absolute terms is dominated by the Aitken mode particles, followed by the accumulation mode. Also the most prominent feature in Figure 48 is exhibited by the particle concentrations in these 2 modes, a clear annual cycle caused by the same underlying physical process. In summer, the vegetation emits gaseous aerosol precursors, which are photo-oxidised and condense onto Aitken-mode particles or form those directly. These particles coagulate, increasing the concentration of accumulation mode particles. These same processes increasing N_{ait} and N_{acc} in summer are also responsible for increasing the scattering Ångström coefficient \hat{a}_{sp} in summer. The processes controlling N_{coa} are decoupled from those controlling N_{ait} and N_{acc} . Coarse mode particles are formed from bulk material, their concentration is affected by wind speed (levitating dust, spores, pollen), snow cover, and rain (both inhibiting dust levitation).

Key findings aerosol physical properties Birkenes: Size segregated aerosol particle concentrations at Birkenes do not exhibit any obvious trend. This corresponds to findings obtained at other Nordic stations within ACTRIS and EMEP. The particle size distribution at Birkenes is governed by 5 major sources: 1) clean Arctic background aerosol; 2) Central and Eastern European aerosol; 3) Arctic haze; 4) fine fraction biogenic aerosol; 5) wood combustion aerosol from domestic heating. An episode observed in late September 2017 can be traced to fires in the South Russian steppe.

3.2.2 Physical aerosol properties measured in situ at the Zeppelin Observatory

A Differential Mobility Particle Sizer (DMPS) instrument, measuring the particle size distribution, was installed at Zeppelin in 2016 primarily to calibrate the ACSM instrument but included continuous monitoring as an additional benefit. The same instrument type, operated in a different mode, is used to measure the particle number size distribution (PNSD) in the particle diameter range 0.01 μm - 0.8 μm at Birkenes and Trollhaugen stations. It was therefore decided to deploy the Zeppelin DMPS in the same way when not needed for calibrating the ACSM instrument.

As an additional feature, the Zeppelin DMPS has been upgraded late 2017 to also provide the PNSD of refractory (non-volatile) particles. The refractory particle fraction is often used as a proxy for the absorbing aerosol particle fraction, which will allow to study changes in particle absorption on a microphysical basis in the Arctic environment on a routine basis. The corresponding data [are already now available in real-real-time](#) with only 1 hour delay.

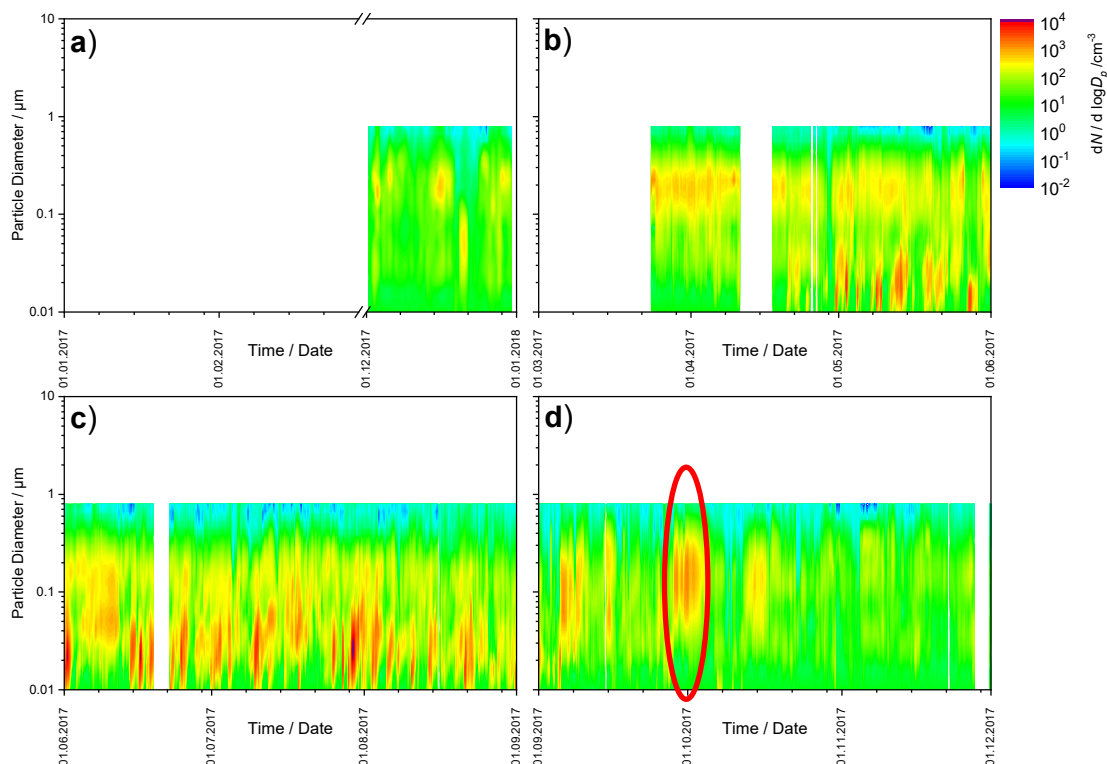


Figure 49: 2017 time series of particle number size distribution at Zeppelin, panel a) winter, panel b) spring, panel c) summer, panel d) autumn. An episode occurring around 1 October 2017 is marked with a red oval.

At Zeppelin, the DMPS is not combined with an instrument covering the coarse particle size range larger $1 \mu\text{m}$. This limits the available PNSD information the DMPS size range. Figure 49 presents the Zeppelin PNSD data available for this report for 2017 in the same way as for Birkenes and Trollhaugen, with one PNSD time series panel per season. The coverage gap in early 2017 is due to a pump failing despite predictive maintenance, combined with the challenge of accessing the station that time of year.

In contrast to mid-latitudes, air masses don't shift quite as rapidly at polar latitudes due to a smaller influence of frontal systems, making weather patterns more persistent at least in parts of the year. This allows to interpret the information on air mass types contained in PNSD data in more detail. Focussing in on Panel a) of Figure 49 displaying the PNSD data of December 2017, periods with a uni-modal size distribution are visible. This means the PNSD is dominated by a single mode, in this case the accumulation mode with a peak around $0.25 \mu\text{m}$ particle diameter. Such a PNSD is typical of early-phase Arctic haze (Heintzenberg, 1980), which is also detectable in late autumn (Panel d) of Figure 49). Arctic haze is formed by auto-processing of aerosol particles trapped under the winter Arctic vortex while the particle mass increases from industrial emissions under the vortex (e.g. Law & Stohl, 2007).

The late spring and summer PNSD at Zeppelin (Figure 49 panels c) and d) is somewhat more variable than in winter due to less stable atmospheric conditions. Particle formation events can be observed, with peak in the PNSD at particle diameters between $0.01 - 0.02 \mu\text{m}$. These are triggered by photo-chemical production of chemical species that condense into the particle phase, and don't find enough existing particle surface to condense on, thus forming new particles. A more unusual event can be observed in the 2017 Zeppelin PNSD data around 1 October. The PNSD is dominated by a single mode peaking around $0.15 \mu\text{m}$ particle diameter. This indicates again strong auto-processing of the aerosol, which is unusual for

Arctic summer. The peak in the PNSD coincides with a peak in particle absorption of about 2 Mm^{-1} . Trajectory analysis indicate fires in the South Russian steppe as source of this event, i.e. the same fires that were registered in Birkenes 3 days before.

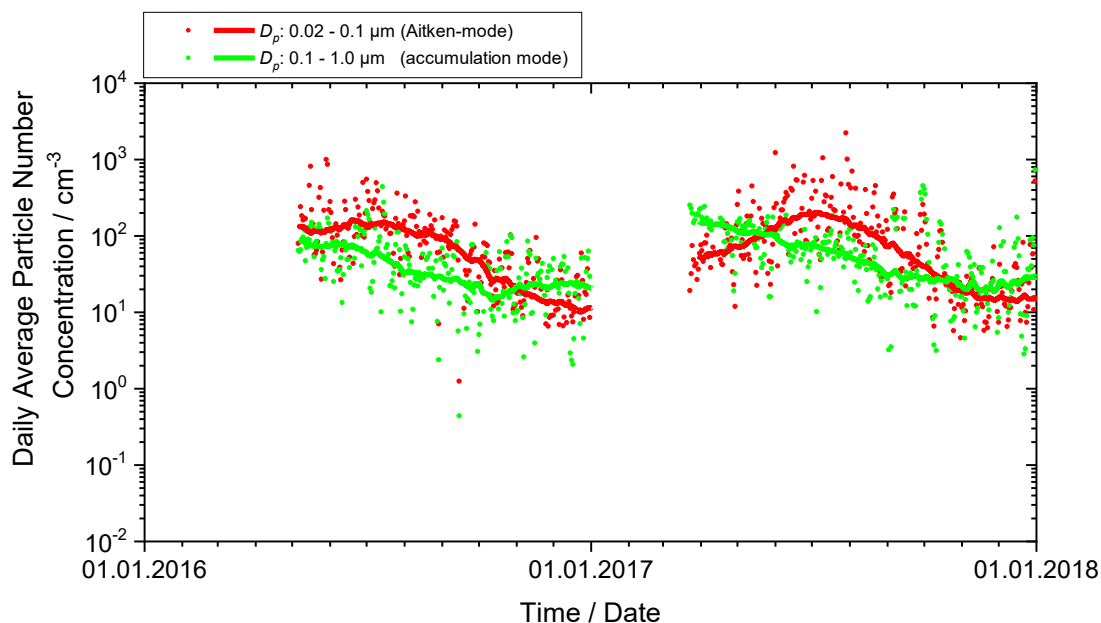


Figure 50: 2016-2017 time series of particle number concentration integrated over selected size ranges representing the different physical processes governing the atmospheric aerosol. The dotted graphs represent daily averages of the respective size range, the lines the 8-week running median.

Figure 50 summarises the 2016 Zeppelin PNSD data by displaying time series of the PNSD size integrals for the Aitken mode N_{ait} ($0.02 \mu\text{m} < D_p < 0.1 \mu\text{m}$) and N_{acc} ($0.1 \mu\text{m} < D_p < 0.8 \mu\text{m}$). Apart from particle concentrations an order of magnitude lower than at boreal Birkenes, the most prominent feature of the graph is the point in early November where the N_{acc} running median becomes consistently larger than N_{ait} running median. This behaviour is typical for auto-processed aerosols, and occurs normally only for shorter events. At Zeppelin, this point marks the onset of the Arctic haze period.

Key findings aerosol physical properties Zeppelin: Zeppelin particle size distribution data so far available for this report exhibit the well-known Arctic haze pattern. Trends in this pattern cannot be studied in this context due to too short the time series. An episode occurring at Zeppelin around 1 October 2017 can be traced back to wildfires in the Southern Russian steppe. The same wildfire emissions have been detected at Birkenes 3 days earlier.

3.2.3 Physical aerosol properties measured in situ at the Trollhaugen Observatory

In contrast to mid-latitudes, polar latitudes exhibit more stable atmospheric conditions with less influence of frontal systems. This is especially true for the Antarctic continent due to colder inland temperatures and the absence of land masses in the mid- to high-latitude

Southern hemisphere that could disturb the atmospheric air flow, as compared to the Northern hemisphere. This makes air masses well-defined in these regions, and their associated aerosol properties easier to interpret than under shifting Northern mid-latitude conditions.

This section only covers data collected at the station after the relocation in January 2014 from the Troll main base to Trollhaugen located above and upwind of the previous location. At the old station location, up to 80% of the collected data were locally contaminated by diffuse (unavoidable) emissions from the main station buildings, making a statistically meaningful interpretation of the data from the old location difficult.

The DMPS system measuring the PNSD at Trollhaugen station has been re-built on-site during the station maintenance visit of January / February 2016, causing a gap in the PNSD time series in January 2016. After the re-built, the system conforms to the quality standards of the ACTRIS research infrastructure, and the observed size range has been extended from 0.03 - 0.8 μm to 0.01 - 0.8 μm , making it now suitable to study formation of new particles. As a consequence, absolute particle concentrations measured by the system before and after remodelling are not directly comparable. This disadvantage has been accepted in favour of a wider range of applications for the DMPS system.

Figure 51 plots the 2017 Trollhaugen PNSD data in the same way as for Birkenes. Due to the rather well-defined atmospheric conditions over the Antarctic continent, 2016 is rather representative also for previous years. Please observe that the times for the panels are the same as for the Northern hemisphere stations, but that seasons in the Southern hemisphere are shifted by 6 months.

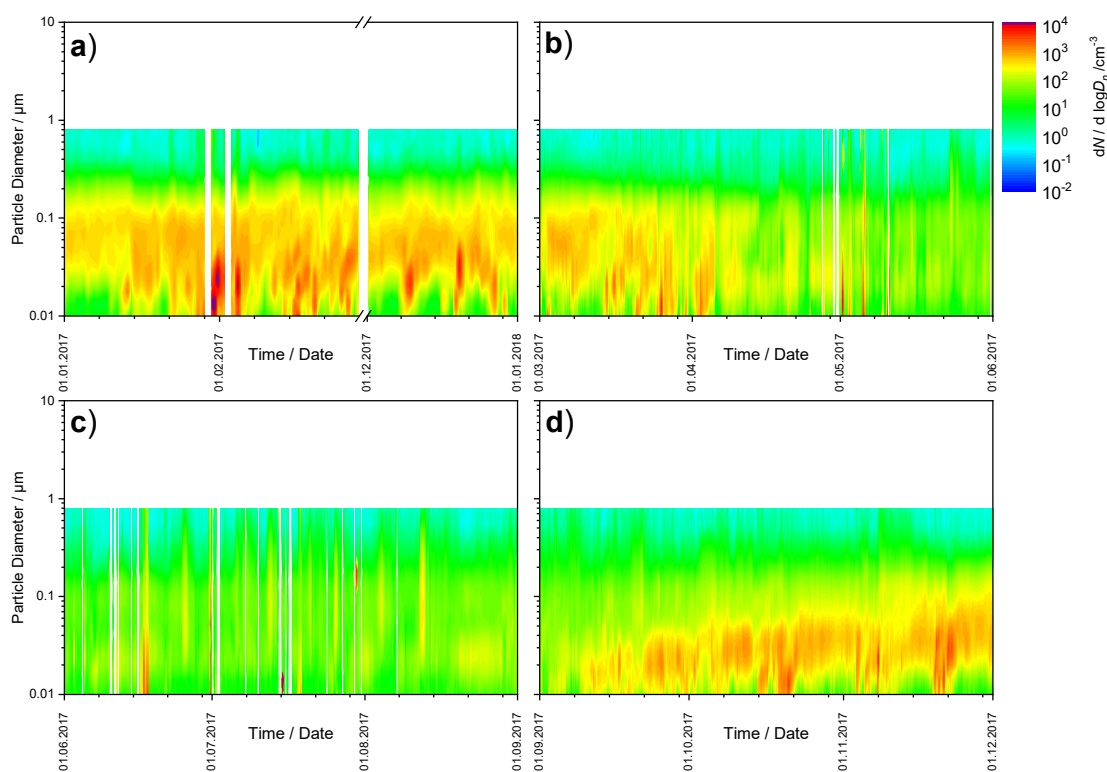


Figure 51: 2017 time series of particle number size distribution at Trollhaugen, panel a) summer, panel b) autumn, panel c) winter, panel d) spring.

Even though the Antarctic continent is among the most pristine regions on the globe, new particle formation events triggered by photo-chemical oxidation of precursor substances can be observed in Antarctic summer. Formation of new particles is not a function of available condensable vapour alone, but also of the available particle surface. If the available particle surface becomes too small to accept the condensing vapour, new particles are formed. This ratio doesn't depend on the absolute amount of aerosol particles. Thus, formation of new particles can occur anywhere on the globe.

A year-round feature observed at Trollhaugen are intrusions of marine air associated with marine storms. These have been observed even at South Pole, and are visible in the PNSD when the particle concentration at the upper end of the particle size range increases.

The probably most prominent feature in the Trollhaugen annual PNSD time series data is the annual cycle visible in the background aerosol. Particle concentrations are low in winter over the whole observed PNSD size range, and nucleation events lacking. In spring, particle concentrations in the diameter range $0.01 - 0.03 \mu\text{m}$ increase, and the central particle diameter of this peak increases throughout spring until reaching values around $0.09 \mu\text{m}$ in summer. Towards autumn and winter, the cycle reverses.

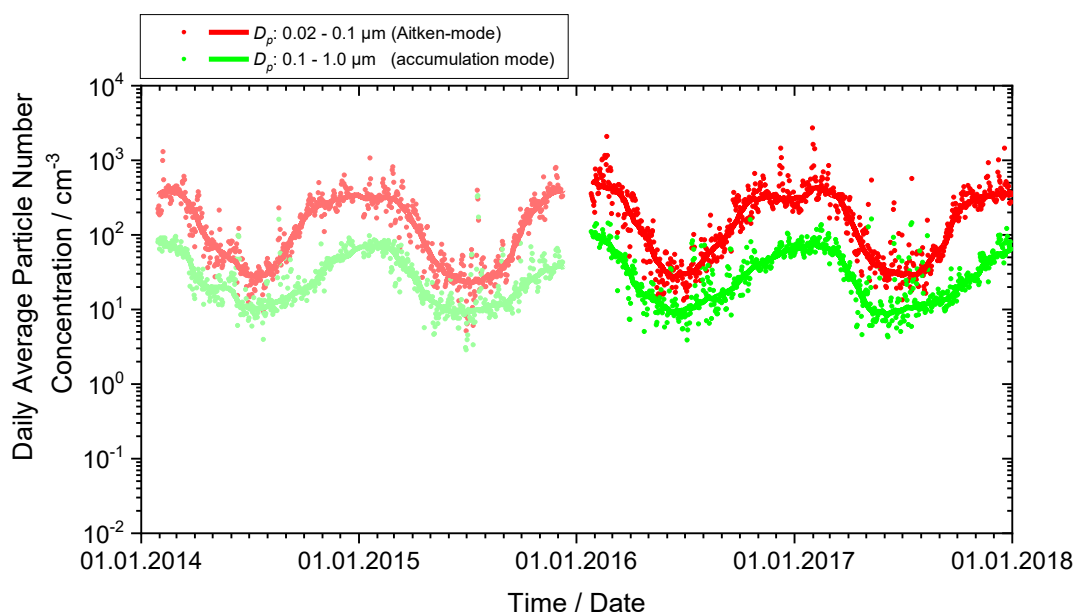


Figure 52: 2014-2017 time series of particle number concentration integrated over selected size ranges representing the different physical processes governing the atmospheric aerosol. The dotted graphs represent daily averages of the respective size range, the lines the 8-week running median. Data of the DMPS instrument prior to its upgrade are plotted in lighter colour as reminder that data before and after remodelling aren't directly comparable due to extension of the particle size range observed by the instrument.

This annual cycle in the Antarctic background PNSD is reflected by the PNSD size integrals for the Aitken mode N_{ait} ($0.02 \mu\text{m} < D_p < 0.1 \mu\text{m}$) and N_{acc} ($0.1 \mu\text{m} < D_p < 0.8 \mu\text{m}$) plotted in Figure 52, with higher particle concentrations in summer than in winter. A study based on data collected at Troll (Fiebig et al., 2014) connects the annual cycle to transport patterns seen by Antarctic background air. Antarctic background air descends over the central continent from the upper troposphere / lower stratosphere region, where it has been transported from mid-latitudes or even the tropics at the same altitude. At these lower

latitudes, it had been uplifted by frontal conveyor belts (mid-latitudes) or tropical anvil clouds. The uplift process, associated with extensive wet removal of particles in the frontal or anvil cloud system, explains the cleanliness of the air.

Fiebig et al. (2014) raise the hypothesis that the annual cycle of the Antarctic background aerosol is caused by the annual variation of solar insolation seen by the air during this transport pattern. The hard UV radiation in the upper troposphere would oxidise water-insoluble vapours that survived wet removal during uplift of the air. The oxidation products would subsequently condense into the particle phase, causing the observed annual cycle in the Antarctic background PNSD. Studying this hypothesis further is subject of the currently ongoing AtmosCAir project funded by the Norwegian Antarctic Research Expedition (NARE) programme.

Key findings aerosol physical properties Trollhaugen: The length of the time series of reliable data on the aerosol particle number size distribution at Trollhaugen is **too short for drawing conclusions on trends**. The annual cycle of the PNSD at Trollhaugen has been studied in detail, and is associated to a natural, hemispheric-scale atmospheric pattern. Antarctic background air descends over the Central continent from the upper troposphere / lower stratosphere. At that altitude, it has been transported to the pole after being uplifted (and cleaned by wet removal) at mid- or tropical latitudes.

3.3 Summary of physical and optical aerosol properties

Aerosol particle absorption and scattering are decisive for the cooling or warming effect of aerosols in climate. All types of aerosols are scattering solar radiation, but the higher fraction of aerosol absorption, the more warming is the aerosol. The absorption is depending on the composition; black carbon (e.g. soot), certain organic compounds (brown carbon), and minerals absorb radiation. The number size distribution of the aerosol particles also determine the scattering properties. In order to summarise the measurements of physical and optical aerosol properties observed at Birkenes, Trollhaugen, and Zeppelin stations, annual and seasonal means of the main parameters are collected in Table 5 for the physical parameters, and in Table 6 for the optical parameters.

Focussing first on the columns for Birkenes, Aitken-mode particle concentrations N_{ait} for the summer and winter show surprisingly little variation over the years. Relative to previous years, particle concentrations in 2017 for these seasons are close to the multi-year average. Differences appear in comparison to the previous years for spring and autumn. For these seasons, the N_{ait} average concentrations are at the lower end of the variability range. Variations of this kind are not unusual, and depend on the actual transition from winter to summer weather and vice versa. Late onset of summer weather in spring and early onset of winter weather in autumn will decrease the N_{ait} particle concentrations for these seasons. Also for the accumulation mode seasonal average particle concentrations N_{acc} , spring and autumn values are at the lower end of the variability range, whereas summer and winter values don't show exceptional deviations.

At Birkenes, N_{ait} values for winter are typically around 33% of their summer values due to lack of biogenic particle production from the gas-phase. They depend on emissions from domestic

heating, with higher values for colder winters. With spring and autumn as transition periods between the winter and summer extreme values, the variability of N_{ait} in these seasons depends on the pace of transition between summer and winter. The same tendencies apply to the accumulation mode particle concentration N_{acc} .

Coarse mode particle concentrations at Birkenes have a tendency to be up to a factor of 2 smaller in winter than in summer due to less crustal particle production with a snow-covered ground. Particle concentrations at Birkenes don't exhibit any obvious trend over time.

Aitken and accumulation mode particle concentrations at Trollhaugen are an order of magnitude lower than at Birkenes, reflecting the pristine conditions in Antarctica. Their pronounced annual cycle and its cause has been discussed in section 3.2.3. Both concentrations show little variability over the years, reflecting the fact that atmospheric composition in Antarctica is still mostly governed by natural processes whose large scale pattern changes slowly.

The conclusion of no obvious trends applies also to the optical aerosol properties observed at Birkenes and Trollhaugen. At Birkenes, the annual and inter-annual variability is governed by the same processes mentioned when discussing the particle concentrations. More specifically, the aerosol is more absorbing in winter, and hence more warming, due to emissions from wood burning for domestic heating, which come on top of a baseline of absorbing aerosol emitted from traffic. This is reflected in higher particle absorption coefficient values in winter, and even more significantly in lower winter values of the single scattering albedo (lower meaning higher average particle absorption).

A long-range transport episode of wildfire emissions originating from the Southern Russian steppe has been observed in 2017 at both Birkenes and Zeppelin stations. The episode begins on 27 September at Birkenes, and 3 days later at Zeppelin. The aerosol during this episode is characterised by a uni-modal particle size distribution and a peak in high particle absorption values.

Table 5: 2010 - 2017 seasonal and annual means of integral particle concentrations in the ultrafine, fine and coarse particle size range for Birkenes, Trollhaugen, and Zeppelin stations, as far as available.

Year	Season	Birkenes				Trollhaugen ¹		Zeppelin	
		N _{ait} / cm ⁻³	N _{acc} / cm ⁻³	N _{coa} / cm ⁻³	N _{tot} / cm ⁻³	N _{ait} / cm ⁻³	² N _{acc} / cm ⁻³	N _{ait} / cm ⁻³	² N _{acc} / cm ⁻³
2009/10	Winter	440	384	0.087	824				
2010	Spring	1030	324	0.311	1354				
2010	Summer	1511	488	0.323	1999				
2010	Autumn	835	299	0.260	1135				
2010	Whole Year	973	362	0.256	1336				
2010/11	Winter	454	285	0.311	739				
2011	Spring	1127	369	0.639	1496				
2011	Summer	1391	438	0.572	1829				
2011	Autumn	1594	464	0.966	2059				
2011	Whole Year	1047	371	0.565	1418				
2011/12	Winter	424	213	0.305	637				
2012	Spring	1107	271	0.386	1378				
2012	Summer	1314	392	0.485	1706				
2012	Autumn	661	152	0.365	814				
2012	Whole Year	889	263	0.375	1152				
2012/13	Winter	383	183	0.183	566				
2013	Spring	1190	352	0.411	1543				
2013	Summer	1519	447	0.467	1967				
2013	Autumn	701	162	0.417	864				
2013	Whole Year	1020	304	0.391	1324				
2013/14	Winter	699	333	0.347	1033				
2014	Spring	1464	402	0.334	1866	183	32		
2014	Summer	1723	625	0.343	2349	43	19		
2014	Autumn	1122	446	0.385	1568	207	28		
2014	Whole Year	1279	456	0.338	1735	183	34		
2014/15	Winter	549	192	0.307	741	368	67		
2015	Spring	1425	332	0.348	1757	134	25		
2015	Summer	1979	559	0.395	2539	38	23		
2015	Autumn	1130	422	0.257	1553	221	28		
2015	Whole Year	1326	390	0.340	1717	171	32		
2015/16	Winter	542	233	0.297	776				
2016	Spring	1328	442	0.449	1770	170	26		
2016	Summer	1455	484	0.517	1940	47	18	156	64
2016	Autumn	895	290	0.312	1185	262	35	47	31
2016	Whole Year	1063	357	0.392	1421	231	37	---	---
2016/17	Winter	526	223	0.232	750	473	74	---	---
2017	Spring	855	216	0.271	1072	157	31	96	126
2017	Summer	1605	402	0.424	2007	51	20	282	70
2017	Autumn	852	250	0.228	1103	265	27	56	63
2017	Whole Year	943	272	0.267	1216	238	38	---	---

Cells shaded in grey mark values obtained with an older instrument version that can't be compared directly with later values.

¹Numbers given for the time when the respective season is present in the Northern hemisphere. Actual seasons in Southern hemisphere are shifted by 6 months.

²The accumulation mode integral particle concentration N_{acc} at Trollhaugen and Zeppelin extends only up to 0.8 µm particle diameter due to lack of an instrument covering larger particles. For Birkenes, N_{acc} includes particles up to 1 µm diameter

Table 6: 2010 - 2017 seasonal and annual means of optical aerosol properties scattering coefficient, absorption coefficient, and single scattering albedo for Birkenes, Trollhaugen, and Zeppelin stations, as far as available.

Year	Season	Birkenes			Trollhaugen ¹			Zeppelin		
		σ_{sp} (550 nm) / Mm ⁻¹	σ_{ap} (550 nm) / Mm ⁻¹	ω_0 (550 nm)	σ_{sp} (550 nm) / Mm ⁻¹	σ_{ap} (550 nm) / Mm ⁻¹	ω_0 (550 nm)	σ_{sp} (550 nm) / Mm ⁻¹	σ_{ap} (550 nm) / Mm ⁻¹	ω_0 (550 nm)
2009/10	Winter	16.82	3.09	0.88						
2010	Spring	12.33	0.78	0.93						
2010	Summer	11.30	0.70	0.94						
2010	Autumn	7.26	0.71	0.90						
2010	Whole Year	11.52	1.24	0.91						
2010/11	Winter	16.96	2.18	0.89						
2011	Spring	18.67	1.26	0.93						
2011	Summer	15.43	0.74	0.95						
2011	Autumn	29.74	2.87	0.92						
2011	Whole Year	20.26	1.69	0.93						
2011/12	Winter	11.29	1.00	0.91						
2012	Spring	15.10	0.86	0.93						
2012	Summer	12.62	0.67	0.95						
2012	Autumn	9.80	0.65	0.92						
2012	Whole Year	12.22	0.83	0.92						
2012/13	Winter	12.48	1.84	0.84						
2013	Spring	17.03	1.48	0.90						
2013	Summer	13.81	1.15	0.92						
2013	Autumn	8.89	1.25	0.85						
2013	Whole Year	13.73	1.40	0.88						
2013/14	Winter	22.89	2.64	0.87						
2014	Spring	12.95	2.09	0.87	0.74	-0.05	0.95			
2014	Summer	15.85	1.26	0.92	1.39	0.04	0.98			
2014	Autumn	18.76	3.41	0.82	1.02	0.15	0.93			
2014	Whole Year	16.99	2.30	0.87	1.01	0.09	0.95			
2014/15	Winter	13.98	1.30	0.89	0.74	0.04	0.94			
2015	Spring	12.72	1.48	0.89	0.65	0.02	0.97			
2015	Summer	12.45	1.46	0.90	2.44	0.02	0.98		0.30	
2015	Autumn	15.69	2.45	0.95	1.32	0.07	0.94		0.14	
2015	Whole Year	14.36	1.56	0.90	1.32	0.04	0.96			
2015/16	Winter	13.59	1.24	0.88	0.87	0.05	0.94		0.38	
2016	Spring	14.86	1.10	0.91	0.78	0.17	0.97		0.39	
2016	Summer	11.93	0.77	0.94	2.01	0.00	0.99		0.09	
2016	Autumn	11.47	1.46	0.85	1.54	0.04	0.95		0.12	
2016	Whole Year	12.26	1.12	0.89	1.31	0.06	0.97		0.24	
2016/17	Winter	12.27	2.24	0.81	0.82	0.05	0.94		0.40	
2017	Spring	8.71			0.76	0.02	0.97		0.48	
2017	Summer	8.58			1.57	0.01	0.99		0.10	
2017	Autumn	8.09	1.21	0.85	1.22	0.05	0.94		0.29	
2017	Whole Year	9.07			1.10	0.03	0.96		0.33	

Values stated in cells shaded in grey are re-evaluated according to latest inter-comparisons within ACTRIS and therefore deviate from values given in earlier years.

¹Numbers given for the time when the respective season is present in the Northern hemisphere. Actual seasons in Southern hemisphere are shifted by 6 months.

3.4 Column optical aerosol properties measured by ground-based remote sensing

Ground-based remote sensing of the optical characteristics of aerosols in the atmospheric total column is conducted with multi-wavelength sun-photometers. A sun-photometer is oriented towards the sun to detect the solar radiation attenuated along the slant path from the top-of-atmosphere to the ground. The atmospheric aerosol load leads to a decrease in the solar radiation transmitted through the atmosphere. This decrease depends on the aerosol optical depth (AOD), which is given by the integral of the volume aerosol extinction coefficient along the vertical path of the atmosphere. The wavelength dependence of AOD, described by the Ångström exponent (\AA) is a qualitative indicator of the particle size and contains information about the aerosol type. The larger the Ångström exponent, the smaller the size of the particles measured.

Photos of instruments used for monitoring of spectral resolved AOD at Birkenes and Ny-Ålesund, their main characteristics are given in Appendix II, and detailed Tables with monthly data for all years are given in Appendix I.

3.4.1 Column optical aerosol properties measured by ground-based remote sensing at Birkenes Observatory

AOD measurements started at the Birkenes Observatory in spring 2009, utilizing an automatic sun and sky radiometer (CIMEL type CE-318, instrument #513). The retrieval method is that of the AERONET version 2 direct sun algorithm (for details: <http://aeronet.gsfc.nasa.gov>). Quality assured (Level 2) data are available for the nine years of operation, 2009 - 2017. Due to technical problems with both the instrument and the communication system at the observatory, usable measurements resumed only by the end of April 2017. They continued until mid-October, followed by several weeks of unfavourable weather conditions. In early November, the instrument was dismantled and sent to the University of Valladolid for annual calibration. Also this measurement gap turned out to be long-lasting (until mid-April 2018), and for this reason, it was decided to purchase a second Cimel instrument, which, from winter 2018/19, will be operated and calibrated alternately with the current instrument in order to gain truly continuous observations. With this, we hope to fill the current lack of winter observations as far as illumination and weather conditions allow it.

Starting in 2017, we also now use level 2 data from version 3 (V3) of the Aeronet data analysis. The version provides significantly more data than version 2 (V2). Results from a comparison between the V3 and the V2 data sets (AOD@500 nm, (470nm, 800nm) Ångström coefficient) and implications for trends are described in appendix II.

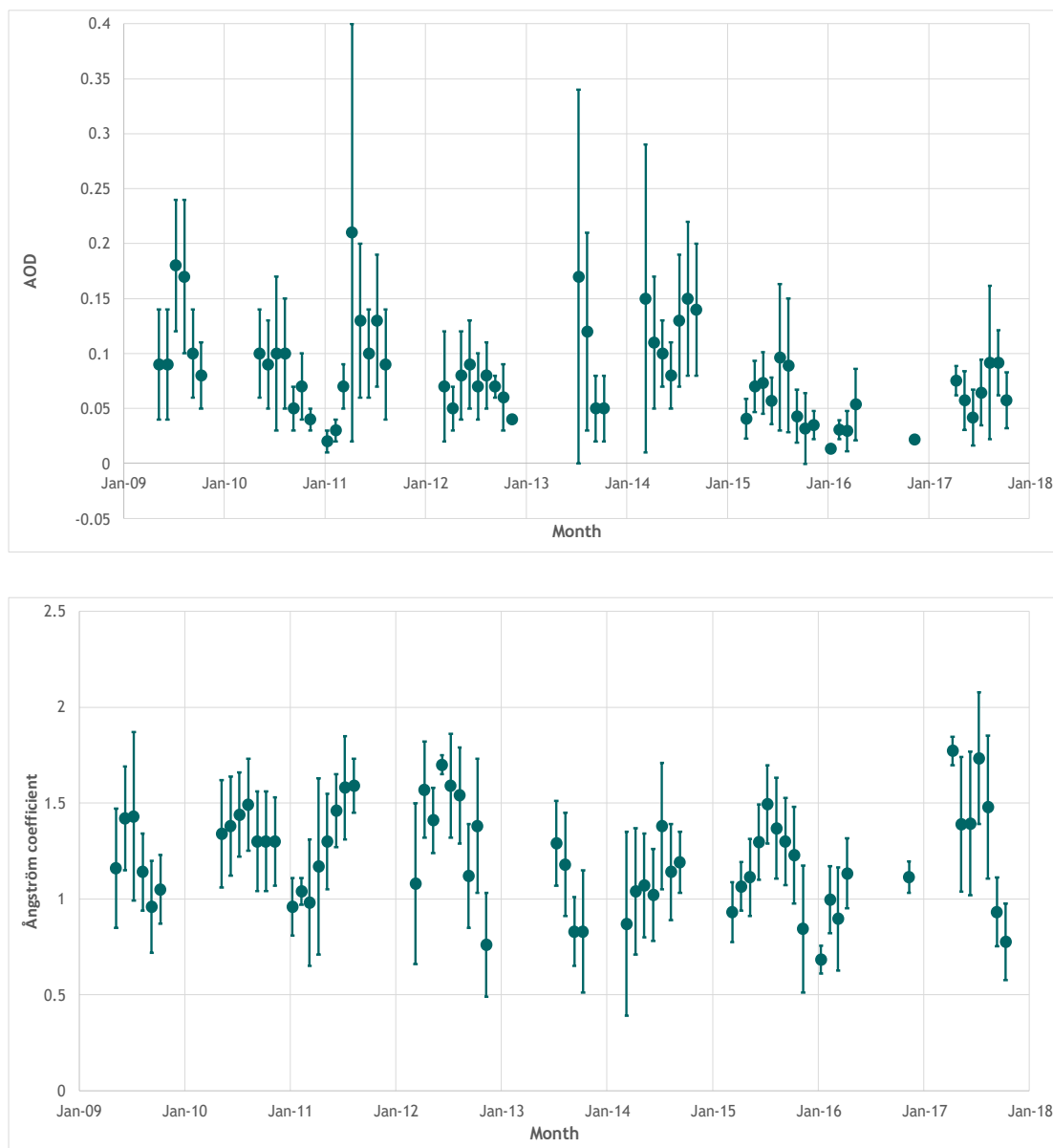


Figure 53: 2009 - 2017 time series of aerosol optical depth (AOD) at 500 nm wavelength in the atmospheric column above Birkenes (upper panel) and (470 nm, 800 nm) Ångström coefficient describing the AOD wavelength dependence (lower panel). Mean values and standard deviations are given.

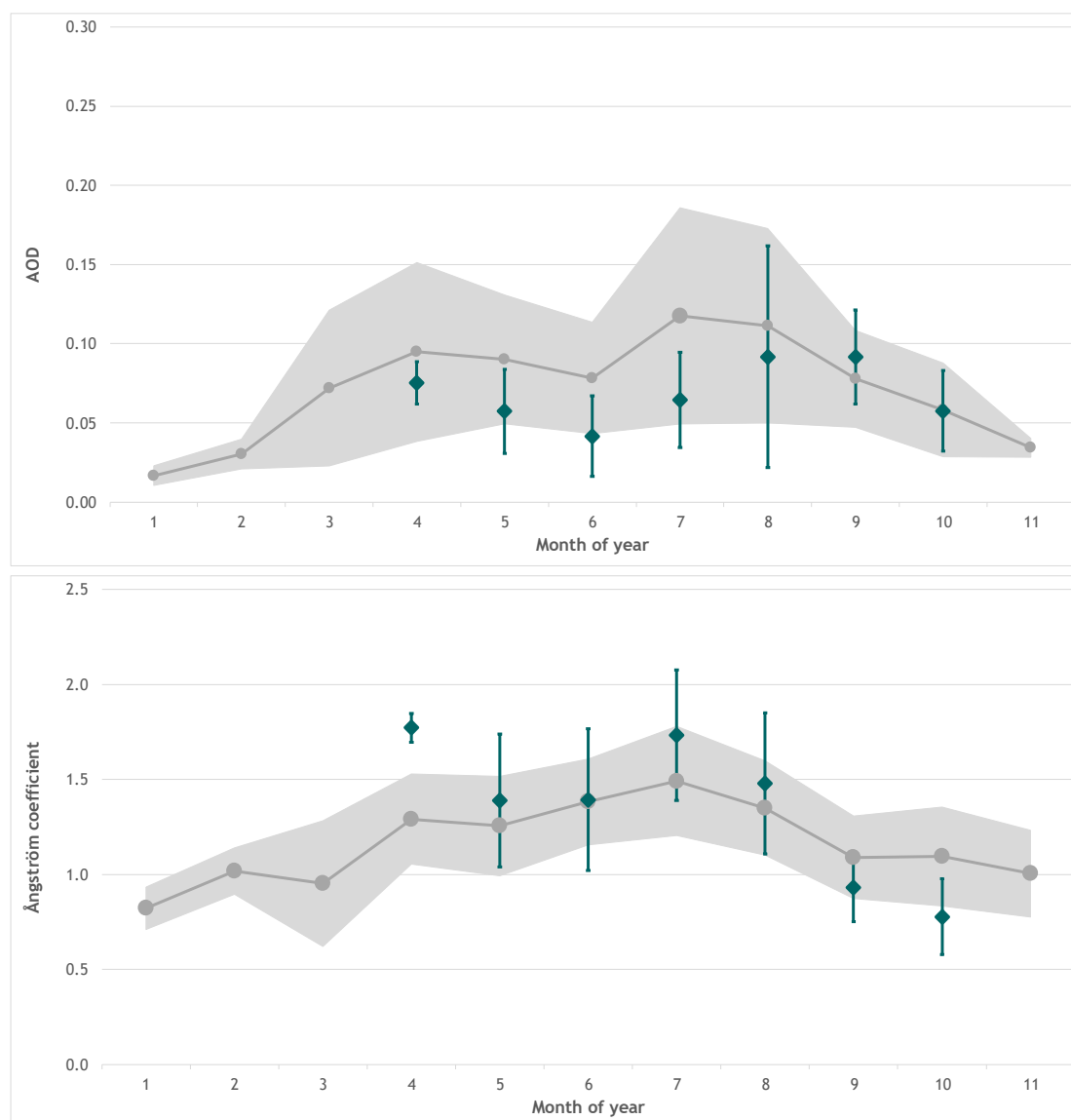


Figure 54: Monthly mean aerosol optical depth (AOD) at 500 nm wavelength in the atmospheric column above Birkenes (upper panel) and (470, 800 nm) Ångström coefficient describing the AOD wavelength dependence (lower panel). Mean values and standard deviations are given. Values marked in grey are the mean and standard deviations for the time period 2009-2017.

The AOD and Ångström coefficient time series and seasonal variations for 2017 are shown in Figure 53 and Figure 54. The 2017 monthly mean and mean values for all years are shown in Table 7. Data for all years are given in Appendix I. There are no obvious (and statistically significant) trends visible in the nine years of observations. In general, AOD measured at Birkenes are relatively low, compared to central European observations. E.g., AERONET climatological values for Cabauw, the Netherlands, vary between 1.2 ± 0.06 in December and 3.1 ± 0.19 in April). In 2017, monthly-mean AOD values at Birkenes were markedly below the long-term means in spring and summer, and close to the means in autumn (September, October). The highest monthly average AOD of 0.09 was observed in September, which is slightly above the long-term mean, but well within its standard deviations. There was one episode with significantly higher AOT values in late August (maximum AOT 0.24) which may have been due to forest fires, but it has not been investigated in detail yet.

Ångström exponent monthly means varied considerably with significant deviations from long-term means, but mostly within the 2σ range of long-term means. The large positive deviation in April can be explained by the fact that this means only contains two days (29, 30 April), while the very small Ångström exponents in September and especially October are not due to poor statistics, but consistent throughout this period.

Table 7: Monthly mean values for 2017 and mean for the time period 2009-2017, plus standard deviations, for aerosol optical depth (AOD) and Ångström coefficient observed in Birkenes. In addition, the number of days with cloud free and quality assured observations are given.

Month/Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
Aerosol optical depth (AOD)											
2017				0.08 ±0.01	0.06 ±0.03	0.04 ±0.03	0.06 ±0.03	0.09 ±0.07	0.09 ±0.03	0.06 ±0.03	
Mean 09-17	0.02 0.01	0.03 ±0.01	0.07 ±0.05	0.09 ±0.06	0.09 ±0.04	0.08 ±0.04	0.12 ±0.07	0.11 ±0.06	0.08 ±0.03	0.06 ±0.03	0.03 ±0.01
Ångström coefficient (Å)											
2017				1.77 ± 0.07	1.39 ± 0.35	1.39 ± 0.37	1.73 ± 0.34	1.48 ± 0.37	0.93 ± 0.18	0.78 ± 0.20	
Mean 09-17	0.82 ±0.11	1.02 ±0.12	0.95 ±0.33	1.29 ±0.24	1.25 ±0.26	1.38 ±0.23	1.49 ±0.29	1.35 ±0.25	1.09 ±0.22	1.09 ±0.26	1.00 ±0.23
Number of days with cloud-free and quality assured observations (AERONET level 2)											
2017				2	17	15	17	11	6	8	
Total 09-17	9	13	61	65	110	129	144	118	74	60	13

Key findings column-integrated optical aerosol properties Birkenes: There is no statistically significant trend in the nine years of AOD observations at Birkenes, but 2017 is the third year in a row with low AOD values through the measurement season. In August, a short episode of higher AOD was observed.

3.4.2 Column optical aerosol properties measured by ground-based remote sensing at Ny-Ålesund

In 2002, Physikalisch-Meteorologisches Observatorium Davos/*World Radiation Center* (PMOD/WRC) in collaboration with NILU, started AOD observations in Ny-Ålesund (at the Sverdrup station, 46 m a.s.l.) as part of the global AOD network on behalf of the WMO GAW programme. A precision filter radiometer (PFR) measures the extinction in four narrow spectral bands at 368 nm, 415 nm, 500 nm and 862 nm. Data quality control includes instrumental control like detector temperature and solar pointing control as well as objective cloud screening. Ångström coefficients are derived for each set of measurements using all four PFR channels. Calibration is performed annually at PMOD/WRC. Quality assured data are available at the World Data Centre of Aerosols (WDCA), hosted at NILU (see <https://ebas.nilu.no>)

In Ny-Ålesund, the solar elevation is less than 5° before 4 March and after 10 October, limiting the period with suitable sun-photometer observations to the spring-summer-early autumn seasons (NILU contributes to a Lunar Arctic initiative to fill the gap in the wintertime AOD climatology by using Lunar photometer, see Appendix II). In 2017, sun-photometer observations started on 1 April and lasted until 4 October; reliable AOD values are available on 65 days. The AOD and Ångström coefficients time series of monthly means and standard deviation are shown in Figure 55, while the 2017 values on the background of the average data and their standard deviation from the whole 16-year period are shown in Figure 56. The 2017 monthly mean values and standard deviations for all years are given in Table 8. Data for all years are given in Appendix I.

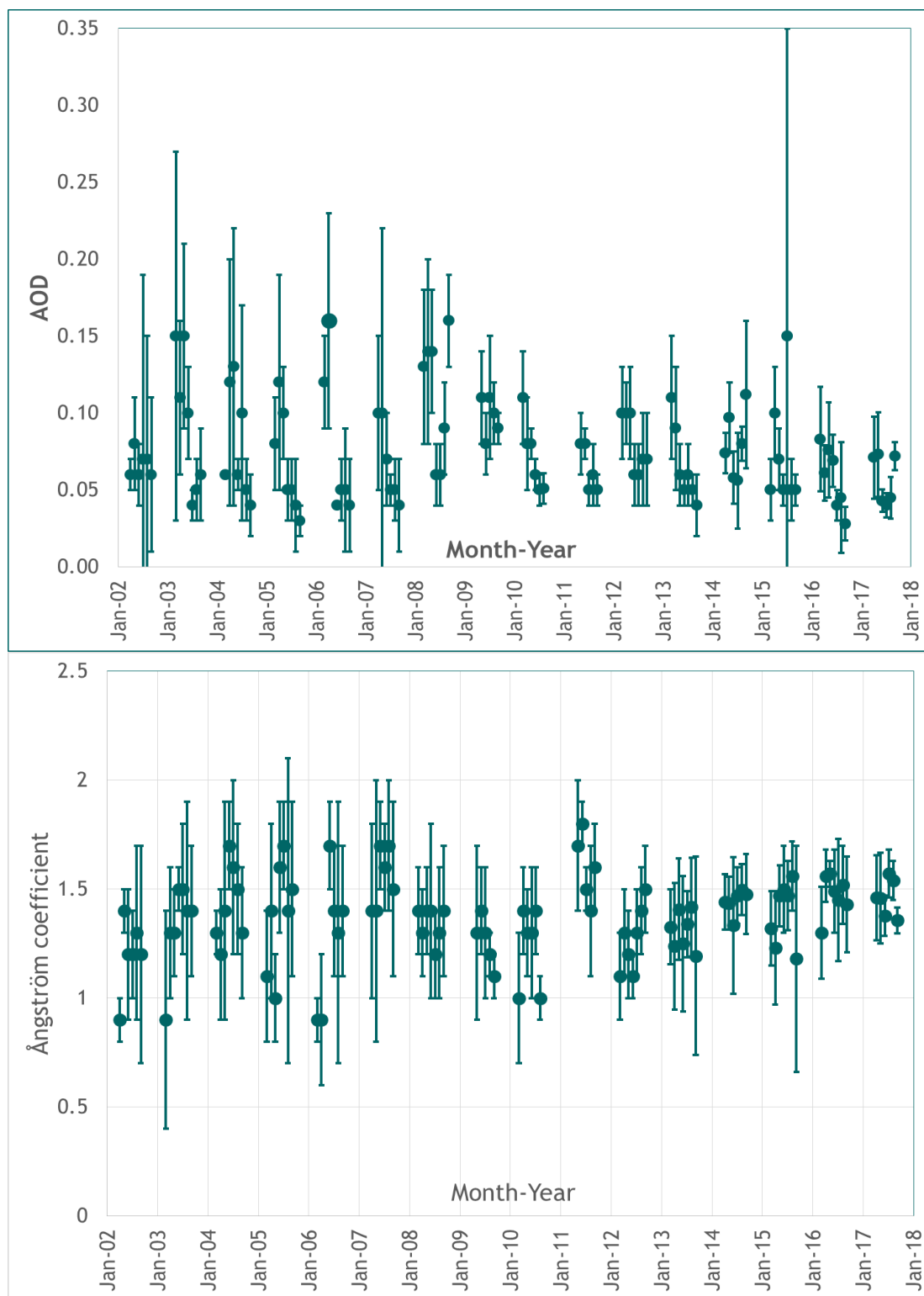


Figure 55: 2002 - 2017 time series of aerosol optical depth (AOD) at 500.5 nm wavelength in the atmospheric column above Ny-Ålesund (upper panel) and Ångström coefficient (lower panel). Monthly mean values and standard deviations are given.

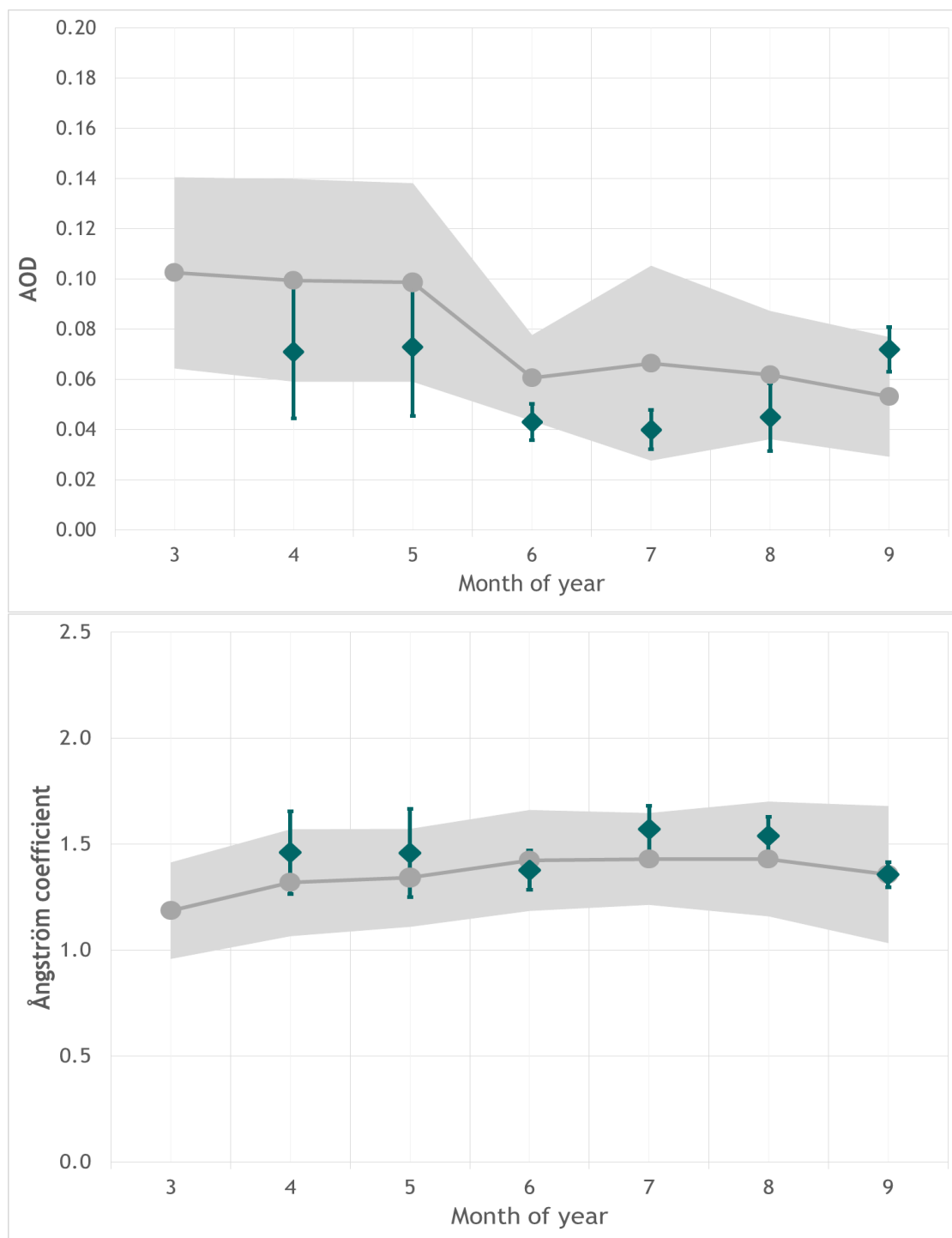


Figure 56: Seasonal variation of the aerosol optical depth (AOD) (upper panel) and Ångström coefficient (lower panel) observed in Ny-Ålesund. Values marked in grey are the mean and standard deviations for the time period 2009-2017; the 2017 monthly mean and standard deviations are shown in green.

In all months except September (with only three days of data), monthly mean AOD values were equal to or smaller than the long-term mean; in June and July the value was at the lower end of the standard deviation of all years. None of the daily average AOD values exceeded 0.2 and only 2 (one in April and one in May) exceeded 0.1. One should also note that the maximum monthly mean (0.073 in May) was the lowest in all the years of

observations so far. As in 2016, monthly mean Ångström coefficients were relatively stable throughout almost the whole measurement period and close to a value of 1.5 (slightly below 1.5 in April - June, slightly above in July/August). In September, the Ångström coefficient dropped to 1.36. Generally, in Ny-Ålesund seasonal variation of the Ångström coefficient is very weak, and the monthly mean Ångström coefficients in 2017 lie within the standard deviation range of the long-term monthly means.

Table 8: Monthly mean values for 2017 and mean for the time period 2002-2017 for March to September, plus standard deviations, for aerosol optical depth (AOD) and Ångström coefficient observed in Ny-Ålesund. In addition, the number of days with cloud free and quality assured observations are given.

Month/Year	Mar	Apr	May	Jun	Jul	Aug	Sep
Aerosol optical depth (AOD)							
2017		0.07 ±0.03	0.07 ±0.03	0.04 ±0.01	0.04 ±0.01	0.05 ±0.01	0.07 ±0.01
Mean 2002-2017	0.10 ±0.04	0.10 ±0.04	0.10 ±0.04	0.06 ±0.02	0.07 ±0.04	0.06 ±0.03	0.05 ±0.02
Ångström coefficient (Å)							
2017		1.46 ±0.20	1.46 ±0.21	1.38 ±0.09	1.57 ±0.11	1.54 ±0.09	1.36 ±0.06
Mean 2002-2017	1.19 ±0.23	1.32 ±0.25	1.34 ±0.23	1.42 ±0.24	1.43 ±0.22	1.43 ±0.27	1.36 ±0.32
Number of days with cloud-free and quality assured observations							
2017		13	19	11	12	6	3
Total 2002-2017	67	187	180	163	191	141	121

Key findings column-integrated optical aerosol properties Ny-Ålesund: In 2017, Ny-Ålesund monthly mean AOD values in all months were equal to or smaller than the long-term mean. The maximum monthly mean in 2017 was smaller than any year before since start of the measurements in 2002.

4 References

- Aas, W., Fiebig, M., Solberg, S., Yttri, K. E. (2018) Monitoring of long-range transported air pollutants in Norway, annual Report 2017. Kjeller, NILU (Miljødirektoratet rapport, M-1064/2018) (NILU report, 10/2018)..
- Asmi, A., Collaud Coen, M., Ogren, J. A., Andrews, E., Sheridan, P., Jefferson, A., Weingartner, E., Baltensperger, U., Bukowiecki, N., Lihavainen, H., Kivekäs, N., Asmi, E., Aalto, P. P., Kulmala, M., Wiedensohler, A., Birmili, W., Hamed, A., O'Dowd, C., G Jennings, S., Weller, R., Flentje, H., Fjaeraa, A. M., Fiebig, M., Myhre, C.E.L., Hallar, A. G., Swietlicki, E., Kristensson, A., Laj, P. (2013) Aerosol decadal trends - Part 2: In-situ aerosol particle number concentrations at GAW and ACTRIS stations. *Atmos. Chem. Phys.*, 13, 895-916. doi:10.5194/acp-13-895-2013.
- Baasandorj, M., Hall, B. D., Burkholder, J. B. (2012) Rate coefficients for the reaction of O(¹D) with the atmospherically long-lived greenhouse gases NF₃, SF₅CF₃, CHF₃, C₂F₆, c-C₄F₈, n-C₅F₁₂, and n-C₆F₁₄. *Atmos. Chem. Phys.*, 12, 11753-11764. doi:10.5194/acp-12-11753-2012.
- Coen, M. C., Andrews, E., Asmi, A., Baltensperger, U., Bukowiecki, N., Day, D., Fiebig, M., Fjaeraa, A. M., Flentje, H., Hyvärinen, A., Jefferson, A., Jennings, S. G., Kouvarakis, G., Lihavainen, H., Myhre, C. L., Malm, W. C., Mihapopoulos, N., Molenaar, J. V., O'Dowd, C., Ogren, J. A., Schichtel, B. A., Sheridan, P., Virkkula, A., Weingartner, E., Weller, R., Laj, P. (2013) Aerosol decadal trends - Part 1: In-situ optical measurements at GAW and IMPROVE stations. *Atmos. Chem. Phys.*, 13, 869-894. doi:10.5194/acp-13-869-2013.
- Coen, M.C., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S. (2010) Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms. *Atmos. Meas. Tech.*, 3, 457-474. doi: 10.5194/amt-3-457-2010.
- Dalsøren, S. B., Myhre, C. L., Myhre, G., Gomez-Pelaez, A. J., Søvde, O. A., Isaksen, I. S. A., Weiss, R. F., Harth, C. M. (2016) Atmospheric methane evolution the last 40 years. *Atmos. Chem. Phys.*, 16, 3099-3126. doi:10.5194/acp-16-3099-2016.
- Dalsøren, S. D., Myhre, G., Hodnebrog, Ø., Myhre, C. L., Stohl, A., Pizzo, I., Schwietzke, S., Höglund-Isaksson, L., Helmig, D., Reimann, S., Sauvage, S., Schmidbauer, N., Read, K. A., Carpenter, L. J., Lewis, A. C., Punjabi, S., Wallasch, M. (2018) Discrepancy between simulated and observed ethane and propane levels explained by underestimated fossil emissions. *Nature Geosci.*, 11, 178-184. doi:10.1038/s41561-018-0073-0.
- Delene, D.J., Ogren, J.A. (2002) Variability of aerosol optical properties at four North American surface monitoring sites. *J. Atmos. Sci.*, 59, 1135-1150.
- Dlugokencky, E.J., Hall, B. D., Montzka, S. A., Dutton, G., Mühle, J., Elkins, J. W. (2018) Long-lived greenhouse gases [in "State of the Climate in 2017"]. *Bull. Amer. Meteor. Soc.*, 99, S46-49. doi:10.1175/2018BAMSStateoftheClimate.1.
- Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A., Hansen, A. D. A. (2015) The "dual-spot" Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation. *Atmos. Meas. Tech.*, 8, 1965-1979. doi:10.5194/amt-8-1965-2015.

- Etiopio, G., Ciccioli, P. (2009) Earth's Degassing: A Missing Ethane and Propane Source. *Science* 323, 478. doi:10.1126/science.1165904.
- Etminan, M., Myhre, G., Highwood, E. J., Shine, K. P. (2016) Radiative forcing of carbon dioxide, methane, and nitrous oxide: A significant revision of the methane radiative forcing, *Geophys. Res. Lett.*, 43, 12,614-12,623, doi:10.1002/2016GL071930.
- Fiebig, M., Hirdman, D., Lunder, C. R., Ogren, J. A., Solberg, S., Stohl, A., Thompson, R. L. (2014) Annual cycle of Antarctic baseline aerosol: controlled by photooxidation-limited aerosol formation. *Atmos. Chem. Phys.*, 14, 3083-3093. doi: 10.5194/acp-14-3083-2014
- Fisher, R. E., France, J. L., Lowry, D., Lanoisellé, M., Brownlow, R., Pyle, J. A., Cain, M., Warwick, N., Skiba, U. M., Drewer, J., Dinsmore, K. J., Leeson, S. R., Bauguitte, S. J.-B., Wellpott, A., O'Shea, S. J., Allen, G., Gallagher, M. W., Pitt, J., Percival, C. J., Bower, K., George, C., Hayman, G. D., Aalto, T., Lohila, A., Aurela, M., Laurila, T., Crill, P. M., McCalley, C. K. and Nisbet, E. G. (2017) Measurement of the ^{13}C isotopic signature of methane emissions from northern European wetlands. *Global Biogeochem. Cycles*, 31, 605-623, doi:10.1002/2016GB005504.
- France, J. L., Cain, M., Fisher, R. E., Lowry, D., Allen, G., O'Shea, S. J., Illingworth, S., Pyle, J., Warwick, N., Jones, B. T., Gallagher, M. W., Bower, K., Le Breton, M., Percival, C., Muller, J., Wellpott, A., Bauguitte, S., George, C., Hayman, G. D., Manning, A. J., Myhre, C. L., Lanoisellé, M., Nisbet, E. G. (2016) Measurements of $\delta^{13}\text{C}$ in CH_4 and using particle dispersion modeling to characterize sources of Arctic methane within an air mass. *J. Geophys. Res. Atmos.*, 121, 14,257-14,270, doi:10.1002/2016JD026006.
- IPCC (2013) Summary for policymakers. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Ed. by Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. Cambridge, Cambridge University Press. pp. 3-29.
- Hall, B. D., Montzka, S. A., Dutton, G., Mühle, J., Elkins, J. W. (2017) Long-lived greenhouse gases [in "State of the Climate in 2016"]. *Bull. Amer. Meteor. Soc.*, 98 (8), S43-S46, doi:10.1175/2017BAMSStateoftheClimate.1.
- Hartmann, D.L., Klein Tank, A. M. G., Rusticucci, M., Alexander, L.V., Brönnimann, S., Charabi, Y., Dentener, F. J., Dlugokencky, E. J., Easterling, D. R., Kaplan, A., Soden, B. J., Thorne, P. W., Wild, M., Zhai, P. M. (2013) Observations: atmosphere and surface. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Ed. by Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. Cambridge, Cambridge University Press. pp. 159-254.
- Heintzenberg, J. (1980) Particle size distribution and optical properties of Arctic haze. *Tellus*, 32, 251-260. doi: 10.3402/tellusa.v32i3.10580
- Helmig, D., Rossabi, S., Hueber, J., Tans, P., Montzka, S. A., Masarie, K., Thoning, K., Plass-Duelmer, C., Claude, A., Carpenter, L. J., Lewis, A. C., Punjabi, S., Reimann, S., Vollmer, M. K., Steinbrecher, R., Hannigan, J. W., Emmons, L. K., Mahieu, E., Franco, B., Smale, D., Pozzer, A. (2016) Reversal of global atmospheric ethane and propane trends largely due to US oil and natural gas production. *Nature Geosci.*, 9, 490-495. doi:10.1038/ngeo2721.

- Hewitt, C. Nicholas (ed.) (1999) *Reactive Hydrocarbons in the Atmosphere*. San Diego, CA, Academic Press. p. 313.
- Hossaini, R., Chipperfield, M. P., Montzka, S. A., Leeson, A. A., Dhomse, S. and Pyle, J. A (2017) The increasing threat to stratospheric ozone from dichloromethane. *Nat. Commun.*, *8*, 15962, doi:10.1038/ncomms15962.
- Isaksen, I. S. A., Hov, Ø. (1987) Calculation of trends in the tropospheric concentration of O₃, OH, CO, CH₄ and NO_x. *Tellus*, *39B*, 271-285.
- Montzka, S. A., Dutton, G. S., Yu, P., Ray, E., Portmann, R. W., Daniel, J. S., Kuijpers, L., Hall, B. D., Mondeel, D., Siso, Nance, J. D., Rigby, M., Manning, A. J., Hu, L., Moore, F., Miller, B. R., Elkins, J. W. (2018) An unexpected and persistent increase in global emissions of ozone-depleting CFC-11. *Nature*, *557*, 413-417.
- Myhre, C. L., Hermansen, O., Fiebig, M., Lunder, C., Fjæraa, A. M., Svendby, T., Platt, M., Hansen, G., Schmidbauer, N., Krognes, T. (2016) Monitoring of greenhouse gases and aerosols at Svalbard and Birkenes in 2015 - Annual report. Kjeller, NILU (Miljødirektoratet rapport, M-694/2016) (NILU report, 31/2016).
- Myhre, C. L., Ferré, B., Platt, S. M., Silyakova, A., Hermansen, O., Allen, G., Pisso, I., Schmidbauer, N., Stohl, A., Pitt, J., Jansson, P., Greinert, J., Percival, C., Fjæraa, A. M., O'Shea, S., Gallagher, M., Le Breton, M., Bower, K., Bauguitte, S., Dalsøren, S., Vadakkepuliambatta, S., Fisher, R., Nisbet, E., Lowry, D., Myhre, G., Pyle, J., Cain, M., Mienert, J. (2016) Large methane release from the Arctic seabed west of Svalbard, but small release to the atmosphere. *Geophys. Res. Lett.*, *43*, 4624-4631. doi:10.1002/2016GL068999.
- Myhre, G., Myhre C. L., Forster, P. M., Shine, K. P. (2017) Halfway to doubling of CO₂ radiative forcing. *Nature Geosci.*, *10*, 710-711. doi:10.1038/ngeo3036.
- Myhre, G., Myhre, C. L., Samset, B. H., Storelvmo, T. (2013a) Aerosols and their relation to global climate and climate sensitivity. *Nature Education Knowledge*, *4*, 5,7. URL: <http://www.nature.com/scitable/knowledge/library/aerosols-and-their-relation-to-global-climate-102215345>
- Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., Zhang, H. (2013b) Anthropogenic and natural radiative forcing. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Ed. by Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. Cambridge, Cambridge University Press. pp. 659-740.
- Ng, N. L., Kroll, J.H., Chan, A. W. H., Chhabra, P. S., Flagan, R. C., Seinfeld, J. H. (2007) Secondary organic aerosol formation from m-xylene, toluene, and benzene. *Atmos. Chem. Physics*, *7*, 3909-3922.
- Nicewonger, M. R., Verhulst, K. R., Aydin, M., Saltzman, E. S. (2016) Preindustrial atmospheric ethane levels inferred from polar ice cores: A constraint on the geologic sources of atmospheric ethane and methane. *Geophys. Res. Lett.*, *43*, 214-221. doi:10.1002/2015GL066854.
- Oram, D. E., Mani, F. S., Laube, J. C., Newland, M. J., Reeves, C. E., Sturges, W. T., Penkett, S. A., Brenninkmeijer, C. A. M., Röckmann, T., and Fraser, P. J. (2012) Long-term

- tropospheric trend of octafluorocyclobutane (c-C₄F₈ or PFC-318), *Atmos. Chem. Phys.*, **12**, 261-269. doi:10.5194/acp-12-261-2012.
- Platt, S. M., Eckhardt, S., Ferré, B., Fisher, R. E., Hermansen, O., Jansson, P., Lowry, D., Nisbet, E. G., Pisso, I., Schmidbauer, N., Silyakova, A., Stohl, A., Svendby, T. M., Vadakkepuliambatta, S., Mienert, J., and Lund Myhre, C. (2018) Methane at Svalbard and over the European Arctic Ocean. *Atmos. Chem. Phys. Discuss.*, *In press*. doi:10.5194/acp-2018-597.
- Prather, M., Ehhalt, D., Dentener, F., Derwent, R. G., Dlugokencky, E., Holland, E., Isaksen, I. S. A., Katima, J., Kirchhoff, V., Matson, P., Midgley, P. M., Wang, M. (2001) Atmospheric chemistry and greenhouse gases. In: *Climate Change 2001: The Scientific Basis, Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*. Ed. by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., Johnson, C. A. Cambridge, Cambridge University Press. pp. 239-287.
- Pisso, I., Myhre, C. L., Platt, S. M., Eckhardt, S., Hermansen, O., Schmidbauer, N., Mienert, J., Vadakkepuliambatta, S., Bauguitte, S., Pitt, J., Allen, G., Bower, K.N., O'Shea, S., Gallagher, M. W., Percival, C.J., Pyle, J., Cain, M., Stohl, A. (2016) Constraints on oceanic methane emissions west of Svalbard from atmospheric in situ measurements and Lagrangian transport modelling. *J. Geophys. Res.*, **121**, 14188-14200. doi:10.1002/2016JD025590.
- R Core Team (2018) R: A language and environment for statistical computing. Vienna, Austria, R Foundation for Statistical Computing. URL: <https://www.R-project.org/>.
- Repo, M. E., Susiluoto, S., Lind, S. E., Jokinen, S., Elsakov, V., Biasi, C., Virtanen, T., Pertti, J., Martikainen, P. J. (2009) Large N₂O emissions from cryoturbated peat soil in tundra. *Nature Geosci.*, **2**, 189-192.
- Simmonds, P. G., Manning, A. J., Cunnold, D. M., Fraser, P. J., McCulloch, A., O'Doherty, S., Krummel, P. B., Wang, R. H. J., Porter, L. W., Derwent, R. G., Grealley, B., Salameh, P., Miller, B. R., Prinn, R. G., Weiss, R. F. (2006) Observations of dichloromethane, trichloroethene and tetrachloroethene from the AGAGE stations at Cape Grim, Tasmania, and Mace Head, Ireland. *J. Geophys. Res.*, **111**, D18304. doi:10.1029/2006JD007082.
- Svendby, T.M., Hansen, G.H., Stebel, K., Bäcklund, A., Dahlback, A. (2018) Monitoring of the atmospheric ozone layer and natural ultraviolet radiation. Annual report 2017. (Miljødirektoratet rapport, M-1089/2018) (NILU report 23/2018).
- Thompson, R. L., Sasakawa, M., Machida, T., Aalto, T., Worthy, D., Lavric, J. V., Myhre, C. L., Stohl, A. (2017) Methane fluxes in the high northern latitudes for 2005-2013 estimated using a Bayesian atmospheric inversion. *Atmos. Chem. Phys.*, **17**, 3553-3572. doi:10.5194/acp-2016-660.
- Thompson, R. L., Dlugokencky, E., Chevallier, F., Ciais, P., Dutton, G., Elkins, J. W., Langenfelds, R. L., Prinn, R. G., Weiss, R. F., Tohjima, Y., Krummel, P. B., Fraser, P., Steele, L. P. (2013) Interannual variability in tropospheric nitrous oxide. *Geophys. Res. Lett.*, **40**, 4426-4431. doi:10.1002/grl.50721.
- Tørseth, K., Aas, W., Breivik, K., Fjæraa, A. M., Fiebig, M., Hjellbrekke, A. G., Myhre, C. L., Solberg, S., Yttri, K. E. (2012) Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972-2000. *Atmos. Chem. Phys.*, **12**, 5447-5481. doi:10.5194/acp-12-5447-2012.

- Umezawa, T., Baker, A. K., Oram, D., Sauvage, C., O'Sullivan, D., Rauthe-Schöch, A., Montzka, S. A., Zahn, A., Brenninkmeijer, C. A. M. (2014) Chloromethane in the upper troposphere observed by the CARIBIC passenger aircraft observatory: Large-scale distributions and Asian summer monsoon outflow. *J. Geophys. Res. Atmos.*, *119*, 5542-5558, doi:10.1002/2013JD021396.
- Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjåraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J. A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Grüning, C., Faloon, F., Beddows, D., Harrison, R., Monahan, C., Jennings, S. G., O'Dowd, C. D., Marinoni, A., Horn, H.-G., Keck, L., Jiang, J., Scheckman, J., McMurry, P.H., Deng, Z., Zhao, C. S., Moerman, M., Henzing, B., de Leeuw, G., Löschau, G., Bastian, S. (2012) Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions. *Atmos. Meas. Tech.*, *5*, 657 - 685. doi:10.5194/amt-5-657-2012.
- WMO (2017) Greenhouse Gas Bulletin. The state of greenhouse gases in the atmosphere using global observations through 2016. Geneva, World Meteorological Organization (GHG Bulletin No. 13, 30 October 2017). URL: https://library.wmo.int/opac/doc_num.php?explnum_id=4022
- WMO (2016) Greenhouse Gas Bulletin. The state of greenhouse gases in the atmosphere using global observations through 2015. Geneva, World Meteorological Organization (GHG Bulletin No. 12, 24 October 2016). URL: http://library.wmo.int/pmb_ged/ghg-bulletin_12_en.pdf
- WMO (2015) Greenhouse Gas Bulletin. The state of greenhouse gases in the atmosphere using global observations through 2014. Geneva, World Meteorological Organization (GHG Bulletin No. 11, 9 November 2015). URL: http://library.wmo.int/pmb_ged/ghg-bulletin_11_en.pdf
- WMO (2014) Greenhouse Gas Bulletin. The state of greenhouse gases in the atmosphere using global observations through 2013. Geneva, World Meteorological Organization (GHG Bulletin No. 10, 9 September 2014). URL: http://library.wmo.int/opac/index.php?lvl=notice_display&id=16396#.VGHPz5-gQtK
- WMO (2014b) Scientific assessment of ozone depletion: 2014. Geneva, World Meteorological Organization (Global ozone research and monitoring project, Report No. 55). URL: <https://www.esrl.noaa.gov/csd/assessments/ozone/2014/chapters/2014OzoneAssessment.pdf>
- WMO (2013) Greenhouse Gas Bulletin. The state of greenhouse gases in the atmosphere using global observations through 2012. Geneva, World Meteorological Organization (GHG Bulletin No. 9, 6 November 2013). URL: http://www.wmo.int/pages/prog/arep/gaw/ghg/documents/GHG_Bulletin_No.9_en.pdf
- WMO (2012) Greenhouse Gas Bulletin. The state of greenhouse gases in the atmosphere using global observations through 2011. Geneva, World Meteorological Organization (GHG Bulletin No. 8, 19 November 2012). URL: http://www.wmo.int/pages/prog/arep/gaw/ghg/documents/GHG_Bulletin_No.8_en.pdf

WMO (2011) Scientific assessment of ozone depletion: 2010. Geneva, World Meteorological Organization (Global ozone research and monitoring project, Report No. 52).

Xu, Y., Zaelke, D., Velders, G.J.M., Ramanathan, V. (2013) The role of HFCs in mitigating 21st century climate change. *Atmos. Chem. Phys.*, 13, 6083-6089. doi:10.5194/acp-13-6083-2013.

Zeileis A. (2004) Econometric Computing with HC and HAC Covariance Matrix Estimators. *J. Stat. Software*, 11, 1-17. doi: 10.18637/jss.v011.i10.

Zeileis, A. (2006) Object-oriented computation of sandwich estimators. *J. Stat. Software*, 16, 1-16. doi:[10.18637/jss.v016.i09](https://doi.org/10.18637/jss.v016.i09).

APPENDIX I: Data Tables

Table A 1: Annual mean concentration for all greenhouse gases included in the programme at Zeppelin and Birkenes. All concentrations are mixing ratios in ppt, except for methane and carbon monoxide (ppb) and carbon dioxide (ppm). The annual means are based on a combination of the measurements and the fitted values; during periods with lacking observations we have used the fitted mixing ratios in the calculation of the annual mean. All underlying measurement data are open and accessible and can be downloaded directly from the database: <http://ebas.nilu.no/>

Component	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
Carbon dioxide - Zeppelin											394.8	397.4	399.6	401.5	404.4	408.0	
Carbon dioxide - Birkenes									391.9	393.9	396.6	397.9	400.7	402.8	405.2	409.8	411.3
Methane - Zeppelin	1843.5	1842.5	1855.4	1853.6	1852.6	1853.3	1863.6	1873.5	1888.2	1881.0	1879.6	1891.9	1898.0	1910.0	1920.3	1932.1	1938.9
Methane - Birkenes									1882.4	1887.3	1895.7	1900.5	1902.5	1917.3	1926.2	1941.9	1945.3
Carbon monoxide	124.9	126.1	140.5	131.4	128.8	126.8	120.2	120.3	117.9	128.2	115.5	120.5	112.9	113.4	113.2	112.9	114.3
Nitrous oxide											323.7	324.2	325.0	326.1	327.2	328.1	329.0
Chlorofluorocarbons																	
CFC-11	259.0	257.2	255.2	253.5	251.3	249.4	246.7	244.7	242.7	240.7	238.4	236.8	235.1	233.9	232.3	231.3	230.2
CFC-12	547.0	547.5	547.8	545.8	547.0	546.3	542.4	541.5	537.6	534.4	531.5	528.4	525.2	522.8	518.6	516.0	512.5
CFC-113	81.5	80.8	80.0	79.4	78.8	77.9	77.4	76.8	76.2	75.5	74.6	74.1	73.5	73.0	72.3	71.7	71.1
CFC-115	8.21	8.18	8.22	8.28	8.41	8.39	8.38	8.40	8.43	8.42	8.41	8.44	8.43	8.46	8.50	8.53	8.59
Hydrochlorofluorocarbons																	
HCFC-22	159.1	164.3	170.5	175.5	181.2	188.9	196.2	204.5	212.5	219.8	225.9	231.4	236.5	241.3	245.4	248.5	252.0
HCFC-141b	16.8	17.9	18.7	19.3	19.5	19.9	20.5	21.2	21.6	22.2	23.1	24.0	24.7	25.4	25.6	26.0	25.9
HCFC-142b	14.5	15.1	15.9	16.5	17.2	18.2	19.3	20.3	21.3	22.0	22.7	23.1	23.3	23.4	23.5	23.5	23.4
Hydrofluorocarbons																	
HFC-125	2.2	2.6	2.9	3.3	4.0	4.9	5.8	6.9	8.0	9.6	11.8	13.5	15.6	17.9	20.3	22.9	25.9
HFC-134a	20.9	26.0	30.8	35.6	40.0	44.2	48.7	53.5	57.8	63.5	68.5	73.5	78.9	84.4	90.1	96.6	103.2
HFC-152a	2.7	3.5	4.2	5.0	5.7	6.8	7.8	8.6	9.0	9.5	10.0	10.3	10.2	10.2	10.1	10.2	10.2

Component	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
HFC-23										23.6	24.7	25.6	26.7	27.7	28.8	29.6	30.7
HFC-365mfc										0.74	0.79	0.87	0.92	1.02	1.10	1.19	1.24
HFC-227ea										0.71	0.79	0.88	0.99	1.10	1.21	1.34	1.47
HFC-236fa										0.09	0.10	0.11	0.13	0.14	0.15	0.16	0.17
HFC-245fa										1.58	1.80	1.99	2.20	2.39	2.57	2.79	3.04
HFC-32										5.20	6.56	7.74	9.29	10.92	12.77	15.11	18.26
HFC-4310mee										0.21	0.22	0.24	0.25	0.26	0.27	0.28	0.28
HFC-143a										11.88	13.18	14.57	16.03	17.62	19.08	20.73	22.52
Perfluorinated compounds																	
PFC-14															82.43	83.28	84.26
PFC-116										4.12	4.20	4.27	4.37	4.45	4.55	4.64	4.74
PFC-218										0.56	0.57	0.59	0.60	0.61	0.63	0.64	0.66
PFC-318										1.27	1.33	1.38	1.43	1.47	1.53	1.59	1.66
Sulphurhexafluoride	4.95	5.14	5.37	5.61	5.82	6.09	6.31	6.64	6.93	7.19	7.49	7.78	8.11	8.43	8.75	9.09	9.46
Nitrogen trifluoride																1.61	1.76
Sulfuryl fluoride										1.72	1.80	1.89	2.01	2.11	2.21	2.33	2.46
Halons																	
H-1211	4.39	4.43	4.47	4.52	4.52	4.48	4.43	4.40	4.33	4.26	4.18	4.09	3.97	3.87	3.77	3.65	3.55
H-1301	2.99	3.07	3.14	3.17	3.22	3.23	3.24	3.28	3.29	3.32	3.32	3.35	3.36	3.37	3.39	3.39	3.39
H-2402										0.47	0.46	0.45	0.44	0.43	0.43	0.42	0.41
Other halocarbons																	
Chloromethane	505.5	521.0	527.0	523.4	519.8	521.1	523.5	525.0	526.2	520.4	509.5	514.2	519.1	514.2	513.3	522.8	517.3
Bromomethane	9.04	9.11	8.89	8.89	8.56	8.59	8.33	7.78	7.37	7.28	7.19	7.03	6.95	6.82	6.62	6.69	6.54
Dichloromethane	31.38	31.45	32.57	32.41	32.06	33.44	35.51	37.59	38.53	42.00	42.02	45.06	53.66	54.34	54.12	56.75	61.40
Trichloromethane	10.95	10.65	10.83	10.47	10.53	10.48	10.68	10.44	10.84	11.44	11.96	12.08	12.70	13.45	13.77	14.42	15.39

Component	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
Carbon tetrachloride										86.78	85.26	84.42	83.49	82.60	81.87	80.66	79.53
Trichloroethane	38.07	32.04	27.26	22.84	19.13	15.93	13.31	11.05	9.23	7.75	6.48	5.40	4.52	3.80	3.32	2.80	2.33
Trichloroethene	0.75	0.67	0.56	0.51	0.49	0.47	0.33	0.37	0.54	0.54	0.55	0.50	0.56	0.49	0.44	0.41	0.32
Tetrachloroethene	4.70	4.22	4.06	3.87	3.37	2.90	3.12	2.71	2.97	3.13	2.83	2.68	2.55	2.58	2.60	2.56	2.48
Volatile Organic Compounds (VOC)																	
Ethane										1433.4	1470.4	1583.6	1571.2	1651.5	1651.3	1583.4	1573.8
Propane										513.9	529.6	560.1	577.5	568.4	546.1	569.8	584.0
Butane										180.2	187.6	196.2	203.9	193.0	183.6	169.8	193.0
Pentane										58.8	61.6	62.0	67.6	64.3	63.0	58.9	60.8
Benzene										84.9	72.7	74.5	69.5	71.4	68.2	66.7	59.1
Toluene										34.3	29.3	28.5	26.5	28.6	26.1	25.5	17.8

Table A 2: All calculated trends per year, error and regression coefficient for the fit. The trends are all in ppt per year, except for CH₄, N₂O, and CO which are in ppb and CO₂ is in ppm. The negative trends are in blue, and the positive trends are shown in red. Generally the period is from 2001 to 2017, but for some compounds the measurements started after September 2010. For these compounds the trends are in general more uncertain.

Component	Formula	Trend /yr	Error	R ²
Carbon dioxide - Zeppelin	CO ₂	2.63	0.22	0.85
Carbon dioxide - Birkenes		2.46	0.08	0.67
Methane - Zeppelin	CH ₄	5.89	0.13	0.86
Methane - Birkenes		8.17	0.34	0.56
Carbon monoxide	CO	-1.25	0.27	0.78
Nitrous oxide	N ₂ O	0.93	0.01	0.98
Chlorofluorocarbons				
CFC-11	CCl ₃ F	-1.89	0.010	0.99
CFC-12	CF ₂ Cl ₂	-2.36	0.033	0.96
CFC-113	CF ₂ ClCFCl ₂	-0.65	0.003	0.99
CFC-115	CF ₃ CF ₂ Cl	0.02	0.001	0.55
Hydrochlorofluorocarbons				
HCFC-22	CHClF ₂	6.21	0.032	0.997
HCFC-141b	C ₂ H ₃ FCl ₂	0.59	0.024	0.970
HCFC-142b	CH ₃ CF ₂ Cl	0.63	0.009	0.991
Hydrofluorocarbons				
HFC-125	CHF ₂ CF ₃	1.46	0.008	0.998
HFC-134a	CH ₂ FCF ₃	5.01	0.007	0.999
HFC-152a	CH ₃ CHF ₂	0.50	0.010	0.972
HFC-23	CHF ₃	1.00	0.005	0.997
HFC-365mfc	CH ₃ CF ₂ CH ₂ CF ₃	0.08	0.001	0.975
HFC-227ea	CF ₃ CHFCF ₃	0.11	0.000	0.997
HFC-236fa	CF ₃ CH ₂ CF ₃	0.01	0.000	0.976
HFC-245fa	CHF ₂ CH ₂ CF ₃	0.20	0.001	0.996
HFC-32	CH ₂ F ₂	1.80	0.009	0.998
HFC-4310mee	C ₅ H ₂ F ₁₀	0.01	0.000	0.922
HFC-143a	CH ₃ CF ₃	1.52	0.006	0.995
Perfluorinated compounds				
PFC-14	CF ₄	0.390	0.4596	0.987
PFC-116	C ₂ F ₆	0.088	0.0004	0.993
PFC-218	C ₃ F ₈	0.014	0.0001	0.960
PFC-318	c-C ₄ F ₈	0.054	0.0002	0.990
Sulphurhexafluoride	SF ₆	0.282	0.0005	0.998
Nitrogen trifluoride	NF ₃	-	-	-
Sulfuryl fluoride	SO ₂ F ₂	0.106	0.0009	0.992

Component	Formula	Trend /yr	Error	R ²
Halons				
H-1211	CBrClF ₂	-0.058	0.0004	0.992
H-1301	CBrF ₃	0.022	0.0004	0.762
H-2402	CBrF ₂ CBrF ₂	-0.008	0.0001	0.942
Halogenated compounds				
Chloromethane	CH ₃ Cl	-0.167	0.2346	0.875
Bromomethane	CH ₃ Br	-0.183	0.0067	0.885
Dichloromethane	CH ₂ Cl ₂	1.951	0.0593	0.936
Trichloromethane	CHCl ₃	0.274	0.0098	0.842
Carbon tetrachloride	CCl ₄	-0.978	0.0271	0.915
Trichloroethane	CH ₃ CCl ₃	-2.051	0.0089	0.999
Trichloroethene	CHClCCl ₂	-0.013	0.0041	0.392
Tetrachloroethene	CCl ₂ CCl ₂	-0.120	0.0088	0.474
Volatile Organic Compounds (VOC)				
Ethane*	C ₂ H ₆	22.56	6.33	0.88
Propane*	C ₃ H ₈	8.76	4.87	0.82
Butane*	C ₄ H ₁₀	-0.29	2.38	0.74
Pentane*	C ₅ H ₁₂	0.09	0.83	0.69
Benzene*	C ₆ H ₆	-2.68	0.66	0.85
Toluene*	C ₆ H ₅ CH ₃	-1.63	0.46	0.71

* The trend is very uncertain and can change dramatically if one or two years are omitted from the time series.

Table A 3: Monthly means and standard deviation of aerosol optical depth (AOD) at 500 nm at Ny-Ålesund.'

Month/Year	Mar	Apr	May	Jun	Jul	Aug	Sep
Aerosol optical depth (AOD)							
2002		0.06±0.01	0.08±0.03	0.06±0.02	0.07±0.12	0.07±0.08	0.06±0.05
2003	0.15±0.12	0.11±0.05	0.15±0.06	0.10±0.03	0.04±0.01	0.05±0.02	0.06±0.03
2004	0.06±0.00	0.12±0.08	0.13±0.09	0.06±0.01	0.10±0.07	0.05±0.02	0.04±0.02
2005	0.08±0.03	0.12±0.07	0.10±0.03	0.05±0.02	0.05±0.02	0.04±0.03	0.03±0.01
2006	0.12±0.03	0.16±0.07		0.04±0.00	0.05±0.02	0.05±0.04	0.04±0.03
2007		0.10±0.05	0.10±0.12	0.07±0.03	0.05±0.01	0.05±0.02	0.04±0.03
2008	0.13±0.05	0.14±0.06	0.14±0.04	0.06±0.02	0.06±0.02	0.09±0.03	0.16±0.03
2009			0.11±0.03	0.08±0.02	0.11±0.04	0.10±0.02	0.09±0.01
2010	0.11±0.03	0.08±0.03	0.08±0.01	0.06±0.01	0.05±0.01	0.05±0.01	
2011			0.08±0.02	0.08±0.01	0.05±0.01	0.06±0.02	0.05±0.01
2012	0.10±0.03	0.10±0.02	0.10±0.03	0.06±0.02	0.06±0.02	0.07±0.03	0.07±0.03
2013	0.11±0.04	0.09±0.04	0.06±0.02	0.05±0.01	0.06±0.02	0.05±0.01	0.04±0.02
2014		0.07±0.01	0.10±0.02	0.06±0.02	0.06±0.03	0.08±0.01	0.11±0.05
2015	0.05±0.02	0.10±0.03	0.07±0.02	0.05±0.01	0.15±0.20	0.05±0.02	0.05±0.01
2016	0.08±0.03	0.06±0.02	0.08±0.03	0.07±0.02	0.04±0.01	0.05±0.04	0.03±0.01
2017		0.07 ±0.03	0.07±0.03	0.04±0.01	0.04±0.01	0.05±0.01	0.07±0.01
Mean (2002-2017)	0.10±0.04	0.10±0.04	0.10±0.04	0.06±0.02	0.07±0.04	0.06±0.03	0.05±0.02

Table A 4: Monthly means and standard deviation of the Ångström coefficient (Å) at Ny-Ålesund. There are no observations of this parameter during the winter months due to polar night.

Month/Year	Mar	Apr	May	Jun	Jul	Aug	Sep
Ångström coefficient (Å)							
2002		0.9±0.1	1.4±0.1	1.2±0.3	1.2±0.2	1.3±0.4	1.2±0.5
2003	0.9±0.5	1.3±0.3	1.3±0.2	1.5±0.1	1.5±0.3	1.4±0.5	1.4±0.3
2004	1.3±0.1	1.2±0.3	1.4±0.5	1.7±0.2	1.6±0.4	1.5±0.3	1.3±0.3
2005	1.1±0.3	1.4±0.4	1.0±0.2	1.6±0.3	1.7±0.2	1.4±0.7	1.5±0.4
2006	0.9±0.1	0.9±0.3		1.7±0.2	1.4±0.3	1.3±0.6	1.4±0.3
2007		1.4±0.4	1.4±0.6	1.7±0.2	1.6±0.2	1.7±0.3	1.5±0.4
2008	1.4±0.2	1.3±0.2	1.4±0.2	1.4±0.4	1.2±0.2	1.3±0.3	1.4±0.3
2009			1.3±0.4	1.4±0.2	1.3±0.3	1.2±0.1	1.1±0.1
2010	1.0±0.3	1.4±0.2	1.3±0.2	1.3±0.3	1.4±0.2	1.0±0.1	
2011			1.7±0.3	1.8±0.1	1.5±0.1	1.4 ±0.3	1.6±0.2
2012	1.1±0.2	1.3±0.2	1.2±0.2	1.1±0.1	1.3±0.2	1.4±0.2	1.5±0.2
2013	1.3±0.2	1.2±0.3	1.4±0.2	1.6±0.3	1.3±0.2	1.4±0.2	1.2±0.5
2014		1.4±0.1	1.4±0.1	1.3±0.3	1.5±0.1	1.5±0.1	1.5±0.2
2015	1.32±0.17	1.23±0.26	1.47±0.14	1.50±0.20	1.47±0.16	1.56±0.16	1.18±0.52
2016	1.30±0.21	1.56±0.12	1.57±0.06	1.49±0.19	1.45±0.28	1.52±0.18	1.43±0.22
2017		1.46±0.20	1.46±0.21	1.38±0.09	1.57±0.11	1.54±0.09	1.36±0.06
Mean (2002 -2017)	1.19±0.23	1.32±0.25	1.34±0.23	1.42±0.24	1.43±0.22	1.43±0.27	1.36±0.32

Table A 5: Number of days with AOD observations at Ny-Ålesund made within the months.

Month/Year	Mar	Apr	May	Jun	Jul	Aug	Sep
Number of days with cloud-free and quality assured observations							
2002		4	15	11	6	9	14
2003	3	12	16	8	15	17	12
2004	2	8	13	9	5	12	12
2005	12	17	24	15	10		11
2006	6	12		5	12	4	13
2007		16	9	12	17	10	9
2008	15	12	14	20	16	13	2
2009			7	10	17	8	8
2010	7	18	7	10	12	3	1
2011			2	2	7	4	6
2012	6	18	12	15	16	11	4
2013	5	13	10	10	8	7	9
2014		13	9	9	9	14	4
2015	5	17	15	9	17	13	6
2016	6	14	8	7	12	10	7
2017		13	19	11	12	6	3
Total (2002-2017)	67	187	180	163	191	141	121

Table A 6: Monthly means and standard deviation of aerosol optical depth (AOD) at 500 nm at Birkenes.

Month/Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
Aerosol optical depth (AOD)											
2009				0.29 ±0.00	0.09 ±0.05	0.09 ±0.05	0.18 ±0.06	0.17 ±0.07	0.10 ±0.04	0.08 ±0.03	
2010					0.10 ±0.04	0.09 ±0.04	0.10 ±0.07	0.10 ±0.05	0.05 ±0.02	0.07 ±0.03	0.04 ±0.01
2011	0.02 ±0.01	0.03 ±0.01	0.07 ±0.02	0.21 ±0.19	0.13 ±0.07	0.10 ±0.04	0.13 ±0.06	0.09 ±0.05			
2012			0.07 ±0.05	0.05 ±0.02	0.08 ±0.04	0.09 ±0.04	0.07 ±0.03	0.08 ±0.03	0.07 ±0.01	0.06 ±0.03	0.04 ±0.00
2013							0.17 ±0.17	0.12 ±0.09	0.05 ±0.03	0.05 ±0.03	
2014			0.15 ±0.14	0.11 ±0.06	0.10 ±0.03	0.08 ±0.03	0.13 ±0.06	0.15 ±0.07	0.14 ±0.06		
2015			0.04 ±0.02	0.07 ±0.02	0.07 ±0.03	0.06 ±0.02	0.10 ±0.07	0.09 ±0.06	0.04 ±0.02	0.03 ±0.03	0.04 ±0.01
2016	0.01 ±0.00	0.03 ±0.01	0.03 ±0.02	0.05 ±0.03							0.02 ±0.00*
2017*				0.08 ±0.01	0.06 ±0.03	0.04 ±0.03	0.06 ±0.03	0.09 ±0.07	0.09 ±0.03	0.06 ±0.03	
Mean 2009-2017	0.02 0.01	0.03 ±0.01	0.07 ±0.05	0.10 ±0.07	0.09 ±0.04	0.08 ±0.04	0.12 ±0.07	0.11 ±0.06	0.08 ±0.03	0.06 ±0.03	0.013 ±0.01

* version 3 data analysis (Aeronet)

Table A 7: Monthly means and standard deviation of the Ångström coefficient (Å) at Birkenes

Month/Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
Ångström coefficient (Å)											
2009				1.5 ±0.0	1.2 ±0.3	1.4 ±0.3	1.4 ±0.4	1.1 ±0.2	1.0 ±0.2	1.1 ±0.2	
2010					1.3 ±0.3	1.4 ±0.3	1.4 ±0.2	1.4 ±0.2	1.3 ±0.3	1.3 ±0.3	1.3 ±0.23
2011	1.0 ±0.2	1.0 ±0.1	1.0 ±0.3	1.2 ±0.5	1.3 ±0.3	1.5 ±0.3	1.6 ±0.3	1.6 ±0.1			
2012			1.1 ±0.4	1.6 ±0.3	1.4 ±0.4	1.7 ±0.1	1.6 ±0.3	1.5 ±0.3	1.1 ±0.3	1.4 ±0.4	0.8 ±0.3
2013							1.3 ±0.2	1.2 ±0.3	0.8 ±0.2	0.8 ±0.3	
2014			0.87 ±0.48	1.04 ±0.33	1.07 ±0.27	1.02 ±0.24	1.38 ±0.33	1.14 ±0.25	1.19 ±0.16		
2015			0.93 ±0.16	1.06 ±0.13	1.11 ±0.20	1.30 ±0.20	1.49 ±0.20	1.37 ±0.26	1.30 ±0.23	1.23 ±0.25	0.84 ±0.33
2016	0.68 ±0.07	1.00 ±0.2	0.90 ±0.3	1.13 ±0.2							1.11 ±0.08*
2017*				1.77 ±0.07	1.39 ±0.35	1.39 ±0.37	1.73 ±0.34	1.48 ±0.37	0.93 ±0.18	0.78 ±0.20	
Mean 2009-2017	0.82 ±0.11	1.02 ±0.12	0.95 ±0.33	1.29 ±0.24	1.26 ±0.26	1.38 ±0.23	1.49 ±0.29	1.37 ±0.25	1.09 ±0.22	1.09 ±0.26	1.00 ±0.23

* version 3 data analysis (Aeronet)

Table A 8: Number of days with AOD observations at Birkenes made within the months.

Month/Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
Number of days with cloud-free and quality assured observations (lev 2; lev 1.5 for 2013)											
2009				1	22	25	11	13	15	12	
2010					13	16	18	15	16	10	6
2011	7	2	20	23	18	20	15	13			
2012			11	12	10	7	16	18	9	12	2
2013							26	18	13	7	
2014			12	17	16	25	20	13	7		
2015			6	1	14	21	21	16	9	11	3
2016	1	9	11	8							1*
2017*				2	17	15	17	11	6	8	
Total	8	11	60	64	110	129	144	118	75	60	12

* version 3 data analysis (Aeronet)

APPENDIX II

Description of instruments and methodologies

ON THE INSTRUMENTAL METHODS USED FOR THE MEASUREMENTS OF THE VARIOUS GREENHOUSE GASES AT BIRKENES AND ZEPPELIN OBSERVATORIES

In this section of the appendix, the instrumental methods used for the measurements of the various greenhouse gases are presented. Additionally we explain the theoretical methods used in calculation of the trends.

Details about greenhouse gas measurements and recent improvement and extensions

Table A 9: Instrumental details for greenhouse gas measurements at Zeppelin and Birkenes.

Component		Instrument and method	Time res.	Calibration procedures	Start - End	Comment
Methane (Birkenes)	CH ₄	Picarro CRDS G1301 CO ₂ /CH ₄ /H ₂ O	5 s	Working std. calibrated against GAW stds at EMPA	19. May 2009 ->	Data coverage in 2017: 96.3%
Methane (Zeppelin)	CH ₄	GC-FID	1h	NOAA reference standards	Aug 2001- Apr 2012	
Methane (Zeppelin)	CH ₄	Picarro CRDS	1 h 5 sec	ICOS reference standards	20.Apr. 2012 ->	Data coverage 2017: 97%
Nitrous oxide (Zeppelin)	N ₂ O	GC-ECD	30 min	Hourly, working std. calibrated vs. NOAA stds		Data coverage 2017 70%
Carbon monoxide (Zeppelin)	CO	GC-HgO/UV	20 min	Every 20 min, working std. calibrated vs. GAW std.	Sep. 2001 - 2012	
Carbon monoxide (Zeppelin)	CO	Picarro CRDS	1 h 5 sec	ICOS reference standards.	20.Apr 2012 ->	Data coverage 2017: 97%
Carbon dioxide (Zeppelin)	CO ₂	Li-Cor	1 h	NOAA reference standards	1989 - 2012	CO ₂ measured by ITM Stockholm University (SU) until 2012
Carbon dioxide (Zeppelin)	CO ₂	Picarro CRDS from 20.4.2012	1 h 5 sec	ICOS reference standards	20.Apr. 2012 ->	Data coverage 2017: 97%
Carbon dioxide (Birkenes)	CO ₂	Picarro CRDS G1301 CO ₂ /CH ₄ /H ₂ O	5 s	Working std. calibrated against GAW stds at EMPA	Measurements started 19 May 2009.	Data coverage in 2017: 96.1%
CFC-11 CFC-12 CFC-113 CFC-115 HFC-125 HFC-134a HFC-152a HFC-365mfc HCFC-22 HCFC-141b HCFC-142b H-1301 H-1211 H-2402	CFCl ₃ CF ₂ Cl ₂ CF ₂ ClCFCl ₂ CF ₃ CF ₂ Cl CHF ₂ CF ₃ CH ₂ FCF ₃ CH ₃ CHF ₂ CF ₃ CH ₂ CHF ₂ C H ₃ CHF ₂ Cl CH ₃ CFCl ₂ CH ₃ CF ₂ Cl CF ₃ Br CF ₂ ClBr	ADS-GCMS	4 h	Every 4 hours, working std. calibrated vs. AGAGE std.	4. Jan 2001- 2010	The measurements of the CFCs have higher uncertainty and are not within the required precision of AGAGE. See next section for details.

Table cont.:

Component		Instrument and method	Time res.	Calibration procedures	Start - End	Comment
Chloromethane Bromomethane Dichloromethane Trichloromethane Trichloroethane Trichloroethene Tetrachloroethene Sulphurhexafluoride	CH ₃ Cl CH ₃ Br CH ₂ Cl ₂ CHCl ₃ CH ₃ CCl ₃ CHClCCl ₂ CCl ₂ CCl ₂ SF ₆	ADS-GCMS	4h	Every 4 hours, working std. calibrated vs. AGAGE std.	4. Jan 2001-2010	
Nitrogen trifluoride PFC-14 PFC-116 PFC-218 PFC-318 Sulphurhexafluoride Sulfuryl fluoride HFC-23 HFC-32 HFC-125 HFC-134a HFC-143a HFC-152a HFC-227ea HFC-236fa HFC-245fa HFC-365mfc HFC-43-10mee HCFC-22 HCFC-141b HCFC-142b CFC-11 CFC-12 CFC-113 CFC-115 H-1211 H-1301 H-2402 Chloromethane Bromomethane Dichloromethane Trichloromethane Trichloroethane Dibromomethane Trichloroethene Tetrachloroethene Carbon tetrachloride Ethane Propane Butane Pentane Benzene Toluene	NF ₃ CF ₄ C ₂ F ₆ C ₃ F ₈ c-C ₄ F ₈ SF ₆ SO ₂ F ₂ CHF ₃ CH ₂ F ₂ CHF ₂ CF ₃ CH ₂ FCF ₃ CH ₃ CF ₃ CH ₃ CHF ₂ CF ₃ CHFCF ₃ CF ₃ CH ₂ CF ₃ CF ₃ CH ₂ CHF ₂ CH ₃ CF ₂ CH ₂ CF ₃ CF ₃ (CHF) ₂ CF ₂ CF ₃ CHClF ₂ CH ₃ CCl ₂ F CH ₃ CClF ₂ CCl ₃ F CCl ₂ F ₂ CCl ₂ FCClF ₂ CClF ₂ CF ₃ CBrClF ₂ CBrF ₃ C ₂ Br ₂ F ₄ CH ₃ Cl CH ₃ Br CH ₂ Cl ₂ CHCl ₃ CH ₃ CCl ₃ CH ₂ Br ₂ CH ₃ CCl ₃ CCl ₂ CCl ₂ CCl ₄ C ₂ H ₆ C ₃ H ₈ C ₄ H ₁₀ C ₅ H ₁₂ C ₆ H ₆ C ₆ H ₅ CH ₃	Medusa-GCMS No. 19	2 h	Every 2 hours, working std. calibrated vs. AGAGE std	1. September 2010	Data coverage 2017: 92%
Ozone	O ₃		5 min			

DATA QUALITY AND UNCERTAINTIES

HALOCARBONS

In 2001 - 2010 measurements of a wide range of hydrochlorofluorocarbons, hydrofluorocarbons (HCFC-141b, HCFC-142b, HFC-134a etc.), methyl halides (CH₃Cl, CH₃Br, CH₃I) and the halons (e.g. H-1211, H-1301) were measured with good scientific quality by using ADS-GCMS. The system also measured other compounds like the chlorofluorocarbons, but the quality and the precision of these measurements was not at the same level. Table A 10 shows a list over those species measured with the ADS-GCMS at Zeppelin Observatory. The species that are in blue are of acceptable scientific quality and in accordance with recommendations and criteria of the AGAGE network for measurements of halogenated greenhouse gases in the atmosphere. Those listed in red have higher uncertainties and are not within the required precision of AGAGE. There are various reasons for this increased uncertainties; unsolved instrumental problems e.g. possible electron overload in detector (for the CFC's), influence from other species, detection limits (CH₃I, CHClCCl₂) and unsolved calibration problems (CHBr₃). Since 1. September 2010 the ADS-GCMS was replaced by a Medusa-GCMS system. The uncertainties improved for almost all species (Table A 11 for details), but there are periods where measurements of the CFC's were still not satisfactory. This is an unsolved problem within the AGAGE network.

Table A 10: ADS-GCMS measured species at Zeppelin from 4. January 2001 to 1. September 2010. Good scientific quality data in Blue; Data with reduced quality data in Red. The data are available through <http://ebas.nilu.no>. Please read and follow the stated data policy upon use.

Compound	Typical precision (%)	Compound	Typical precision (%)
SF ₆	1.5	H1301	1.5
HFC134a	0.4	H1211	0.4
HFC152a	0.6	CH ₃ Cl	0.6
HFC125	0.8	CH ₃ Br	0.8
HFC365mfc	1.7	CH ₃ I	5.1
HCFC22	0.2	CH ₂ Cl ₂	0.4
HCFC141b	0.5	CHCl ₃	0.3
HCFC142b	0.5	CHBr ₃	15
HCFC124	2.3	CCl ₄	0.5
CFC11	0.3	CH ₃ CCl ₃	0.6
CFC12	0.3	CHClCCl ₂	1.2
CFC113	0.2	CCl ₂ CCl ₂	0.7
CFC115	0.8		

Table below gives an overview over the species measured with the Medusa-GCMS systems at the AGAGE stations and the typical precision with the different instruments. The Medusa-GCMS instrument at the Zeppelin Observatory meet the same criteria as shown in the Table.

Table A 11: AGAGE measured species.

Compound	Typical precision (%)	Compound	Typical precision (%)
NF ₃	1		
CF ₄	0.15	H-1301	1.5
C ₂ F ₆	1	H-1211	0.5
C ₃ F ₈	3	H-2402	2
c-C ₄ F ₈	1.4	CH ₃ Cl	0.2
SF ₆	0.4	CH ₃ Br	0.6
SO ₂ F ₂	2	CH ₂ Cl ₂	0.5
HFC-23	0.7	CH ₂ Br ₂	1.5
HFC-32	3	CCl ₄	1
HFC-134a	0.5	CH ₃ CCl ₃	0.7
HFC-152a	1.4	CHClCCl ₂	2.5
HFC-125	0.7	CCl ₂ CCl ₂	0.5
HFC-143a	1.2	C ₂ H ₆	0.3
HFC-365mfc	5	C ₃ H ₈	0.3
HFC-43-10mee	3	C ₄ H ₁₀	0.6
HCFC-22	0.3	C ₆ H ₆	0.5
HCFC-141b	0.5	C ₇ H ₈	0.6
HCFC-142b	0.4		
CFC-11	0.15		
CFC-12	0.05		
CFC-113	0.2		
CFC-115	0.8		

METHANE

Methane is measured using a Picarro CRDS (Cavity Ring-Down Spectrometer) monitor which is calibrated against ICOS reference standards (NOAA scale). The instrument participates in yearly ring tests and applies to the ICOS system for calibration and measurement control. The continuous data are also compared to weekly flask samples sent to NOAA CMDL, Boulder Colorado. All data will be available for download from EBAS database <http://ebas.nilu.no>.

$\delta^{13}\text{C}_{\text{CH}_4}$

Air samples are collected in 1 L steel or aluminium canisters at the same air inlet as CH_4 . Two samples per week are sent to the Greenhouse Gas Laboratory at Royal Holloway University of London. Methane mole fraction was measured using a Picarro 1301 cavity ring down spectrometer (CRDS). Methane $\delta^{13}\text{C}$ analysis is carried out using a modified gas chromatography isotope ratio mass spectrometry system for all samples (Trace Gas and Isoprime mass spectrometer, Isoprime Ltd.) with 0.05‰ repeatability. All measurements for the canisters are made in triplicate. See Fischer et al, 2017 and Myhre et al, 2016 for more details.

N_2O MEASUREMENTS

N_2O is measured using a gas chromatograph with an electron capture detector. Due to malfunctioning valves, data are missing from mid May to mid August 2017. The instrument is old and worn and has now been replaced by a new mid-IR Cavity Ring Down instrument (December 2017). This new instrument is doing continuous measurements with improved precision and higher measurement frequency (< 1 min).

CO_2 MEASUREMENTS

Carbon dioxide (CO_2) is monitored using a Picarro Cavity Ring-Down Spectrometer for continuous measurements, calibrated against a set of ICOS reference standards (NOAA scale). The instrument participates in yearly ring tests and applies to the ICOS system for calibration and measurement control. The continuous data are also compared to weekly flask samples sent to NOAA CMDL, Boulder Colorado. All data will be available for download from the EBAS database <http://ebas.nilu.no>.

CO MEASUREMENTS

Carbon monoxide (CO) is monitored using a Picarro Cavity Ring-Down Spectrometer for continuous measurements, calibrated against a set of ICOS reference standards (NOAA scale). The instrument participates in yearly ring tests and applies to the ICOS system for calibration and measurement control. The continuous data are also compared to weekly flask samples sent to NOAA CMDL, Boulder Colorado. All data will be available for download from the EBAS database <http://ebas.nilu.no>.

IMPLEMENTATION OF MEASUREMENTS FROM BIRKENES AND ZEPPELIN INTO ICOS RESEARCH INFRASTRUCTURE

Integrated Carbon Observation System (ICOS) is a European research infrastructure forming an observation system that will measure and assess atmospheric CO₂, CH₄ and CO concentrations ensuring independent and reliable measurements. ICOS-Norway (<https://no.icos-cp.eu>) contributes to the network of atmospheric measurements with two observatories, Birkenes and Zeppelin.

ICOS has divided their sites into two levels, Class 1 and Class 2, dependent on their instrumental setup. At Class 1 sites a wider range of measurements of different species are required. Whilst, Class 2 is the low cost option where less parameters are mandatory. The table below shows mandatory and recommended parameters for ICOS Class 1 and Class 2 sites:

Category	Gases, continuous	Gases, periodical	Meteorology, continuous	Eddy Fluxes
Class 1 Mandatory parameters	<ul style="list-style-type: none"> • CO₂, CH₄, CO : at each sampling height 	<ul style="list-style-type: none"> • CO₂, CH₄, N₂O, SF₆, CO, H₂, ¹³C and ¹⁸O in CO₂: weekly sampled at highest sampling height† • ¹⁴C (radiocarbon integrated samples): at highest sampling height 	<ul style="list-style-type: none"> • Air temperature, relative humidity, wind direction, wind speed: at highest and lowest sampling height* • Atmospheric Pressure • Planetary Boundary Layer Height** † 	
Class 2 Mandatory parameters	<ul style="list-style-type: none"> • CO₂, CH₄ : at each sampling height 		<ul style="list-style-type: none"> • Air temperature, relative humidity, wind direction, wind speed: at highest and lowest sampling height* • Atmospheric Pressure 	
Recommended parameters***	<ul style="list-style-type: none"> • ²²²Rn, N₂O, O₂/N₂ ratio • CO for Class 2 stations 	<ul style="list-style-type: none"> • CH₄ stable isotopes, O₂/N₂ ratio for Class 1 stations: weekly sampled at highest sampling height 		<ul style="list-style-type: none"> • CO₂ : at one sampling height

* Atmospheric temperature and relative humidity recommended at all sampling heights

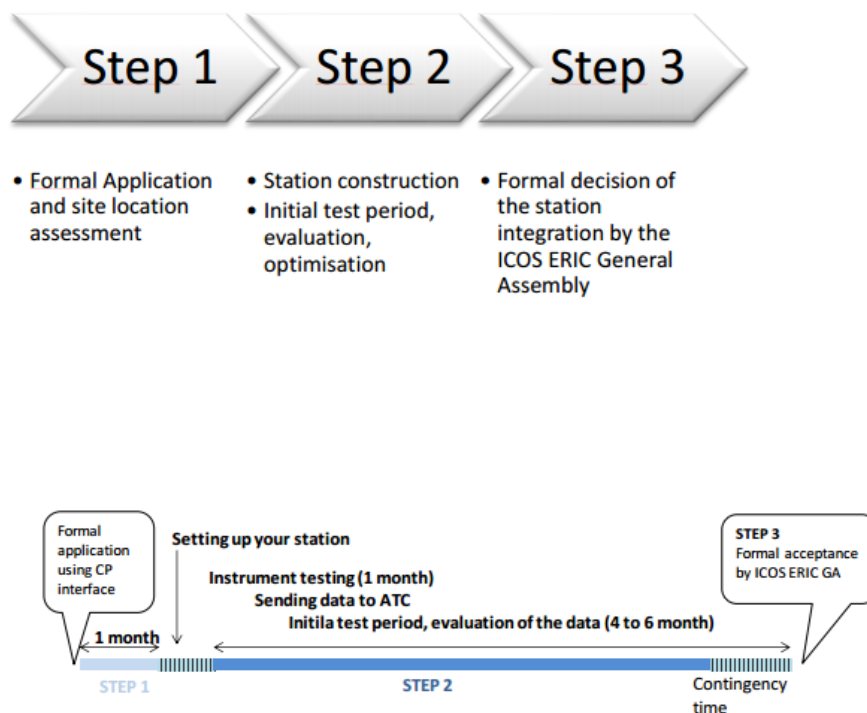
** Only required for continental stations.

*** Recommended for its scientific value but support from ATC in terms of protocols, data base, spare analyzer will not be ensured as long as the parameters are not mandatory.

ICOS-Norway will upgrade Zeppelin Observatory to become an ICOS Class 1 site and Birkenes Observatory to an ICOS Class 2 site, and comply with the ICOS quality assurance and quality control.

To become an ICOS site it requires that the site goes through a so-called labeling process to get a certificate as a qualified ICOS site. The labeling process evaluate the site set up, the instrumentation with its calibration set-up and data handling, as a quality certificate of the data output from the ICOS site. The labeling process is a time consuming process as the

instrument (Picarro) needs to be evaluated for minimum 1 month at the central ICOS lab, ICOS Atmospheric Thematic Centre (ATC) in France. Another central ICOS lab, ICOS Central Analytical Laboratories (CAL) in Germany, provides calibration cylinders and it takes 4 months to produce them. The same lab provides calibration cylinders and it takes 4 months to produce them. After having the instrument and calibration routines established at the site, the measurements will have to go through a 6 months evaluation period before approval as an ICOS site. In total it takes about 1.5 years to get an ICOS certificate.



The figure above shows the three steps in the labeling process of an ICOS station.

Status of the labeling process for Birkenes and Zeppelin Observatories:

- Both stations have their analyzers (Picarro) installed and run continuous measurements of CH₄ and CO₂ at both sites and CO at Zeppelin.
- Both analyzers have been at ICOS ATC for 1 month evaluation.
- ICOS calibration tanks and calibration routines have been implemented at Zeppelin
- Establish automatic data flow to the ICOS Atmospheric Thematic Centre (ATC) and Carbon Portal at Zeppelin.
- Implement ICOS data quality control procedures for Zeppelin data
- At Zeppelin, started sampling of 14C radiocarbon
- Mandatory meteorological parameters are measured with the upgraded equipment to meet the ICOS precision criteria at both sites.
- Have the permission by the landowner and Avinor to install a tall sampling tower (up to 100m) at Birkenes.

Plans for 2018:

- Implement ICOS calibration tanks and routines at Birkenes
- Establish automatic data flow to the ICOS ATC and Carbon Portal for Birkenes data
- Implement ICOS data quality control procedures for Birkenes data.

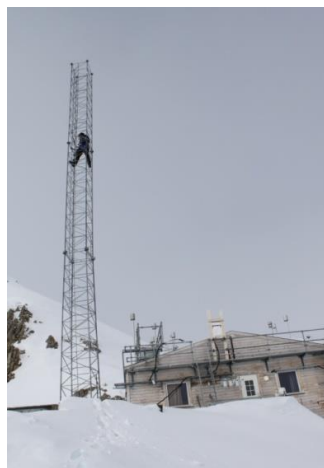
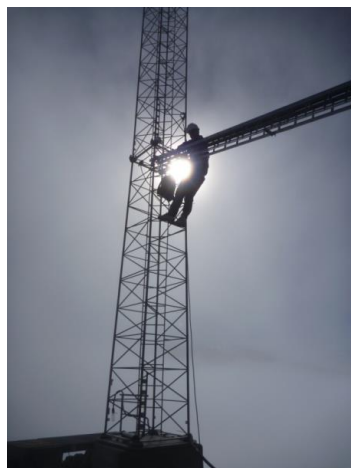
- At Zeppelin, implement N₂O measurements in the ICOS setup and quality routines, including own ICOS calibration tanks.
- Continue the process with local authority to get permission to install a tall sampling tower (up to 100m) and with ICOS ATC to decide the necessary height of the tower at Birkenes.

At class 1 sites an automatic flask sampler is mandatory, but as this is under development by ICOS CAL, it is not clear when the sampler will be ready for installation at Zeppelin. Most likely the flask sampler will be installed in 2019.

AIR INLET AT ZEPPELIN

In 2011 the air inlet for the GHG measurements at Zeppelin were improved to reduce possible influence from the station and visitors at the station. The inlet was moved away from the station and installed in a 15 m tower nearby for the following components:

- N₂O
- CH₄
- CO₂
- CO
- Halogenated compounds
- NOAA flasks sampling program
- Isotope flask sampling of CO₂ and CH₄



DETERMINATION OF BACKGROUND DATA

Based on the daily mean concentrations an algorithm is selected to find the values assumed as clean background air. If at least 75% of the trajectories within +/- 12 hours of the sampling day are arriving from a so-called clean sector, defined below, one can assume the air for that specific day to be non-polluted. The remaining 25% of the trajectories from European, Russian or North-American sector are removed before calculating the background.

CALCULATION OF TRENDS FOR GREENHOUSE GASES AND VOCs

To calculate the annual trends the observations have been fitted as described in Simmonds et al. (2006) by an empirical equation of Legendre polynomials and harmonic functions with linear, quadratic, and annual and semi-annual harmonic terms:

$$f(t) = a + b \left(\frac{N}{12} \right) \cdot P_1 \left(\frac{t}{N} - 1 \right) + \frac{1}{3} \cdot d \left(\frac{N}{12} \right)^2 \cdot P_2 \left(\frac{t}{N} - 1 \right) + c_1 \cdot \cos \left(\frac{2\pi t}{12} \right) + s_1 \sin \left(\frac{2\pi t}{12} \right) + c_2 \cos \left(\frac{4\pi t}{12} \right) + s_2 \sin \left(\frac{4\pi t}{12} \right)$$

The observed f can be expressed as functions of time measures from the 2N-months interval of interest. The coefficient a defines the average mole fraction, b defines the trend in the mole fraction and d defines the acceleration in the trend. The c and s define the annual and inter-annual cycles in mole fraction. N is the mid-point of the period of investigation. P_i are the Legendre polynomials of order i .

This equation is used for the VOCs, while for the GHGs the fit between the empirical equation and observations improves if the semi-annual harmonic terms are replaced by an additional Legendre polynomial.

In this year's report we are applying a new and improved method for estimating the *uncertainties* in the estimated annual trends from the time series regression modelling. In previous years, these uncertainties were estimated using a standard method from ordinary linear regression, where it was implicitly assumed that the residuals of the fitted regression models were uncorrelated. However, the use of such standard methods is known to often underestimate the true uncertainties in the estimated parameters if these assumptions are not true, i.e. if the residuals are autocorrelated, which to a large extent is the case for the regression models being fitted here.

In the current report we have replaced the standard method with a new method for estimation of uncertainties in the estimated annual trend that takes into account the presence of autocorrelated errors in the fitted model residuals. The new method also takes into account possible presence of heteroscedasticity, which means that the variances of the residuals might vary with the level of the time series, which also affects the uncertainties of the estimated trend. To this end we use the routine `vcovHAC` in the `sandwich` package in R (R Core Team, 2018) as described in Zeileis (2006; 2004), to estimate standard deviation of all estimated parameters of the time series regression models. Here HAC is short for Heteroscedastic and Autocorrelation Consistent.

It is important to emphasize that the new method only alters the estimated *uncertainties* of the annual trend estimates. The annual trend estimates themselves are not influenced by this update and have been correctly estimated using standard linear regression also in previous year's reports.

ON THE SURFACE IN SITU OBSERVATIONS OF AEROSOL MICROPHYSICAL AND OPTICAL PROPERTIES AT BIRKENES, ZEPPELIN AND TROLLHAUGEN OBSERVATORY

Table A 12: Overview of atmospheric aerosol parameters measured by surface in situ observations operated at which station

	Birkenes	Trollhaugen	Zeppelin
Particle Number Size Distribution (fine size range $D_p < 0.8 \mu\text{m}$)	X	X	X
Particle Number Size Distribution (coarse size range $D_p > 0.8 \mu\text{m}$)	X		
Aerosol Scattering Coefficient (spectral)	X	X	
Aerosol Absorption Coefficient (spectral)	X	X	X
Cloud Condensation Nuclei (indirect climate effect)	X		

Concerning surface in situ observations of microphysical and optical properties of atmospheric aerosol, the table on the left gives an overview over the parameters observed at Birkenes, Trollhaugen, and Zeppelin stations and operated by NILU.

To achieve high quality data with appropriate uncertainty and precisions, this requires networked instruments to participate in inter-comparisons at ACTRIS aerosol calibration centre in Leipzig, Germany, in regular

intervals. This activity has proven to be necessary in order to ensure comparable measurements within the distributed infrastructure. The frequency of these inter-comparisons, once every 2-3 years, is balanced with minimising the downtime associated with these quality assurance measures. In 2016, instruments targeting the direct aerosol climate effect were in the focus of inter-comparisons. Both the integrating nephelometer and the newer filter absorption photometer, measuring the spectral aerosol particle scattering and absorption coefficients respectively, were scheduled for being inter-compared, with satisfactory outcome in both cases.

With respect to microphysical aerosol properties, the particle number size distribution (PNSD) at surface-level is observed at all 3 stations covered in this report, at least over parts of the relevant range in particle size. The relevant particle sizes cover a range of $0.01 \mu\text{m} - 10 \mu\text{m}$ in particle diameter. The diameter range of $1.0 \mu\text{m} - 10 \mu\text{m}$ is commonly referred to as coarse mode, the range $D_p < 1.0 \mu\text{m}$ as fine mode. The fine mode is separated further into Aitken-mode ($0.01 \mu\text{m} < D_p < 0.1 \mu\text{m}$) and accumulation mode ($0.1 \mu\text{m} < D_p < 1 \mu\text{m}$). The distinction of these modes is justified by different predominant physical processes as function of particle size. In the Aitken-mode, particles grow by condensation of precursor gases from the gas-phase, and coagulate among themselves or with accumulation mode particles. Accumulation mode particles grow by taking up Aitken-mode particles or by mass uptake while being activated as cloud droplets, and they are removed by precipitation. Coarse mode particles in turn are formed by break-up of biological or crustal material, including pollen, bacteria, and fungus spores, and removed by gravitational settling and wet removal. The PNSD of an aerosol is needed for quantifying any interaction or effect of the aerosol since all of them depend strongly on particle size.

To measure the PNSD over the full relevant size range, several measurement principles need to be combined. A Differential Mobility Particle Spectrometer (DMPS) measures the particle

number size distribution, now in the range of 0.01 - 0.8 μm particle diameter after several improvements of the instruments at all three stations, i.e. almost the full fine mode. In a DMPS, the particles in the sample air stream are put into a defined state of charge by exposing them to an ionised atmosphere in thermal equilibrium. The DMPS uses a cylindrical capacitor to select a narrow size fraction of the particle phase. The particle size of the selected size fraction is determined by the voltage applied to the capacitor. The particle number concentration in the selected size fraction is then counted by a Condensation Particle Counter (CPC). A mathematical inversion that considers charge probability, diffusional losses of particles in the system, transfer function of the capacitor, and counting efficiency of the CPC is then used to calculate the particle number size distribution.

The PNSD of particles with diameters $0.25 \mu\text{m} < D_p < 30 \mu\text{m}$ is measured with an Optical Particle Spectrometer (OPS). In the OPS, the particles in the sample stream are focussed through a laser beam. The instrument registers number and amplitude of the pulses of light scattered by the particles. The particle pulses are sorted into a histogram by their amplitude, where the pulse amplitude yields the particle diameter and the pulse number the particle concentration, i.e. together the PNSD. Both, the DMPS and the OPS, yield method specific measures of the particle diameter, the electrical mobility and the optical particle diameter, respectively. When related to the spherical equivalent geometric particle diameter commonly referred to, both particle size measures depend on particle shape (causing a 5% systematic uncertainty in particle diameter), the optical particle diameter in addition on particle refractive index (causing a 20% systematic uncertainty in particle diameter). Where possible, the PNSDs provided by DMPS and OPS are joined into a common PNSD, in this report. To quality assure this process, PNSDs are accepted only if DMPS and OPS PNSD agree within 25% in particle diameter in their overlap size range. Together, both instruments provide a PNSD that spans over 3 orders of magnitude in particle diameter, and over 6 orders of magnitude in particle concentration.

Another measured microphysical property is the Cloud Condensation Nuclei (CCN) Concentration, i.e. the concentration of particles that could act as condensation nuclei for liquid-phase cloud droplets, which is a property depending on the water vapour supersaturation. The corresponding instrument, a Cloud Condensation Nucleus Counter (CCNC), exposes the aerosol sample to an “artificial cloud” of defined supersaturation, and counts the cloud activated particles optically. Observations of the CCN concentration are essential for quantifying the indirect aerosol climate effect. The CCNC at Birkenes has been in operation since 2012, but is currently not included in the monitoring programme.

Optical aerosol parameters quantify the direct aerosol climate effect. The observation programme at Birkenes includes the spectral particle scattering coefficient $\sigma_{sp}(\lambda)$ and the spectral particle absorption coefficient $\sigma_{ap}(\lambda)$. The scattering coefficient quantifies the amount of light scattered by the aerosol particle population in a sample per distance a light beam travels through the sample. The absorption coefficient is the corresponding property quantifying the amount of light absorbed by the particle population in the sample. An integrating nephelometer is used for measuring $\sigma_{sp}(\lambda)$ at 450, 550, and 700 nm wavelength. In this instrument, the optical sensors look down a blackened tube that is filled with aerosol sample. The tube is illuminated by a light source with a perfect cosine intensity characteristic perpendicularly to the viewing direction. It can be shown mathematically that this setup integrates the scattered light seen by the optical sensors over all scattering angles. The nephelometer at Birkenes has successfully undergone quality assurance by intercomparison

within the EU research infrastructure ACTRIS in 2015. In 2017 we detected drift in the older filter absorption photometer operated at Birkenes since 2009 through carefully implemented quality control within ACTRIS. The drift was detected by operating the older filter absorption photometer in parallel with a newer, more stable make and model in order to ensure a continuous, rupture-free aerosol absorption time series at Birkenes. In addition, the newer instrument was sent to an inter-comparison within the European research infrastructure for short-lived climate forcers ACTRIS. These exercises connect individual instruments to a network-wide primary standard, ensuring traceability and comparability of observations at stations in the network. The old instrument exhibiting the drift has since been decommissioned.

For the nephelometer at Trollhaugen, such intercomparisons are impossible because of the remote Antarctic location and associated logistical challenges. However, the Trollhaugen instrument undergoes the same regular on-site quality assurance as the Birkenes instrument, including regular calibration verification traceable to physical first principles (calibration with high-purity carbon dioxide, where scattering coefficient of carbon dioxide can be calculated from fundamental quantum mechanics).

The spectral particle absorption coefficient $\sigma_{ap}(\lambda)$ is measured by filter absorption photometers. A filter absorption photometer infers $\sigma_{ap}(\lambda)$ by measuring the decrease in optical transmissivity of a filter while the filter is loaded with the aerosol sample. The transmissivity time series is subsequently translated into an absorption coefficient time series by using Lambert-Beer's law, the same law also used in optical spectroscopy. The filter absorption photometers deployed at Birkenes are a custom-built 1 wavelength Particle Soot Absorption Photometer (PSAP), and a commercial 3-wavelength PSAP. The 1-wavelength PSAP received quality assurance by intercomparison within ACTRIS in 2013 discovering calibration stability issues. The 3-wavelength PSAP has undergone ACTRIS intercalibration successfully in 2015, i.e. without discovering any issues. Thus, both instruments are interpreted in combination to benefit from both, quality assurance in a research network and spectral capabilities. For 2013 and later, the data of the 3-wavelength PSAP are used, for 2010-2012, the data of the older 1-wavelength are used after being corrected by comparison with the newer instrument during the overlap period. For comparison with the nephelometer, the PSAP data has been transferred to a wavelength of 550 nm using the measured spectral dependence (3-wavelength PSAP), or by assuming an absorption Ångström coefficient α_{ap} of -1 (1-wavelength PSAP, adding 2% systematic uncertainty to the data).

For measuring $\sigma_{ap}(\lambda)$ at Zeppelin, a recently developed filter absorption photometer, an aethalometer type AE33, has been deployed in 2015. The AE33 features a larger spectral range of 370 - 950 nm as compared to other instruments, and offers a 7-wavelength resolution across this range. Previous aethalometer models suffered from high systematic uncertainties due to uncorrected dependencies on filter loading (e.g. Collaud Coen et al., 2010). Comparisons and calibrations within ACTRIS have shown that this systematic uncertainty has been reduced significantly in the AE33 model by an internal loading compensation (Drinovec et al., 2015). Due to this successful instrument evaluation, the instrument programme at Birkenes has been upgraded by an AE33 in 2017 as well.

All in situ observations of aerosol properties representing the ground-level are conducted for the aerosol at dry-state (RH < 40%) for obtaining inter-comparability across the network.

DETAILS ABOUT AEROSOL OPTICAL DEPTH MEASUREMENTS

The amount of particles in the air during sunlit conditions is continuously monitored by means of a Precision-Filter-Radiometer (PFR) sun photometer, located at the Sverdrup station in Ny-Ålesund and a Cimel instrument at Birkenes. The observations in Ny-Ålesund are performed in collaboration with PMOD/WRC (N. Kouremeti, S. Kazadzis), Davos, Switzerland. The main instrument characteristics are given below.



AERONET - Cimel C-318

- Sun (9 channels) and sky radiances
- Wavelength range: 340-1640 nm
- 15 min sampling
- No temperature stabilization
- AOD uncertainty: 0.01-0.02



PFR-GAW- Precision Filter Radiometer

- Direct sun measurements (4 channels)
- Wavelength range: 368 - 862 nm
- 1 min averages
- Temperature stabilized
- AOD uncertainty: 0.01

Figure 57: Photos and typical features of the standard instrument of the AERONET (left panel) and GAW PFR network instruments (right panel)

The sun-photometer measurements in Ny-Ålesund are part of the global network of aerosol optical depth (AOD) observations, which started in 1999 on behalf of the WMO GAW programme. The instrument is located on the roof of the Sverdrup station, Ny-Ålesund, close to the EMEP station on the Zeppelin Mountain (78.9°N, 11.9°E, 474 m a.s.l.). The Precision Filter Radiometer (PFR) has been in operation since May 2002. In Ny-Ålesund, the sun is below 5° of elevation from 10 October to 4 March, limiting the period with sufficient sunlight to the spring-early autumn season. However, during the summer months it is possible to measure day and night if the weather conditions are satisfactory. The instrument measures direct solar radiation in four narrow spectral bands centred at 862 nm, 501 nm, 411 nm, and 368 nm. Data quality control includes instrumental control like detector temperature and solar pointing control as well as objective cloud screening. Measurements made at full minutes are averages of 10 samples for each channel made over a total duration of 1.25 seconds.

SCIAMACHY TOMSOMI and OMI ozone columns and meteorological data from Ny-Ålesund are used for the retrieval of aerosol optical depth (AOD).

At Birkenes Observatory, aerosol optical depth measurements started in spring 2009, utilizing an automatic sun and sky radiometer (CIMEL type CE-318) of the global Aerosol Robotic Network (AERONET) at NASA-GSFC, with spectral interference filters centred at selected wavelengths: 340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, 1020 nm, and 1640 nm. The measurement frequency is approximately 15 minutes (this depends on the air-mass and time of day). Calibration is performed about once per year, at the Atmospheric Optics Group at the University of Valladolid (GOA-UVa), Spain. GOA manages the calibration for the AERONET sun photometers of the European sub-network of AERONET. Raw data are processed and quality assured centrally by AERONET. Data reported for 2009 - 2016 are quality-assured AERONET level 2.0 data, which means they have been pre- and post-field-calibrated, automatically cloud cleared and have been manually inspected by AERONET.

From 2017, only the new analysis algorithm (version 3) is used at the central AERONET analysis unit at NASA GSFC. In this version, the data quality control, including cloud screening, has been improved and a temperature correction has been applied to all wavelength channels. A comparison between the data from all years of operations at Birkenes (2009 - 2016) analysed with version 3 and version 2 revealed an increase of 25% in number of usable data, especially in the winter half-year. While during the summer months, the agreement between the two versions is better than 2% for both AOD and (470, 870nm) Ångström coefficient, there are significant deviations (>10%) in the months with less than 1000 observations over the whole period (November - February). A comprehensive analysis of the differences between the two versions and possible effects on trend studies is in progress; a paper by the NASA GSFC team will be published in the near future.

Due to the large gaps in data acquisition caused by the obligatory annual calibrations at the University of Valladolid combined with technical problems and occasionally unfavourable weather conditions at Birkenes, NILU decided to purchase a second Cimel instrument which will be operated alternately with the current instrument. It will be taken into operation in fall 2018.

OUTLOOK ON OBSERVATIONS OF AEROSOL OPTICAL DEPTH IN NY-ÅLESUND BEYOND 2017

A major obstacle to obtaining a year-round AOD climatology in the Arctic arises from the long polar night. To fill gaps in the aerosol climatology at Ny-Ålesund, a lunar photometer will be operated on a quasi-permanent basis in the frame of the SIOS infrastructure project. This is a collaborative initiative between NILU, PMOD/WRC and ISAC-CNR. Seasonal deployments of a lunar photometer owned by PMOD/WRC were already made in the winters of 2014/15 and 2016/17. A multiple-season deployment is envisaged from autumn 2018.



Figure 58: Moon PFR on the Kipp & Zonen tracker during the day (left, parking position) and during night-time measurements (right).

The PFR instrument modified by PMOD-WRC was installed on a tracker model Kipp & Zonen provided usually hosting a sun photometer. Figure 58 shows the instrument on the tracker during day-time and night-time. Six lunar cycles were monitored: the first during February 2014, while the other 5 during winter 2014-2015. We collected data on 66 measurement periods, from Moon-rise to Moon-set or from minimum-to-minimum elevation as in Polar Regions no set-rise events are possible. Among these, we obtained 17 distinct good measurement periods, due to the frequent occurrence of clouds. For further details see e.g. Mazzola et al., 2015.

APPENDIX III: Abbreviations

Abbreviation	Full name
ACSM-ToF	Aerosol Chemical Speciation Monitor
ACTRIS	Aerosols, Clouds, and Trace gases Research InfraStructure Network
ADS-GCMS	Adsorption-Desorption System - Gas Chromatograph Mass Spectrometer
AeroCom	Aerosol Comparisons between Observations and Models
AERONET	Aerosol Robotic Network
AGAGE	Advanced Global Atmospheric Gases Experiment
AIRS	Atmospheric Infrared Sounder
AMAP	Arctic Monitoring and Assessment
AOD	Aerosol optical depth
AWI	Alfred Wegener Institute
BC	Black carbon
CAMP	Comprehensive Atmospheric Monitoring Programme
CCN	Cloud Condensation Nuclei
CCNC	Cloud Condensation Nucleus Counter
CFC	Chlorofluorocarbons
CICERO	Center for International Climate and Environmental Research - Oslo
CIENS	Oslo Centre for Interdisciplinary Environmental and Social Research
CLTRAP	Convention on Long-range Transboundary Air Pollution
CO	Carbon monoxide
CPC	Condensation Particle Counter
DMPS	Differential Mobility Particle
EMEP	European Monitoring and Evaluation Programme
ENVR ^{plus}	Environmental Research Infrastructures Providing Shared Solutions for Science and Society
EOS	Earth Observing System
ERF	Effective radiative forcing ERF
ERFaci	ERF due to aerosol-cloud interaction
EU	European Union
EUSAAR	European Supersites for Atmospheric Aerosol Research
FLEXPART	FLEXible PARTicle dispersion model
GAW	Global Atmosphere Watch
GB	Ground based

Abbreviation	Full name
GHG	Greenhouse gas
GOA-UVA	Atmospheric Optics Group of Valladolid University
GOSAT	Greenhouse Gases Observing Satellite
GOSAT-IBUKI	Greenhouse Gases Observing Satellite "IBUKI"
GWP	Global Warming Potential
HCFC	Hydrochlorofluorocarbons
HFC	Hydrofluorocarbons
ICOS	Integrated Carbon Observation System
InGOS	Integrated non-CO2 Greenhouse gas Observing System
IPCC	Intergovernmental Panel on Climate Change
ISAC-CNR	Institute of Atmospheric Sciences and Climate (ISAC) of the Italian National Research Council
ITM	Stockholm University - Department of Applied Environmental Science
JAXA	Japan Aerospace Exploration Agency
LLGHG	Well-mixed greenhouse gases
MOCA	Methane Emissions from the Arctic Ocean to the Atmosphere: Present and Future Climate Effects
MOE	Ministry of the Environment
NARE	Norwegian Antarctic Research Expeditions
NASA	National Aeronautics and Space Administration
NEOS-ACCM	Norwegian Earth Observation Support for Atmospheric Composition and Climate Monitoring
NIES	National Institute for Environmental Studies
NOAA	<u>National Oceanic and Atmospheric Administration</u>
NRS	Norsk Romsenter
OC	Organic Carbon
ODS	Ozone-depleting substances
OH	Hydroxyl radical
OPS	Optical Particle Spectrometer
OSPAR	Convention for the Protection of the marine Environment of the North-East Atlantic
PFR	Precision filter radiometer
PMOD/WRC	Physikalisch-Meteorologisches Observatorium Davos/World Radiation Center
PNSD	Particle number size distribution

Abbreviation	Full name
ppb	Parts per billion
ppm	Parts per million
ppt	Parts per trillion
PSAP	Particle Soot Absorption Photometers
RF	Radiative forcing
RI	Research Infrastructure
RIMA	Red Ibérica de Medida fotométrica de Aerosoles
SACC	Strategic Aerosol Observation and Modelling Capacities for Northern and Polar Climate and Pollution
SCIAMACHY	SCanning Imaging Absorption spectroMeter for Atmospheric CHartography
SIS	Strategisk instituttsatsing
SMPS	Scanning Mobility Particle
TES	Tropospheric Emission Spectrometer
TOA	Top Of Atmosphere
TOMS OMI	Total Ozone Mapping Spectrometer Ozone Monitoring instrument
UN	United Nations
UNFCCC	United Nations Framework Convention on Climate Change
VOC	Volatile organic compounds
WDCA	World Data Centre for Aerosol
WDCS	World Data Centre of Aerosols
WMGHG	Well-mixed greenhouse gases
WMO	World Meteorological Organization

Norwegian Environment Agency

Telephone: +47 73 58 05 00 | **Fax:** +47 73 58 05 01

E-mail: post@miljodir.no

Web: www.environmentagency.no

Postal address: Postboks 5672 Sluppen, N-7485 Trondheim

Visiting address Trondheim: Brattørkaia 15, 7010 Trondheim

Visiting address Oslo: Grensesvingen 7, 0661 Oslo

The Norwegian Environment Agency is working for a clean and diverse environment. Our primary tasks are to reduce greenhouse gas emissions, manage Norwegian nature, and prevent pollution.

We are a government agency under the Ministry of Climate and Environment and have 700 employees at our two offices in Trondheim and Oslo and at the Norwegian Nature Inspectorate's more than sixty local offices.

We implement and give advice on the development of climate and environmental policy. We are professionally independent. This means that we act independently in the individual cases that we decide and when we communicate knowledge and information or give advice.

Our principal functions include collating and communicating environmental information, exercising regulatory authority, supervising and guiding regional and local government level, giving professional and technical advice, and participating in international environmental activities.