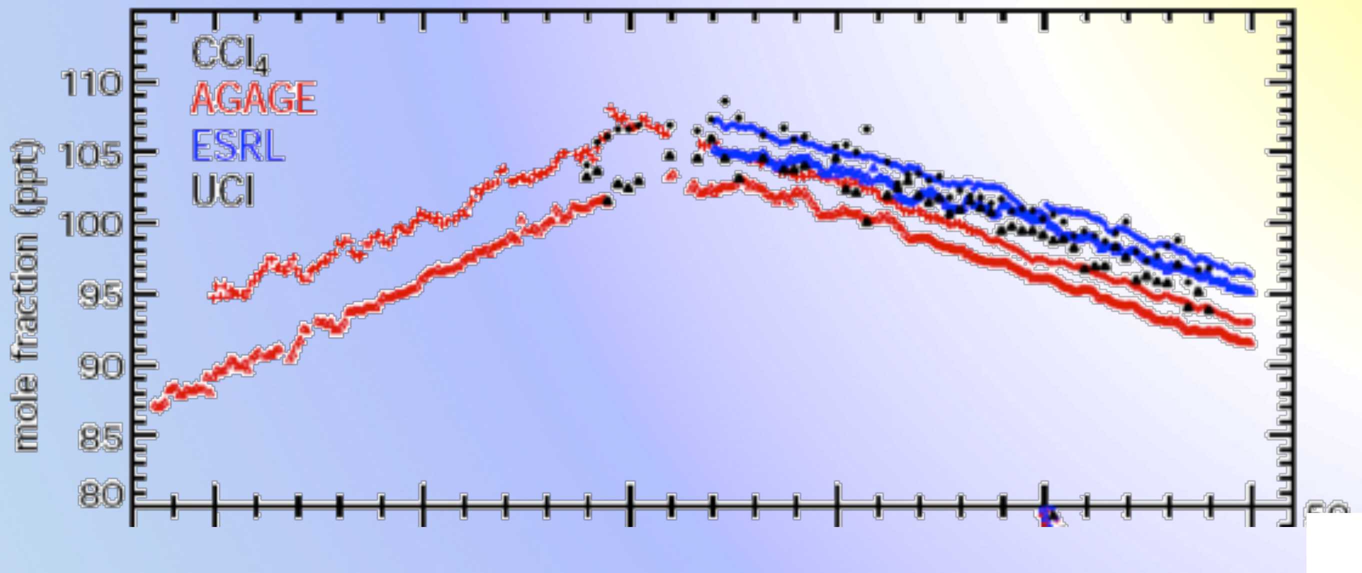
The "lifetime" problem:

The Executive Summary of the 2010 WMO/UNEP Ozone Assessment cites lifetime problems and problems associated with lifetimes:

- "Evidence is emerging that lifetimes for some important ODSs (e.g., CFC-11) may be somewhat longer than reported in past assessments. In the absence of corroborative studies, however, the CFC-11 lifetime reported in this Assessment remains unchanged at 45 years. Revisions in the CFC-11 lifetime would affect estimates of its global emission derived from atmospheric changes and calculated values for Ozone Depletion Potentials (ODPs) and best-estimate lifetimes for some other halocarbons."
- "Carbon tetrachloride (CCl_4) tropospheric abundances have declined less rapidly than expected. Emissions derived from data reported to the United Nations Environment Programme (UNEP) are highly variable and on average appear smaller than those inferred from observed abundance trends. Although the size of this discrepancy is sensitive to uncertainties in our knowledge of how long CCl4 persists in the atmosphere (its "lifetime"), the variability cannot be explained by lifetime uncertainties."
- "A stronger BDC would:
  - decrease the abundance of tropical lower stratospheric ozone,
  - increase poleward transport of ozone, and
  - could reduce the atmospheric lifetimes of long-lived ODSs and other trace gases."

M. Ko, P. Newman, S. Reimann, S. Strahan

Issues of global data series of CCl_4



Stable interhemispheric gradient: sources in N-Hemisphere?

Global sources: 70 Gg (lifetime: 26 years)

Lifetime uncertain (ocean sink, soil sink)

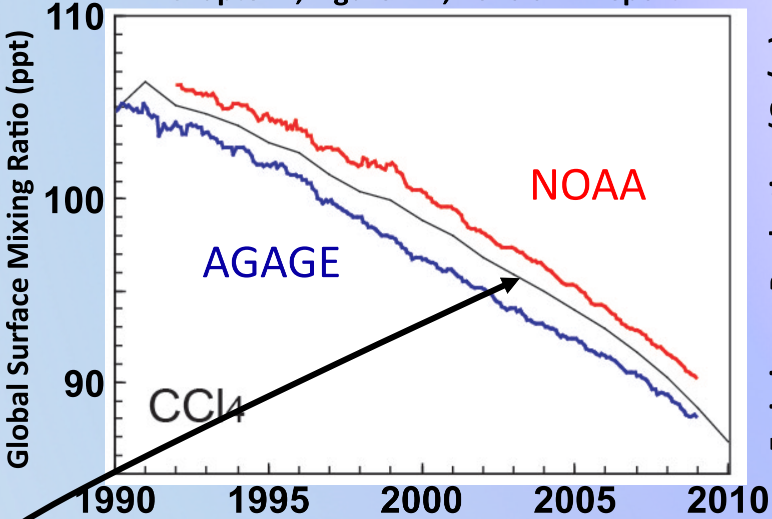
Summed regional sources from inverse modelling : ~20 Gg

Ozone Assessment (2007):
Overall the budget of CCl_4 remains poorly understood.

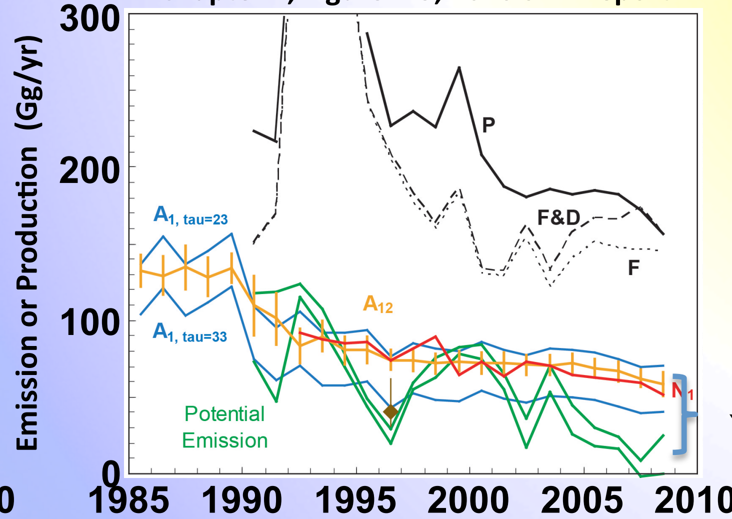
M. Ko, P. Newman, S. Reimann, S. Strahan

The Carbon Tet (CCl_4) puzzle

Chapter 1, Figure 1-1, 2010 SAP Report



Chapter 1, Figure 1-5, 2010 SAP Report



$$\frac{\partial \chi}{\partial t} = E - \frac{\chi}{\tau}$$

- Carbon tetrachloride (CCl_4) continues to decrease in the atmosphere
- … but its abundance is not consistent with reported emissions and known lifetimes.
- Emissions derived from data reported to UNEP are highly variable and on average appear smaller than those inferred from observed trends.
- Discrepancy (~ 40 Gg per year):
  - Cannot be explained by the lifetime. CCl_4 lifetime, τ = 28±5 years.
  - Errors in reporting, or errors in analysis of reported data, possible illegal prod.
  - Unknown sources or poorly estimated sinks

M. Ko, P. Newman, S. Reimann, S. Strahan

The "lifetime" problem

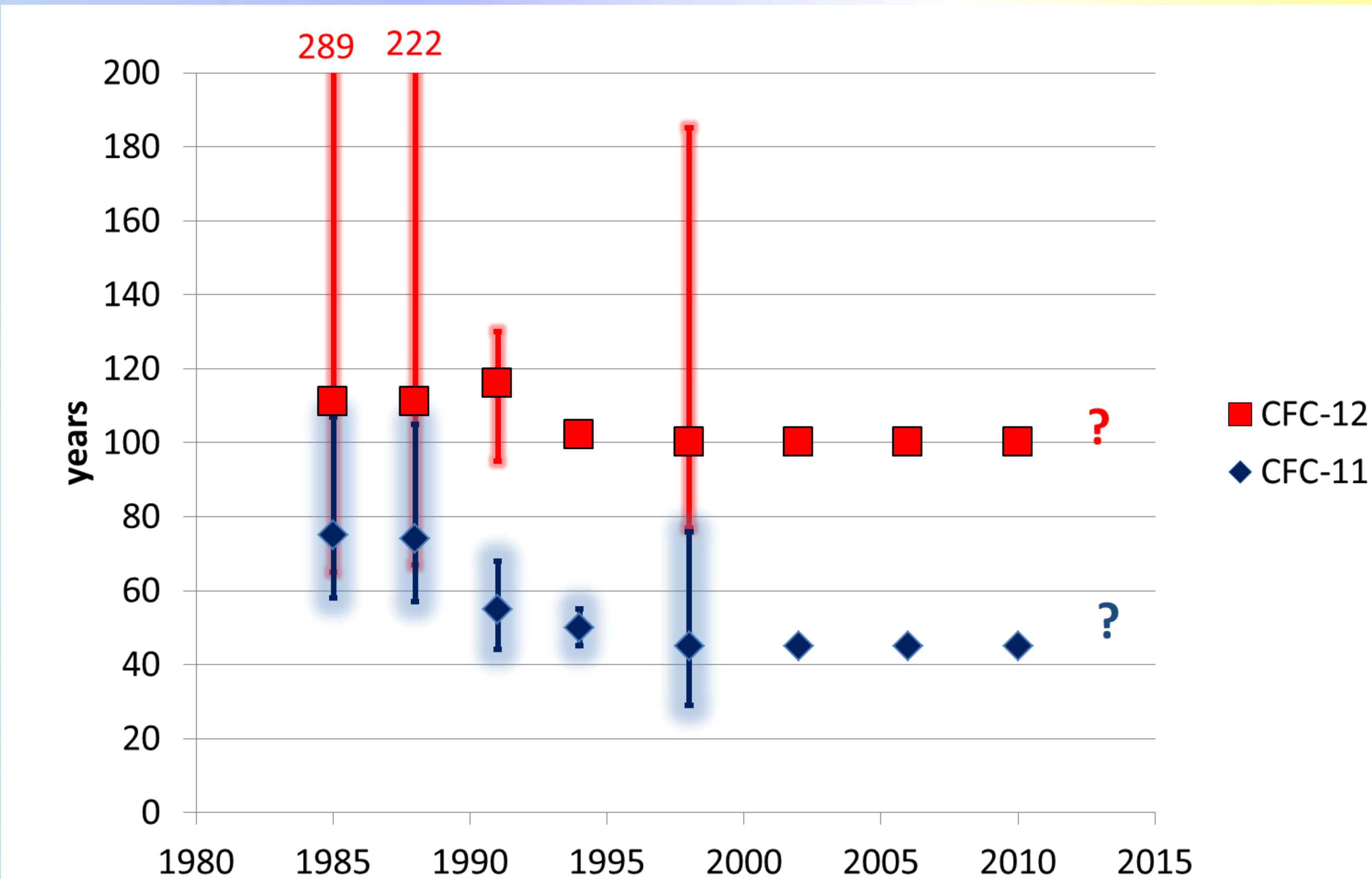
- Lifetimes of many long-lived atmospheric trace gases may be seriously error. At GSFC we have estimated lifetimes using met fields from the GEOS5-GCM coupled chemistry model in an offline GMI simulation. **Our current best models are yielding lifetimes that differ from values estimated from older models and observations.**

Compound	GMI (yrs)	WMO 2007 (yrs)
CFC-11	~61	45
CFC-12	106	100
CH_3CCl_3	7.5	5.0
HCFC-22	17.1	12
CCl_4	59 *	26
CFC-113	92.5	85
CH_4	14.5	–
N_2O	120.	114

*The GMI CTM has no ocean sink for CCl_4 so the GMI lifetime is too long.

M. Ko, P. Newman, S. Reimann, S. Strahan

History of lifetimes in previous ozone assessments



M. Ko, P. Newman, S. Reimann, S. Strahan

Started lifetime re-evaulation within SPARC

Stratospheric **P**rocesses **A**nd their **R**ole in **C**limate

Scope of the re-evaluation

- Estimate the numerical values for lifetimes
- Estimate the uncertainties for numerical values for lifetimes
- Assess the influence/use of different lifetime definitions (e.g. steady-state /instantaneous lifetimes)
- Assess lifetime changes associated with the changing climate

Lifetimes to reevaluate

	Compound	Formula	Lifetime (yrs)
Priority 1:			
1.	CFC-11	CCl_3F	45
2.	CFC-12	CCl_2F_2	100
3.	Carbon Tetrachloride	CCl_4	26
4.	Methyl Chloroform	CH_3CCl_3	5.0
5.	HCFC-22	$CHClF_2$	12
6.	Nitrous oxide	N_2O	114
7.	Methane	CH_4	8.7* (lifetime) 12.0* (pulse decay
Priority 2:			
8.	Halon-1211	$CBrClF_2$	16
9.	Halon-1301	$CBrF_3$	65
10.	CFC-113	CCl_2FCClF_2	85
11.	CFC-115	CF_3CClF_2	1020
12.	HFC-134a	CH_2FCF_3	13.4
13.	HFC-143a	CF_3CH_3	47.1
14.	HFC-23	CHF_3	222
Priority 3:			
15.	CFC-114	$CClF_2CClF_2$	190
16.	HCFC-141b	CH_3CCl_2F	9.2
17.	HCFC-142b	CH_3CClF_2	17.2
18.	Methyl Chloride	CH_3Cl	1.0
19.	Methyl Bromide	CH_3Br	0.8
20.	Halon-1202	CBr_2F_2	2.9
21.	Halon-2402	$CBrF_2CBrF_2$	20.
21.	HFC-125	CHF_2CF_3	28.2

M. Ko, P. Newman, S. Reimann, S. Strahan

Structure of the lifetime re-evaluation

- **Executive Committee:** Malcolm Ko, Susan Strahan, Stefan Reimann, *Paul Newman*
- **Chapter 1: importance of global lifetimes, history of lifetimes**
  - Lead authors: Executive Committee
- **Chapter 2: Theory of estimating lifetimes using models and observations**
  - Lead authors: Alan Plumb, Richard Stolarski
- **Chapter 3: Update on kinetic data that determined lifetimes (cross sections etc.)**
  - Lead authors: James Burkholder, Wahid Mellouki
- **Chapter 4: Inferred lifetimes from observed trace-gas distributions**
  - Lead authors: Andreas Engel, Elliot Atlas
- **Chapter 5: Model estimates of lifetimes**
  - Lead authors: Martyn Chipperfield, Qing Liang
- **Chapter 6: Summary**
  - Lead authors: chapter leads and executive committee

Timetable

2011

Feb. 2011, Presentation to SPARC SG
Feb. 2011, Comments solicited from scientific community on scope
Apr. 2011, Scope redefined, author teams formed.
May 2011, Chapter outlines drafted
Jul. 2011, Begin of model simulations

Dec. 2011, model simulation completed.

2012

May. 2012, 1st drafts complete; circulated for internal review.

Sep. 2012, 2nd drafts complete, start peer review.

Dec. 2012, 3rd draft complete.

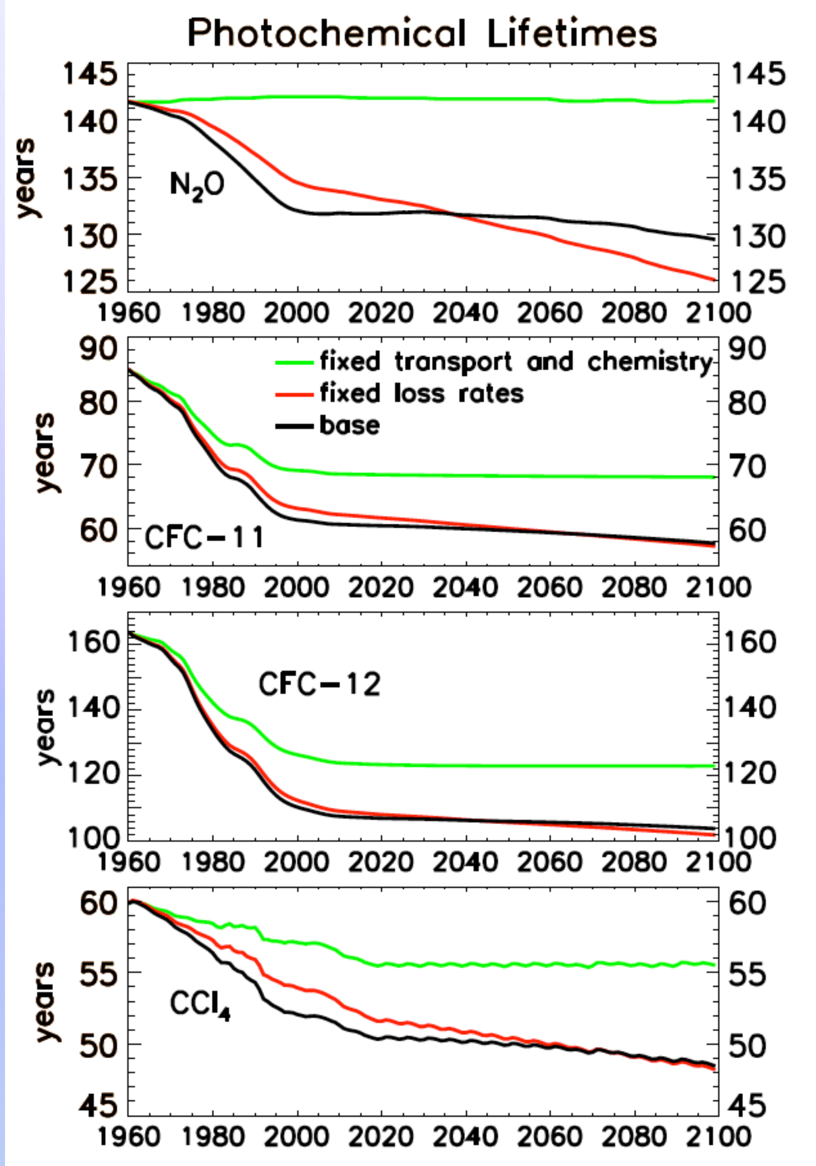
Jan. 2013, Open meeting with reviewers
Feb. 2013, Final draft, including Executive Summary,

Apr. 2013, Document released

M. Ko, P. Newman, S. Reimann, S. Strahan

The lifetime of a specie changes with time

- Fleming et al. (ACPD, 2011) Figure 13
- **Black lines show** lifetimes for N_2O, CFC-11, CFC-12, and CCl_4
- **Red lines** are with a fixed loss rate for 1960 values.
- Green lines use chemistry and transport fixed at 1960 values illustrating the effect of the changing atmospheric burden.



M. Ko, P. Newman, S. Reimann, S. Strahan

Chapter 2

Alan Plumb	Massachusetts Institute of Technology	USA
Rich Stolarski	Johns Hopkins University	USA
Andreas Engel	University of Frankfurt	Germany
Michaela Hegglin	University of Toronto	Canada
Qing Liang	Universities Space Research Association	USA
Michael Prather	University of California, Irvine	USA
John Pyle	University of Cambridge	UK
Michael Volk	University of Frankfurt	Germany
Darryn Waugh	Johns Hopkins University	USA

Chapter 3

• **Jim Burkholder**	NOAA	USA
• **Wahid Mellouki**	ICARE/CNRS	France
• Eric Fleming	Space Systems and Applications, Inc.	USA
• Christian George	CNRS/Université Claude Bernard Lyon 1	France
• Dwayne Heard	University of Leeds	UK
• Charley Jackman	NASA Goddard Space Flight Center	USA
• Matthew Johnson	University of Copenhagen	Denmark
• Mike Kurylo	Universities Space Research Association	USA
• Tim Wallington	Ford Corporation	USA

Chapter 4

Elliot Atlas	University of Miami	USA
Andreas Engel	University of Frankfurt	Germany
Peter Bernath	University of York	UK
Harald Bönisch	University of Frankfurt	Germany
Lambert Kuijpers	Eindhoven University of Technology	Netherlands
Johannes Laube	University of East Anglia	UK
Ken Minschwaner	New Mexico Inst. of Mining & Technology	USA
Steve Montzka	NOAA	USA
Simon O'Doherty	University of Bristol	UK
Ron Prinn	Massachusetts Inst. of Technology	USA
Matt Rigby	Massachusetts Inst. of Technology	USA
Sue Schauffler	National Center for Atmospheric Research	USA
Michael Volk	University of Frankfurt	Germany
Shari Yvon-Lewis	Texas A&M University	USA

M. Ko, P. Newman, S. Reimann, S. Strahan

Chapter 5

• **Martyn Chipperfield**	University of Leeds	UK
• **Qing Liang**	Universities Space Research Association	USA
• Slimane Bekki	LATMOS/IPSL/CNRS	France
• Anne Douglass	NASA Goddard Space Flight Center	USA
• Doug Kinnison	National Center for Atmospheric Research	USA
• David Plummer	University of York	Canada
• Michael Prather	University of California, Irvine	USA
• Björn-Martin Sinnhuber	University of Bremen	Germany