Investigating stratospheric changes between 2009 and 2018 with trace gas data from aircraft, AirCores, a global model and a focus on CFC-11

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What we have done

- AirCores were invented by NOAA* and allow the collection of air samples from altitudes of up to >35 km using weather balloons, which is very cost-effective

- We present data from 15 AirCore flights since 2016 and expand the set of measurable trace gases to include the important ozone-depleting substances CFC-11, CFC-12, HCFC-22, H-1211, H-1301 as well as SF_6

- Due to its recent unusual tropospheric trend** we focus on CFC-11 and use runs of the CLaMS global stratospheric model driven with three reanalyses to

  a) compare with the observational record and

  b) derive CFC-11 strat-trop mass fluxes over a 10-year period

*Karion et al., 2010
** Montzka et al., 2018, Rigby et al., 2019

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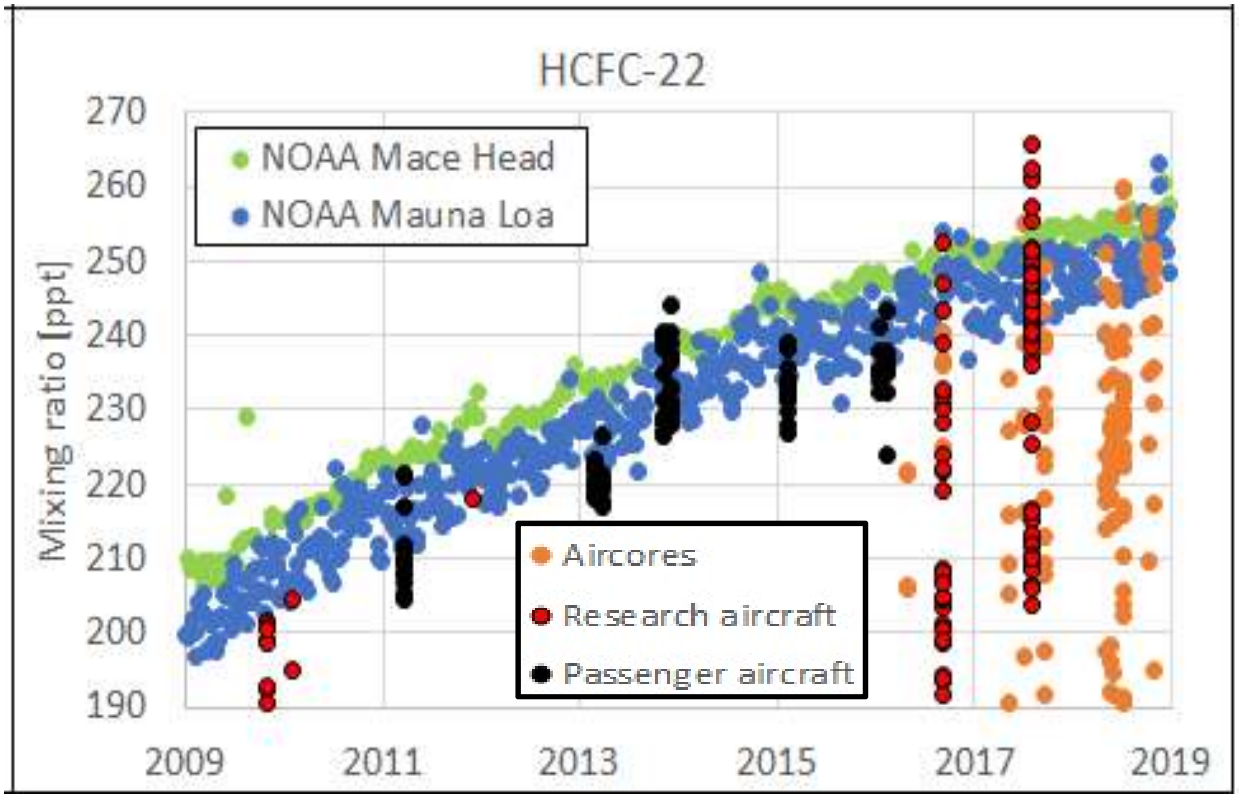
Message 1: Quality assurance

- AirCore data agrees well with NOAA background data from ground-based stations



- Average precisions are slightly worse (e.g., 0.9 vs. 0.4 % for CFC-11) due to the amount of air retrieved by more than an order of magnitude smaller

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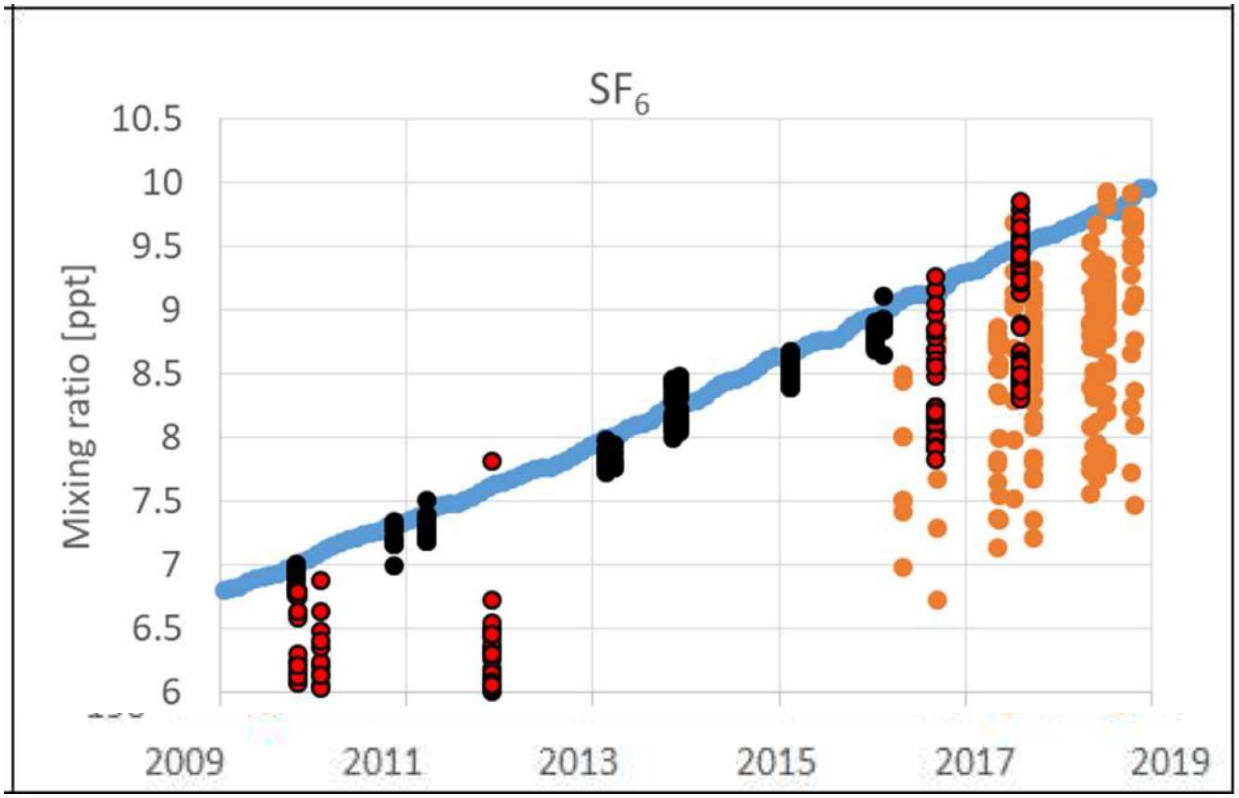

- SF_6 data demonstrates the increased temporal and spatial density of stratospheric observations - and can be used, alongside other gases like C_2F_6 and HFC-125, to derive average stratospheric residence times (Ages of Air, AoAs)

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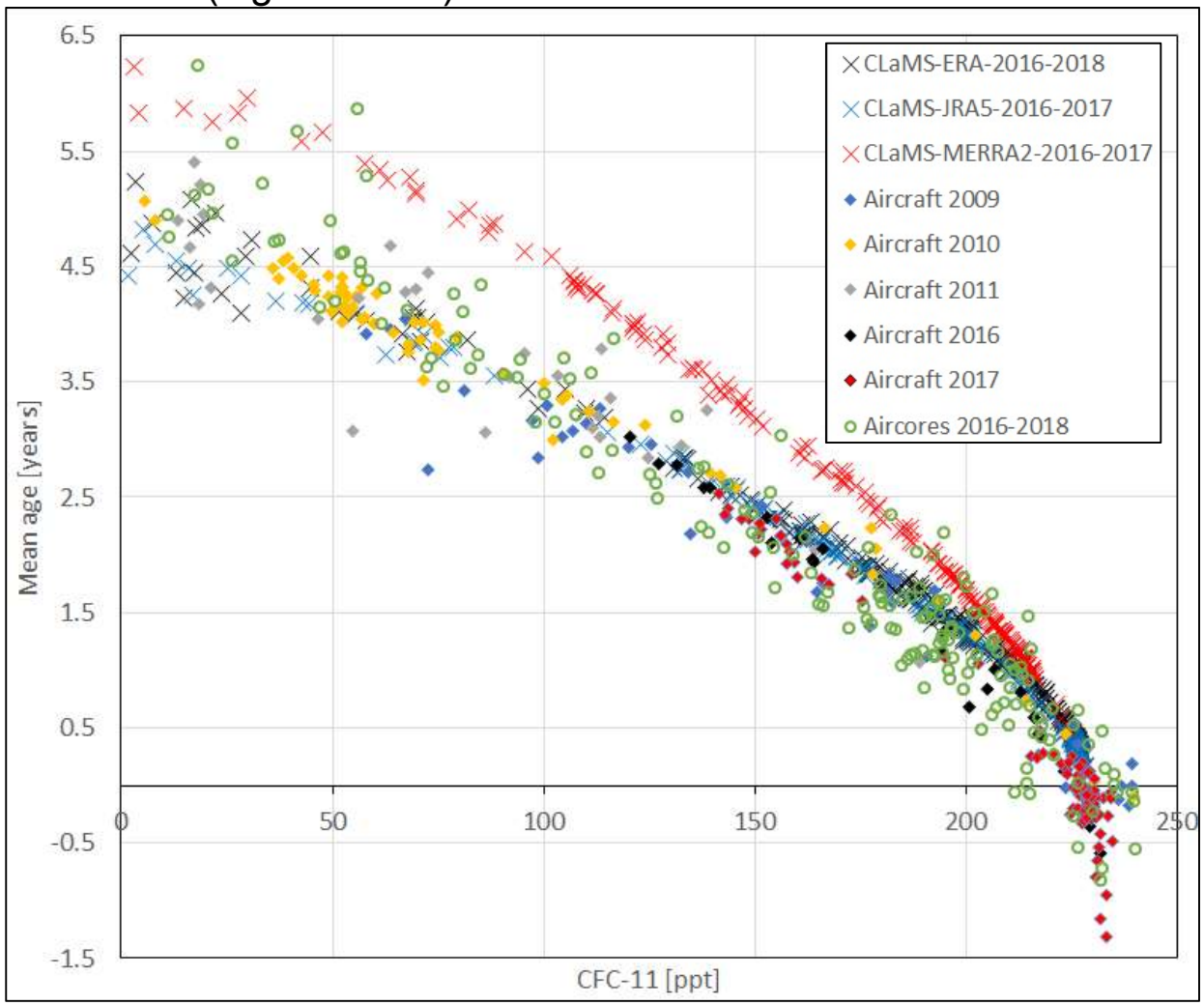
Message 2: Model comparisons

- Correlations of mixing ratios and Ages-of-air from AirCore, aircraft and CLaMS model data driven by JRA-55 and ERA-Interim agree well, but not the model run driven by MERRA-2 (higher AoAs)



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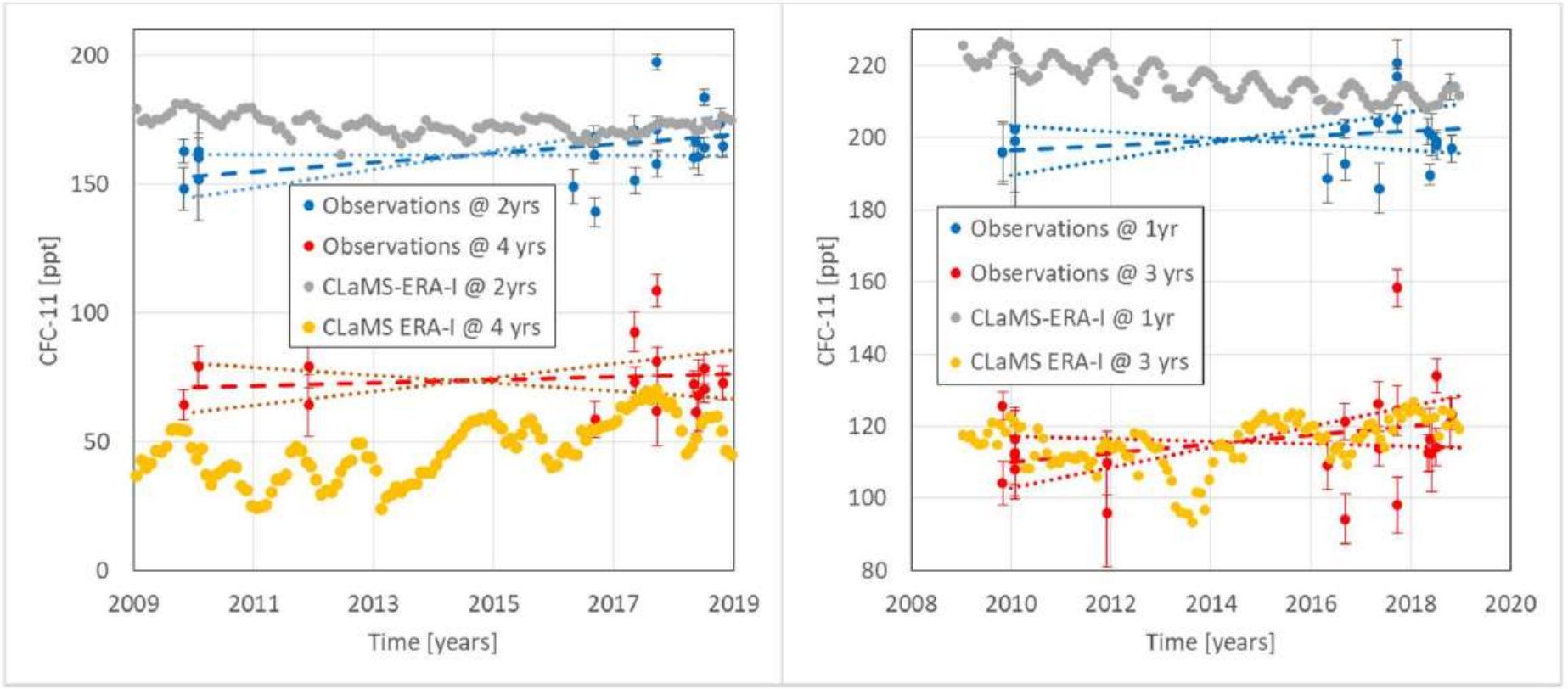
Message 3: Model comparisons

- When determining trends of CFC-11 mixing ratios at certain AoA levels they do not agree well (apart from ERA-I @ 3 yrs AoA) with the trends derived from model runs
- On average we find increases of 3 to 10 % between 2009 and 2018, whereas tropospheric CFC-11 decreased by 6 % during that period



→ This points towards stratospheric circulation changes that could transport increased amounts of CFC-11 back into the troposphere

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Message 3: Model comparisons

- Mhmm, ok, but there's not a lot of observations and they're pretty scattered – so is this proposed stratospheric change actually likely?

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- Mhmm, ok, but there's not a lot of observations and they're pretty scattered – so is this stratospheric change actually significant?

CFC-11/AoA	**1 year**	**2 years**	**3 years**
Slope obs (ppt/year)	0.69	1.77	1.25
Uncertainty (ppt/year)	1.54	1.81	1.60
Trend (%/decade)	3.2	10.4	10.2
CFC-12			
Slope obs (ppt/year)	-1.96	-0.45	-0.38
Uncertainty (ppt/year)	1.90	2.20	2.52
Trend (%/decade)	-3.6	-0.95	-0.93
HCFC-22			
Slope obs (ppt/year)	6.15	6.16	5.98
Uncertainty (ppt/year)	0.15	0.14	0.14
Trend (%/decade)	30.5	33.4	36.0
H-1211			
Slope obs (ppt/year)	-0.031	0.000	0.013
Uncertainty (ppt/year)	0.008	0.008	0.007
Trend (%/decade)	-9.0	0.2	9.1

2 sigma

troposphere: -6 %

troposphere: -6 %

troposphere: +25 %

troposphere: -20 %

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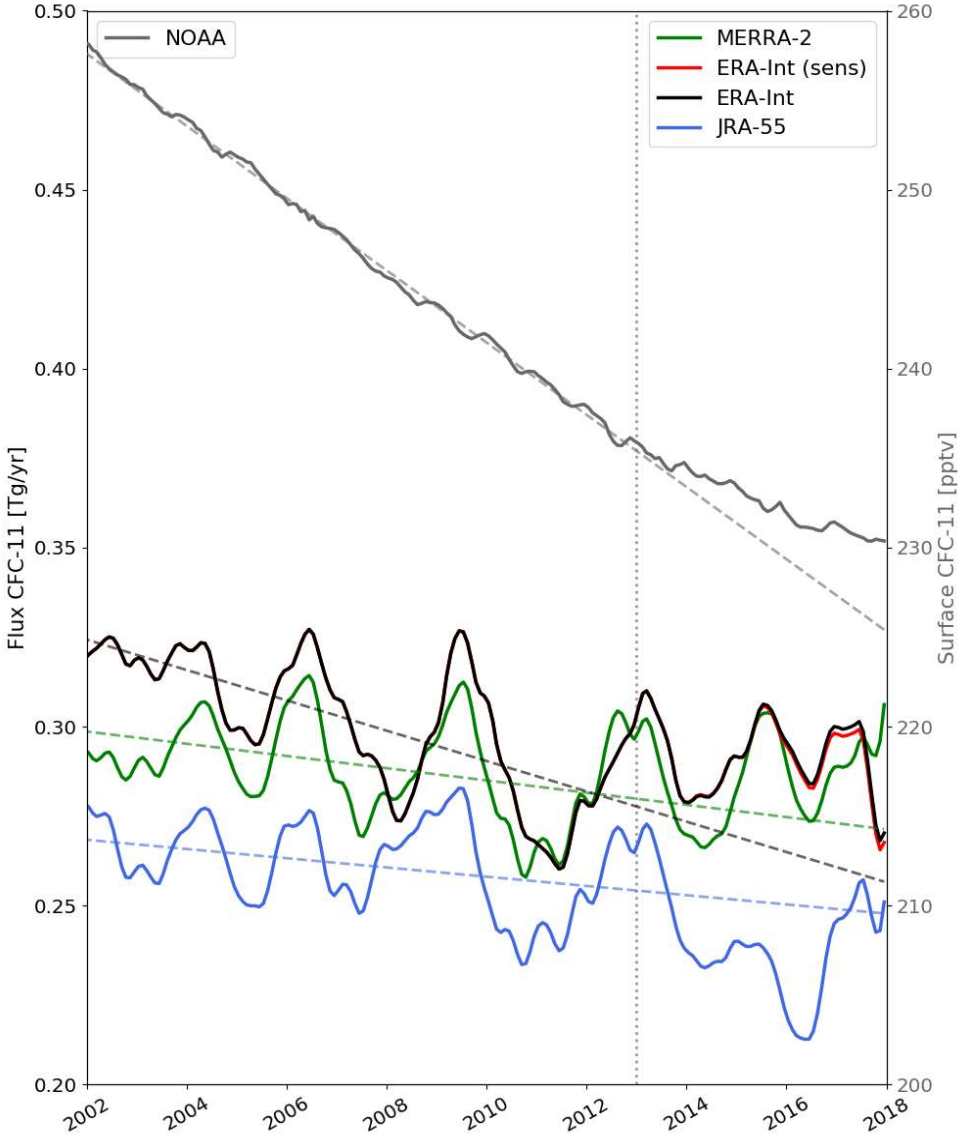
Message 3: Model comparisons

- Overall message: The mixing ratio trend between 2009 and 2018 at certain AoAs in the stratosphere is in the right direction to explain a part of the increased emission signal – but observations are sparse in both space and time

- Still, it demonstrates the potential use of AirCore-based data

- Remaining question: Can we use the CLaMS model data to look at changes of the CFC-11 stratosphere-troposphere mass flux between 2009 and 2018?

Message 4: CFC-11 strat-trop mass fluxes from 3 reanalyses



- Montzka et al., 2018: Stratospheric changes might explain up to 50 % of the tropospheric CFC-11 emission signal
- The CLaMS runs driven by the ERA-Interim, JRA-55, and MERRA-2 reanalyses give very different answers in terms of the amount of CFC-11 transported back to the troposphere - and could explain anything from -3 % to +270 % of the new tropospheric emission signal after 2012

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Conclusions

- The AirCore technique can be used to measure halogenated trace gases in the stratosphere (down to mixing ratios of well below 1 ppt)
- Currently available global meteorological reanalyses are insufficient to constrain the stratospheric part of the CFC-11 budget
- Atmospheric observations are also still too sparse and infrequent to help with this quantification, but do offer qualitative clues
- For more details please see our paper in ACPD:

https://www.atmos-chem-phys-discuss.net/acp-2020-62/

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