Atmospheric Ozone, Air Pollution and Climate

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Geological Time Scales

EO	N ER/	ERA PERIOD		EPOCH		Ma	Eirst migration of	Modorn humans	Migrations of fully	Great European	
	-		Quaternary		Holocene		0.011	fully modern	arrive in	modern humans from	Beginning civilisations:
					Pleistocene Late		-0.011-	humans out of Africa	Australia	South Asia to Europe	of agriculture Greek, Roman
					reiscocciie	Early	- 2.4 -		, lubil ullu		
				he	Pliocene	Early	- 3.6 -	24			
	<u>e</u> .			ge		Late	- 5.3 -	-34			
	R		Tertiary	Neo	Miocene	Middle	16.4				aj s
	2					Early	- 23.0 -				н н н н н н н н н н н н н н н н н н н
	G				Olígocene	Late	- 28.5 -	ā -38 4 1		ALANAL ALAA	- e
	-			aleogen		Late	- 34.0 -	e Welward V		I. PAR WALLARD	it i
					Eocene	Middle	- 41.3 -				
						Early	- 55.8 -	² 0 42	1 UKWANY	FT THE THE TYPE WORKED	20 E
				~	Paleocene	Early	- 61.0 -	-42	M M du de		
U	-				Late	Larry	- 65.5 -				
0			Cretaceou	IS	Early		- 99.6 -	100.000 00			20.000
8	2				Late		- 161 -	100,000 80,	,000 60,0	40,000	20,000 0
e	i i i i		Jurassic		Middle		- 176 -		A	ge (before present)	The Holocene
5	ŭ,				Late		- 200 -				The Holocene
Phi	Σ		Triassic		Middle		228 -		230-65 M	a: Formation	of the Earth
		_			Early		- 251 -		Dinosaur	s Hominids	
			Bormion		Middle		- 260 -	ca. 380 Ma	a.	Mammals	
			rennan		Early		- 271 -	First vertebrate lan	nd animals	Animals	
					Late		- 306 -			Multicellular Eukaryotes	life 4527 Ma:
		1	Pennsylvar	nian	Middle		- 311 -	ca. 530 Ma:		Prokaryotes	Formation of the Moon
		-			Early		- 318 -	750.635 Ma:		65 Ma 4.6 Ga	ca. 4000 Ma: End of the
			Mississippian		Middle		- 326 -	Two Snowball Earths		251	Late Heavy Bombardment;
	<u>.</u>				Farly		- 345 -		1//////////////////////////////////////		
	N				Late		- 359 -		8 1	2 B B	Harr 4 Ga
	8		Devonian		Middle		- 385 -			Č 6	ean ca. 3500 Ma:
	T				Early		- 416 -			8	Photosynthesis starts
	B	-	Silurian		Farly		- 419 -			JAY MARK	
					Late		- 423 -		161		
			Ordovician		Middle		428 -				
					Early		- 488 -				
					Late		- 501 -				
			Cambrian		Middle Early		- 513 -				
							- 542 -				
	2 1 3	te	Neopro	tero	zoic (7)		342		1		
			Heapre		Lore (L)		-1000 -		E I		J. J
i i i	6 Mi	idd	ddle Mesoproterozoic (Y)					Fold	and the second se	3 Ga	
=	ē	and the	by Balance Arrent Park			-1600 -	· · · · · · · · · · · · · · · · · · ·				
ie i		Larry Paleoprotorozon (X)				-2500 -					
ecambr	5 10	Late					2500				
	e						-3200-			Ga	
	E	Early								Contraction and the second second	68
4							- 4000 -			2.5	
											ca. 2300 Ma:
	5									Atmos	phere becomes oxygen-rich; first Spowball Earth

The Anthropocene

THE GREAT ACCELERATION

In September 2015 the nations of the world will meet to agree on Sustainable Development Indicators will be essential to asses progress





Figure 2: Twelve Earth system trends from 1750 to present.

Why observe the atmosphere from space?

Earth has enter and P. J. Crutze

From the Neolithi rose from 4M to Dramatic changes emissions sinc Energy supplied => Release of

- ⇒ Global transpo and land use cl
 ⇒ Climate Change
 ⇒ Global destruct
- ⇔ It is <u>impossible</u>
- ⇒ Environmental/(
- ⇒ Evidence base f

PASTEUR'S QUADRANT



Stoermer ging!



USE-INSPIRED RESEARCH: to pursue fundamental understanding but motivated by a question of use

Figure 1

Spatial and Temporal Scales relevant for measurements from LEO and GEO



European LEO and GEO Passive Remote Sensing of trace consitutents in the Anthropocene - Some Relevant History

1984-1988	Development and Submission to ESA for POEM/ Envisat AO, of SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY concept Burrows et al – hunting light
1988	Proposal of SCIA-mini for ERS-2 later descrped to GOME
1989	Selection of SCIAMACHY for ENVISAT
1990	Selection of GOME for ERS-2
1995 1998	Launch of GOME 20.04.1995 Proposal of GeoSCIA IUP/IFE-UB to ESA EEM-1
2002	Proposal of GeoSCIA++ UV-VIS-NIR-SWIR-TIR/Ligthning/firto ESA EEM-2
2002	Launch of SCIAMACHY on ENVISAT 28.02 2002
2002	Proposal of GeoTROPE UV-VIS-NIR-SWIR-TIR to ESA EEM-3
2004/5 2006	Proposal of GeoSCIA-R and GeoSCIA-Lite EUMESAT Post Metop Committee recommends GOME-2 follow on UVNS
2006	Methane and carbon dioxide Mapper MaMap 01– Aircraft - UB
2006	EU Copernicus funds UVNS/Sentinel 5 Metop Second Generation
2006	Launch of GOME-2 on MetOp A
2008	CarbonSat and CarbonSat Constellation studies at UB - SCIA Heritage
2010	CarbonSat selected for ESA EE8 Phase AB1 Studies
2011	Start of SCIA-ISS studies UB NICT / Decommssioning of ERS-2
2012	Loss of Envisat 9 th April
2012	Launch of GOME-2 on Metop-B 17 th September
2013	Sentinel 5 agreed for Metop Second Generation 2020- 2034
2015	September ESA decision either CarbonSat or FLEX for ESA EE8????

SCIAMACHY: Target Molecules



*EXZELLENT.

Ozone Production & Catalytic Destruction









Update Weber et al. 2011, WMO 2014

Ozone hole above Antarctica 2015



Latitude-altitude dependence of ozone trends Impact of SST / T-hiatus on BDC



 minimum in the tropical 30-35 km range related slowing BDC and changing NO_X Gebhardt et al 2012 OQS and ACP 2013 and Aschmann et al ACP 2014



FIg. 2. Observed and simulated tropical (20°N-20°S) LS O₃ partial columns (17-21 km). Anomalies are deviations from the modelled 1980-2013 averages.



FIg. 5. Linear trends of El surface temperature from 2002–2013. Stippling indicates where the trend exceeds the 95% confidence threshold. Setup adapted from Kosaka and Xie (2013).

Some Key Processes in Global Tropospheric Chemistry/ Chemical Weather ~ 2014



Tropopsheric NO₂ and Sources?



*EXZELLENT.

Satellite NO₂ Trends: The Global View 1995-



A. Richter et al., Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437 2005





*EXZELLENT.

The spatial distribution of satellite NO₂ trends



Many cities can be identified



Hilboll et al., : Long-term changes of tropospheric NO2 over megacities derived from multiple satellite instruments, *Atmos. Chem. Phys.*, 13, 2013

NO₂ Trends over some Megacities/Urban Aglomerations





NO₂ levels are changing in cities throughout the world. Contributing factors are

- Urbanisation
- Population growth
- Increase in standard of living
- changes in fuels used
- Improvements in emission controls







Recent NO₂ Trends above China



- Until 2011, there was continuous increase in NO₂
- After two years of stagnation, 2014 saw a large decrease
- \Rightarrow economic slow down?
- \Rightarrow Improved technology?
- \Rightarrow Switch in fuels used?
- \Rightarrow Other factors?







NOx Emissions from Shipping



With estimate of NO_2 lifetime, NO_x emissions can be estimated => agreement within error bars.

But: error bars mainly from lifetime)

A. Richter et al., Satellite Measurements of NO2 from International Shipping Emissions, *Geophys. Res. Lett.*, 31, L23110, doi:10.1029/2004GL020822, 2004
A. Richter et al.:: An improved NO2 retrieval for the GOME-2 satellite instrument, *Atmos. Meas. Tech.*, 4, 1147-1159, doi:10.5194/amt-4-1147-2011, 2011





Ship emissions:

- large source of $\mathrm{NO}_{\mathrm{x}},\,\mathrm{SO}_{\mathrm{x}}$ and aerosols
- relevant input into marine boundary layer
- well defined NO₂ patterns in Red Sea and Indian Ocean in GOME-2 data
- consistent with pattern of shipping



Latest Aircraft Instrument IUP UB - AirMap instrument









- Push-broom imager
- 48° field of view
- Swath ~ flight altitude
- Acton 300i spectrometer
- Princeton frame transfer CCD
- Fibre optics
- Only narrow spectral range
- Video camera, GPS
- At typical
 - flight altitude (3000m)
 - aircraft speed (60m/s)
 - Integration time (0.5s)
 - \Rightarrow 35 pixels @ 80 x 30 m²



Some Recent AirMap targets – Northern Germany and Shipping











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Spatial resolution – the evolution to meet the needs of tropospheric chemistry spatial and temporal scales?



AirMap: Bucharest VC NO₂ 08.09.2014

- ESA Campaign
- Composite of the results from the flights on on 14Large values
- Low wind speed (≈ 0 1 m/s), alternating directions







MAX-DOAS Measurements in Athens



- 3.2 million inhabitants
- Emissions from industry and transportation
- Intense photochemistry
- Affected by fires and Sahara dust events





- Oct 2012 now
- 330 500 nm
- 8 viewing azimuths
 - Ocean
 - Airport
 - City x 5
 - Background
- -1°.. 30° elevation + zenith
- 15 minutes cycle
- Closest zenith reference



Spatial Gradients City Pollution





Weekly Cycle in NO2 for Athens





- Very clear weekly cycle
- Most pronounced over city directions
- Most pronounced in lowest elevation angles
- Best seen during summer break





NO₂ Trends above Europe









SCIAMACHY SO₂: The global Picture







***EXZELLENT.**

SO₂ columns above China



- SO₂ increase in similar regions as NO₂ increase
- Main reason is increase in power generation using coal



- Legislation made flue gas desulphurisation mandatory after 2006
- Marked decrease in SO₂ but small upward trend since 2009 (industrial sources)

*EXZELLENT.





NMVOCs: HCHO and glyoxal



Wittrock, F., et al., (2006), Simultaneous global observations of glyoxal and formaldehyde from space, Geophys. Res. Lett., 33, L16804, doi:10.1029/2006GL026310.

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VC CHOCHO [molec cm⁻²] 1.1 1015 9.5 1014 8.0 1014 6.5 1014 5.0 1014 3.5 1014 2.0 1014

1.8 1016

1.5 1016 1.2 1016 9.0 1015

6.0 1015 3.0 1015 0.0 1000

Sources:

- **Biogenic**
- Fires
- Fossil fuel
- VOC oxidation

Sinks:

- **Photolysis**
- OH

Relevance:

- O_3 production
- SOA



Glyoxal, CHO.CHO columns



- Glyoxal is a VOC with little primary emission
- Main sources are oxidation of biogenic and anthropogenic VOCs, biomass burning
- Seasonality of glyoxal indicates mainly biogenic precursors
- Consistent upward trend over SCIAMACHY time series
- Additional anthropogenic emissions?
- Land use changes?
- More biomass burning?

Vrekoussis, M., et al., Temporal and spatial variability of glyoxal as observed from space, *Atmos. Chem. Phys.*, 9, 4485-4504, 2009



Aerosols in the troposphere



Sources

- Sea-salt
- Dust / sand
- Combustion / fires
- Secondary aerosols (SO₂, HNO₃, SOA, …)

Sinks

• Wet & dry deposition

Relevance

- Health
- Scattering

Yoon, J., Changes in atmospheric aerosol loading retrieved from space based measurements during the past decade, Atmos. Chem. Phys. Discuss., 13, 26001-26041, doi:10.5194/acpd-13-26001-2013, 2013.



Changes in aerosol AOT 2003-2008

2003 - 2008





Figure 13. Time series of atmospheric AOTs normalized to their average mean values from the MODIS-Terra (MOD), MISR-Terra (MIS), SeaWiFS-OrbView-2 (SEA), MODIS-Aqua (MYD), and AERONET (AER) data sets; tropospheric nitrogen dioxide and sulfur dioxide columns from SCIAMACHY (SCIA) over eastern China (region 6); and Chinese GDP from 2003 to 2008.



- Downward trend in Western Europe and Eastern US
- Upward trend in Asia
- Some differences between instruments

Yoon, J., Changes in atmospheric aerosol loading retrieved from space based measurements during the past decade, Atmos. Chem. Phys. Discuss., 13, 26001-26041, doi:10.5194/acpd-13-26001-2013, 2013.



Changes in CTH using observations of O₂ A band and SACURA GOME- SCIAMACHY GOME-2



Figure 13. Global trend β in CTH anomaly, statistically significant at 95% confidence level. Data are gridded onto a mesh of 2°-sided cells.

Fable 4. Overview of zonal trends in CTH [m yr⁻¹], ENSO excluded, masking any data within the box 170–120° W, 5° N–5° S. Bootstrap resamples $n = 10^3$. The zonal values are not weighted by the respective land and water abundances.

	Belt		Land + water	Land	Water
With ENSO	Tropics	5° N-5° S	-4.34 ± 5.65	-1.56 ± 4.02	-5.15 ± 8.21
	Tropics	20° N-20° S	-2.16 ± 2.97	$+1.83 \pm 4.40$	-3.39 ± 5.32
	Mid-latitude	30-60° N	-2.17 ± 1.52	-2.85 ± 4.23	-1.52 ± 3.68
	Mid-latitude	30-60° S	-2.71 ± 2.59	-2.70 ± 9.25	-2.71 ± 2.47
Without ENSO	Tropics	5° N-5° S	-1.80 ± 6.00	-1.43 ± 5.05	-1.99 ± 8.52
	Tropics	20° N-20° S	$+0.53 \pm 3.53$	$+5.93 \pm 5.33$	-1.74 ± 4.36
	Mid-latitude	30-60° N	-2.11 ± 3.09	-2.72 ± 4.60	-1.53 ± 3.70
	Mid-latitude	30-60° S	-2.78 ± 2.54	-3.24 ± 8.77	-2.75 ± 2.35

Natural variability and instrument or algorithm error



SCIAMACHY on ENVISAT: CO₂ & CH₄ from space





Methane SCIAMACHY/WFMD 2003-2005 Harbin Urumai Chengdu Wuhar Shanghai Chongging New Delhi Hong Kong Calcutta Mumbai Vijayawada Mysore Methane column averaged mixing ratio [ppb] 1675 1710 1745 1780 1815 Univ.Bremen, IUP/IFE WFMDv2.0.2/L3(0.6x0.5)/nsm=0











SCIAMACHY XCO₂: Anthropogenic source regions





Temperature response of terrestrial carbon sink (Schneising et al., 2014, ACP)



Highlights GHG

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- Years with higher surface temperatures during the growing season are associated with larger CO₂ growth rates and smaller seasonal cycle amplitudes (reduced net carbon uptake by vegetation)
- Temperature sensitivity: 2.7 ± 0.7 GtC/yr/K
- Positive carbon-climate feedback unless the biosphere adapts its carbon storage under warming conditions in the longer term



European Carbon Dioxide Surface Flux estimated from SCIAMACHY (and some GOSAT data)

European carbon uptake in gigatonnes of carbon in 2010

Previous estimate without satellite CO₂ 0.4±0.4

Reuter et al. (ACP, 2014) with satellite CO₂

1.0±0.3

maximilian reuter@iup.physik.uni-bremen.de



Anthropogenic CO₂ and NO_x emissions (Reuter et al., 2014, nat. geosci.)

- CO₂ and NO_x are co-emitted species in anthropogenic fossil fuel combustion processes.
- A spatial high-pass filtering method is used to derive co-located regional anomalies ΔXCO₂ and ΔNO₂.
- A statistical relationship between ΔXCO₂ and ΔNO₂ allows to conclude on CO₂ with anthropogenic origin.







Anthropogenic CO₂ and NO_x emissions (Reuter et al., 2014, nat. geosci.)



- We find significantly lower ΔXCO₂ levels at weekends in North America and Europe but not in East Asia.
- The weekend effect of XCO₂ is a tiny signal and this is its **first detection from space**.
- It underlines that the analyzed CO₂ signals originate from anthropogenic activities.



Anthropogenic CO₂ and NO_x emissions (Reuter et al., 2014, nat. geosci.)



- North America and Europe: satellite data show a small downward trend in emissions of both, NO_x and CO₂ albeit associated with a large uncertainty.
- East Asia: CO₂ emissions increased on average at a rate of 9.8%/a but NO_x increased "only" by 5.8%/a, i.e., significantly less compared to CO₂ (increasing CO₂-to-NO_x emission ratio F).



 Interpretation: technology used in East Asia is getting cleaner thus emitting less toxic nitrogen gases per amount of fossil fuel burned.

Methane

- Second most important anthropogenic GHG (directly after CO₂)
- Important precursor of O₃ in global tropospheric Chemistry
- Many anthropogenic and natural sources; large uncertainties



Three Decades of Global Methane Sources and Sinks





Fugitive methane emissions from oil and gas production (Schneising et al., 2014, Earth's Future)



• We analyse **methane enhancements** over the fastest growing production regions in the U.S.



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Fugitive methane emissions from oil and gas production (Schneising et al., 2014, Earth's Future)



 To filter out large-scale seasonal variations or global increase, XCH₄ anomalies are computed by subtracting regional monthly means from the individual measurements.



- The shown **differences of the anomalies** for the period **2009-2011** relative to the period **2006-2008** highlight the changes in atmospheric methane abundance.
- Anomaly differences exhibit increases aligning with the analysed oil and gas fields.

Fugitive methane emissions from oil and gas production (Schneising et al., 2014, Earth's Future)



Bakken

Emission Increase: $990 \pm 650 \text{ ktCH}_4/\text{yr}$ Leakage rate: $10.1 \pm 7.3 \%$ Eagle Ford Emission Increase:

- Emission Increase: $530 \pm 330 \text{ ktCH}_4/\text{yr}$ Leakage rate:
- $9.1 \pm 6.2 \%$
- The emission increase is quantified by a mass-balance approach using the net enhancement relative to the background upwind of the prevailing wind direction and average horizontal boundary layer wind speed.
- The leakage rate is defined as the ratio of the emission increase between 2006-2008 and 2009-2011 divided by the production growth between these two periods.



CarbonSat: Methane @ high latitudes





CarbonSat sun-glint mode allows observation of methane in vulnerable high latitude regions including Arctic sea and shelf areas

The MAMAP instrument



→ Methane and carbon dioxide Airborne Mapper a passive remote sensing instrument using absorption NIR and SWIR spectroscopy



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CarbonSat – ESA EE8 Candidate







CarbonSat's unique contribution at national scales



Methane Leackage from Gas Production

Simulation of XCH₄: Emission rate of = 482 ktCH₄/yr on an area of ca. 35 km x 35 km (*), 5 m/s wind speed, instrument resolution and single measurement precision as below:



39.

39.7

109.8 -109.6 Longitude



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C-MAPExp Campaign Results: Power Plant CO₂



MAMAP Airborne remote sensing and in-situ observations on 18.8.2012: Lignite Coal fired power plant Weissweiler



CO₂ emitting Power Plant Weissweiler

MAMAP aircraft observations:

- Remote sensing data at MAMAP resolution (approx. 100m x 100m) including plume inversion result
- Filtered for instrument inclination angle ≤10°
- Derived emission: 16.15 MtCO₂/yr at the time of measurements
- But what would be detected from space?





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CO₂ emitting Power Plant Weissweiler

What CarbonSat will yield:

MAMAP data "converted" to CarbonSat ovservations:

- Recorded remote sensing data gridded to spatial resolution of approx. 2 km x 2 km
 Including plump
- Including plume inversion result
- Derived emission: 15.7 MtCO₂/yr at the time of measurements





COMEX Campaign (USA) Results: Oil Field CH₄

Airborne remote sensing data (MAMAP) from California, August/Sept. 2014





California Oil Field CH₄

MAMAP interpolated:

What CarbonSat will see at 2x2 km² resolution:





CarbonSat Constellation



SCIANACHY 2002-2012 hunting light and shadows



