re-evaluation of lifetime of ozone-depleting substances within SPARC

Scope of the re-evaluation

1) Estimate the numerical values for lifetimes of ODS

2) Estimate the uncertainties for numerical values for lifetimes

3) Assess the influence/use of different lifetime definitions (e.g. steady-state /instantaneous lifetimes)

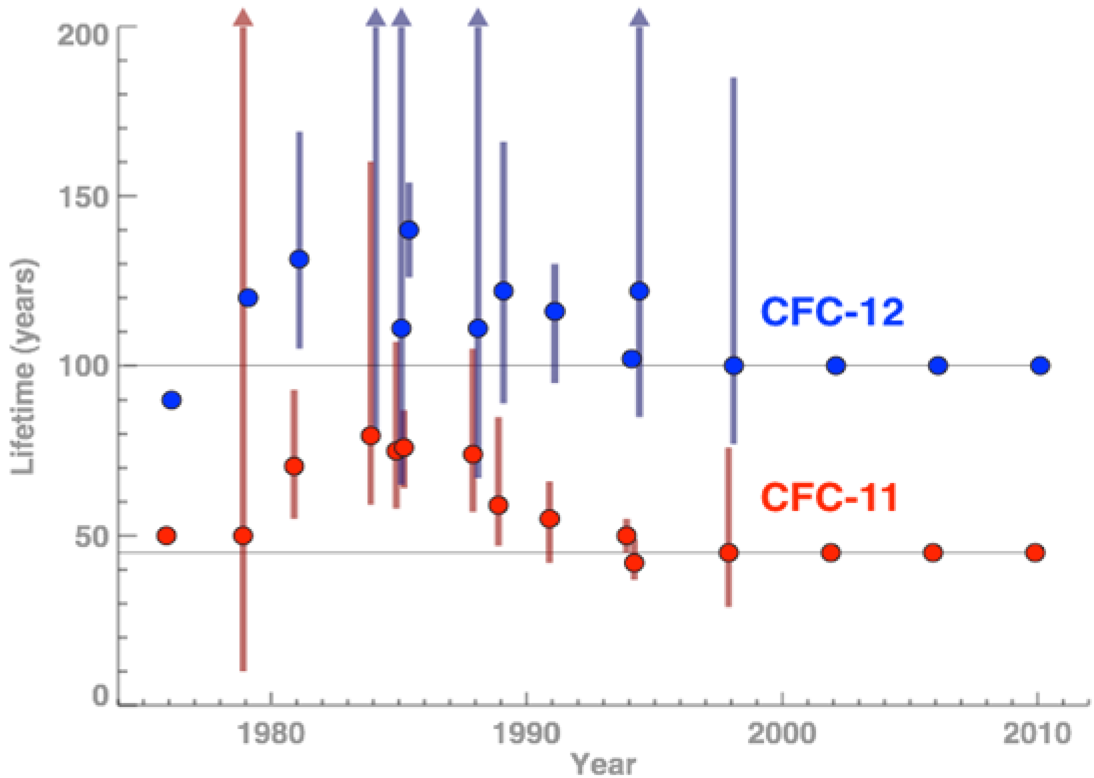
4) Assess lifetime changes associated with the changing climate

The Motivation for the lifetime re-evaulation

Extract from the Executive Summary of the 2010 WMO/UNEP Ozone Assessment

"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**. (…)



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

Examples from the 1998 ozone assessments

CFC-11	45 years	(29-76)	Volk (1997) Tracer-ratio stratosphere + Cunnold (1997) 12-box model
CFC-12	100 years	(77-185)	

Table 1-4. Model-calculated steady-state lifetimes in years.

Species	AER[a]	GSFC[a]	CSIRO[a]	Harvard 2-D[a]	LLNL[a]	SUNY -SPB[a]	UNIVAQ 2-D[a]	LaRC 3-D[a]	GISS-UCI 3-D[b]	MIT 3-D[c]
N_2O	109	130	117	122	106	125	122	175	113	124
CCl_3F (CFC-11)	47	61	53	68	49	49	44	57	35	42
CCl_2F_2	92	111	100	106	92	107	105	149	90	107

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

Structure of the lifetime re-evaluation

Steering Committee:
Malcolm Ko, Paul Newman, Stefan Reimann, Susan Strahan

Chapter 1: importance of global lifetimes, history of lifetimes
Lead authors: *steering commitee*

Chapter 2: Theory of estimating lifetimes using models and observations
Lead authors: *Alan Plumb/Richard Stolarski*

Chapter 3: Update on kinetic/photochemical data that determine lifetimes
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*

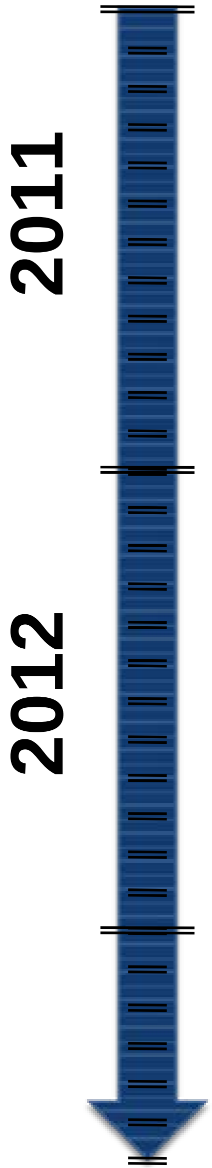
Substances whose lifetime are re-evaluated

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

Compound		Lifetime (yrs)
Priority 3:		
15.	CFC-114	190
16.	HCFC-141b	9.2
17.	HCFC-142b	17.2
18.	Methyl Chloride	1.0
19.	Methyl Bromide	0.8
20.	Halon-1202	2.9
21.	Halon-2402	20
22.	HFC-32	5.7
23.	HFC-125	28.2
24.	HFC-152a	1.5
25.	HFC-227ea	38.9
26.	HFC-245fa	7.7

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

Timetable



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

Jun 2011, Begin of model simulations

Dec. 2011, model simulation completed.

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

June. 2012, 1^{st} review meeting, US East Coast

Sep. 2012, 2^{nd} drafts complete, start peer review.

Dec. 2012, 3^{rd} 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

First results: Chapter 2 (Plumb/Stolarski et al.)
Theory of estimating lifetimes using models and observations

I. Lifetime definition(s)

- Simple residence time vs. coupled lifetimes (e.g. N_2O or CH_4)

II. Loss processes

- Stratospheric loss: photolyis, reaction with OH, O(1D)
- Tropospheric loss: reaction with OH (and evaluation of changes)
- Loss at the surface: soils and oceans.

III. Steady-state lifetimes

- Calculation of steady-state lifetimes from models (e.g. burden over loss)

IV. Instantaneous Lifetimes

- Use mixing ratios, mixing ratio changes and emission inventories to derive instantaneous lifetimes.
- Steady-state lifetime is when the burden is not changing

V. Relative Lifetimes

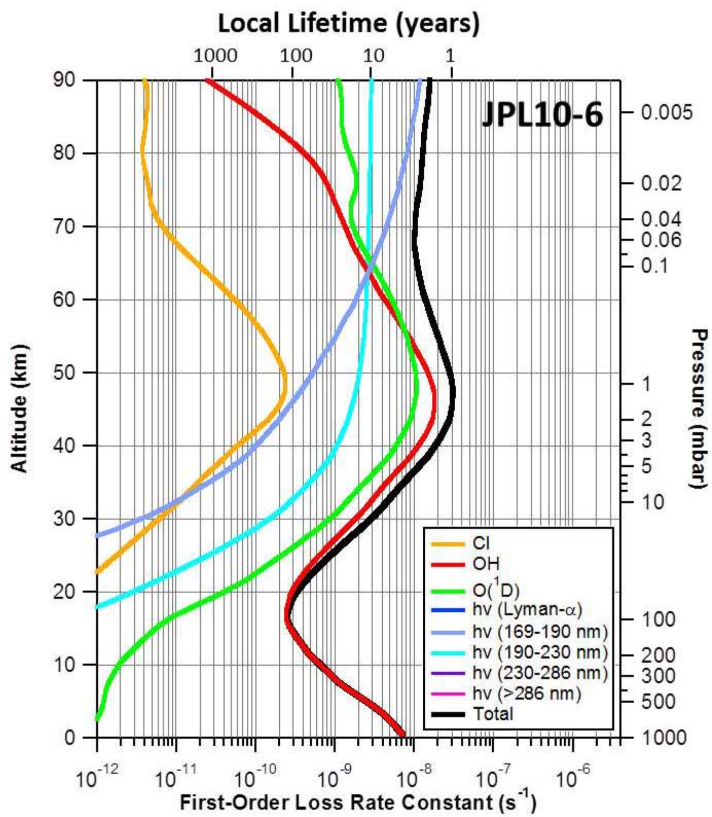
- Tracer-tracer ratio to derive relative lifetimes

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

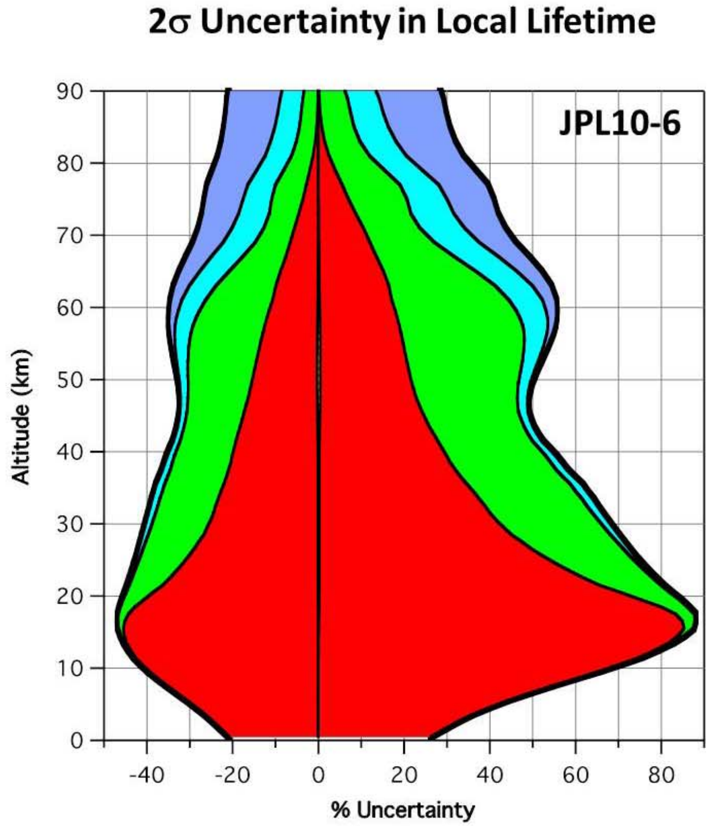
First results: Chapter 3 (Burkholder/Mellouki et al.)
Update on kinetic/photochemical data that determine lifetimes

Preliminary: Refine Previous JPL and IUPAC Data evaluations

Example: HCFC-22 CHF_2Cl



Large Uncertainties throughout the atmosphere !



T-dep of OH reaction is a critical factor

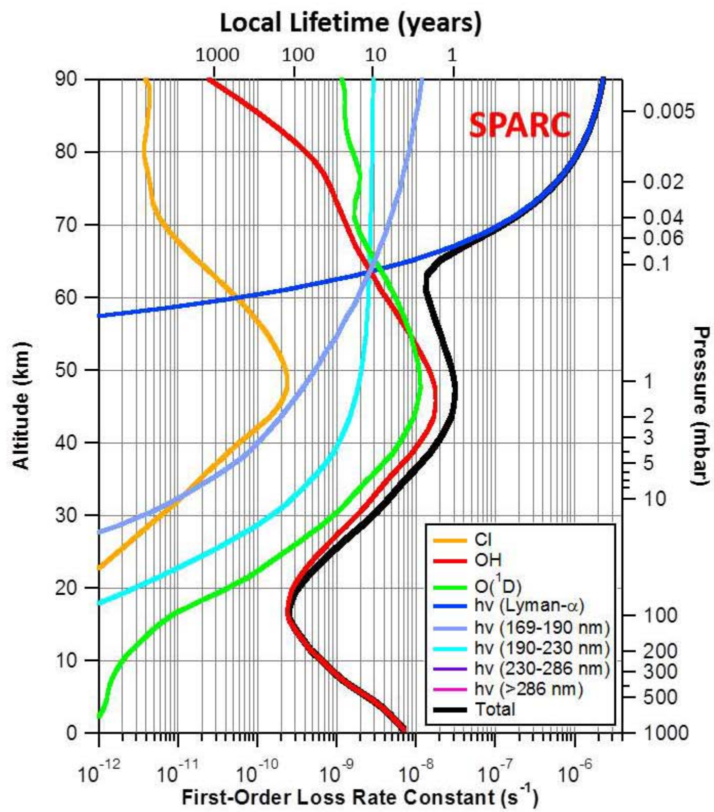
M. Ko, P. Newman, S. Reimann ,S. Strahan

First results: Chapter 3 (Burkholder/Mellouki et al.)

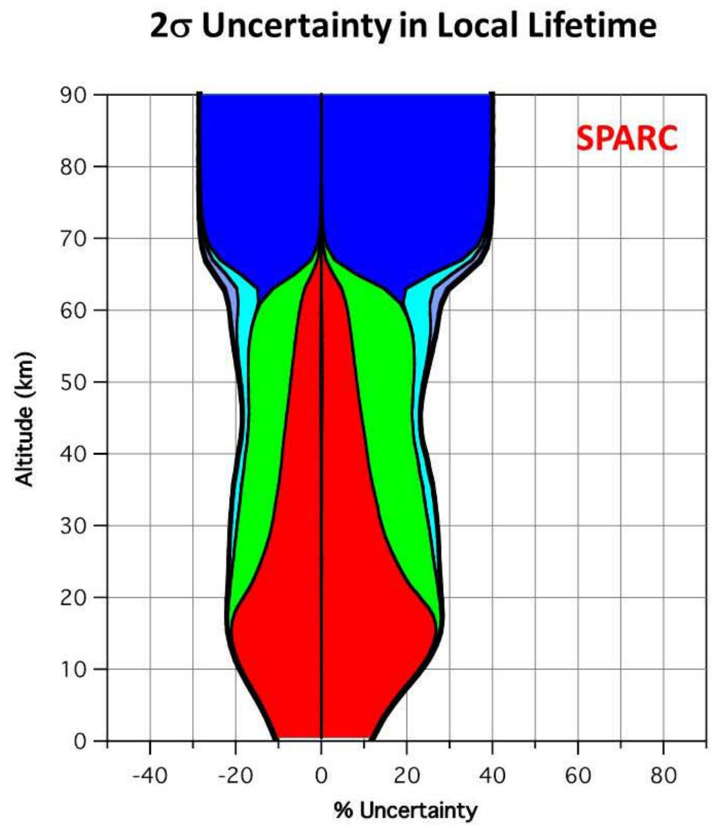
Update on kinetic/photochemical data that determine lifetimes

Preliminary: Refine Previous JPL and IUPAC Data evaluations

Example: HCFC-22 CHF_2Cl



SPARC: Lyman-α photolysis included



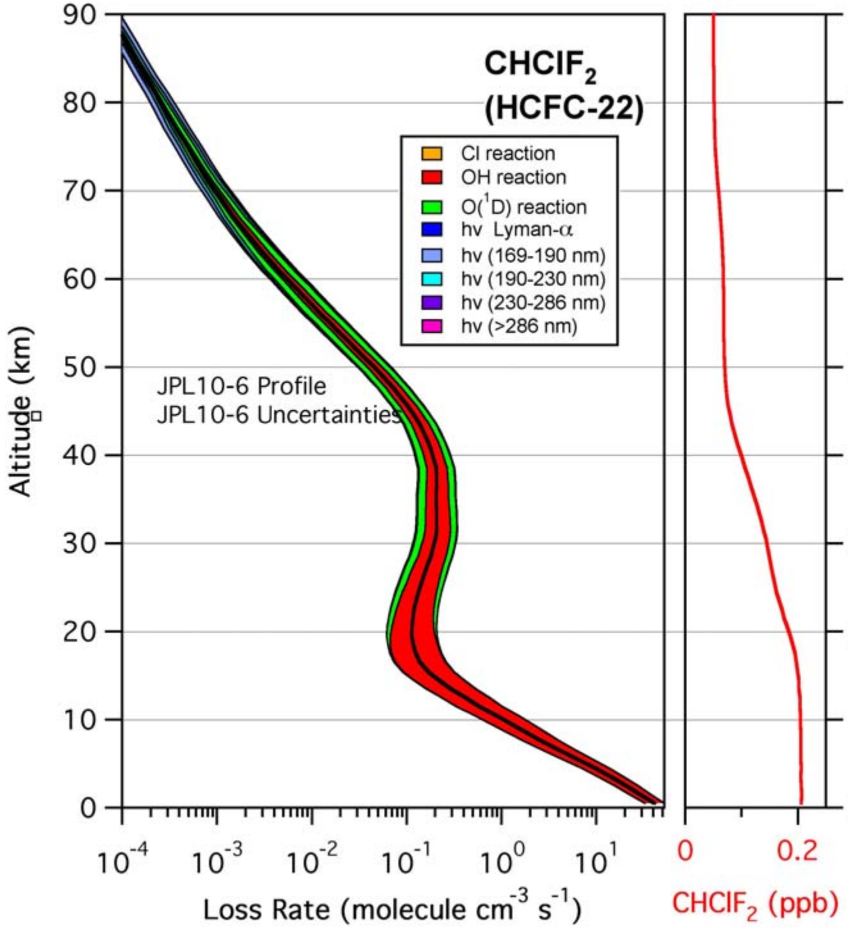
Recommended overall and individual uncertainties evaluated and revised

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

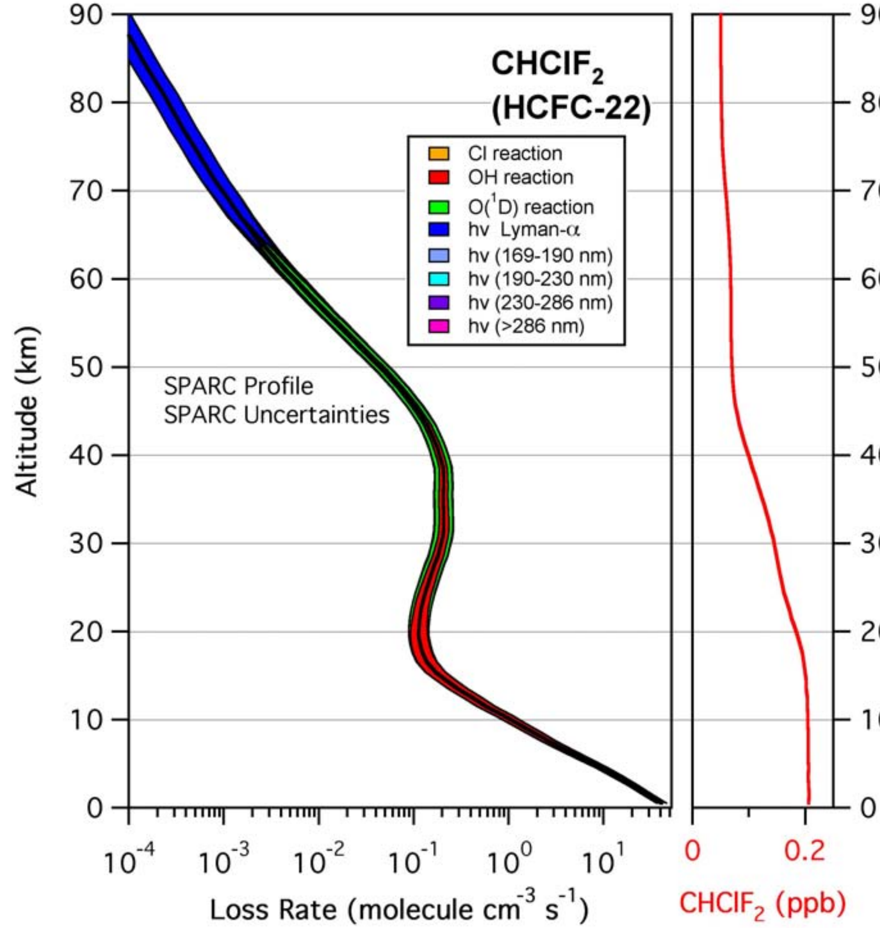
First results: Chapter 3 (Burkholder/Mellouki et al.)
Update on kinetic/photochemical data that determine lifetimes

Preliminary: Resulting smaller uncertainty for loss rates

JPL10-6



SPARC



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

First results: Chapter 4 (Engel/Atlas et al.)

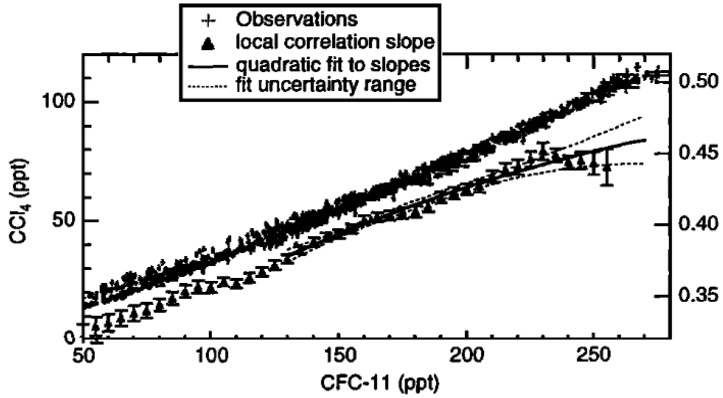
Inferred lifetimes from observed trace-gas distributions

3 different approaches

1. box-models using in-situ data

emissions + Mixing ratios rate of changes → lifetime

2. Tracer ratio at the tropopause (Volk et al., 1997)



3. Satellites

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

First results: Chapter 4 (Engel/Atlas et al.)
Inferred lifetimes from observed trace-gas distributions

1. box-models using in-situ data

Long-lived substances: **12-box model using AGAGE and NOAA observations**

➔ **CFC-11 lifetime could be around 10% higher than before**
➔ **other compounds: ~ existing values**

Short-lived ODSs: **box-modelling**

CH3Br (0.8 yr)/CH3Cl (~1.0 yr) ✓

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

First results: Chapter 4 (Engel/Atlas et al.)

Inferred lifetimes from observed trace-gas distributions

2. Tracer ratio at the tropopause (Volk et al., 1997)

re-evaluation of old and new campaigns

1)ABSOULTE: Tracer-mean age relations: → lifetimes are highly uncertain

2) RELATIVE: Tracer-tracer relations (with slightly higher CFC-11lifetime)

→CFC-12: 100 years ⇒

→CFC-113 : 85 years ⇒

→CCl4: 35 years ⇑

3. Satellites (Stratosphere):

ABSOULTE : CFC-11 ~38-60 years

RELATIVE:

→**CFC-12:** 100 years ⇑

→**CCl4:** 35 years ⇑

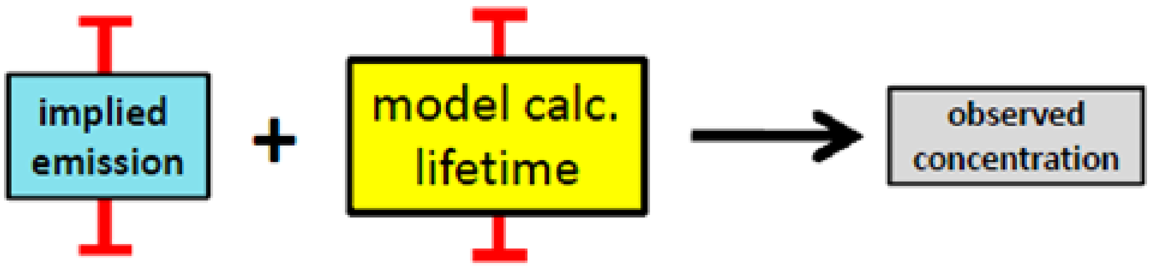
M. Ko, P. Newman, S. Reimann ,S. Strahan

First results: Chapter 5 (Chipperfield/Liang et al.)
Model estimates of lifetimes

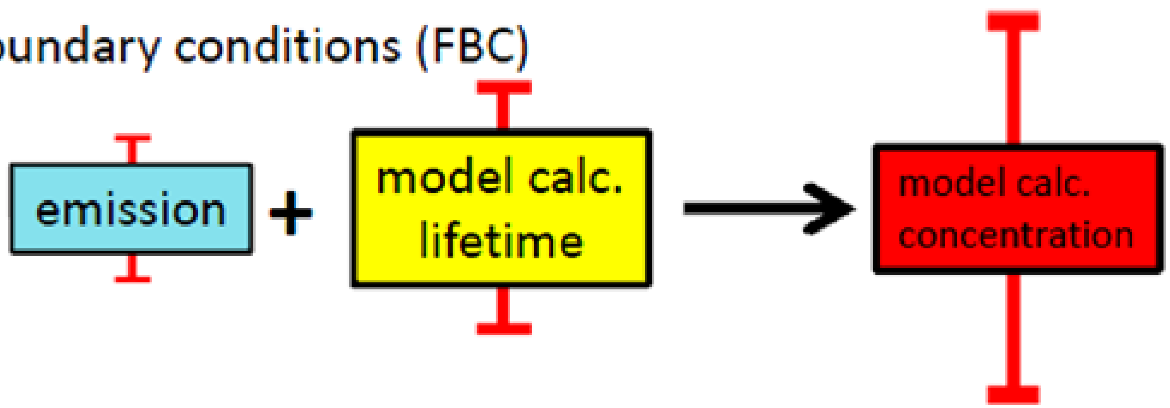
Chemistry-Climate Models (CCMs) to calculate lifetimes

2 different approaches

- Mixing-ratio boundary conditions (MBC)



- Flux boundary conditions (FBC)



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

First results: Chapter 5 (Chipperfield/Liang et al.)
Model estimates of lifetimes

Model Runs

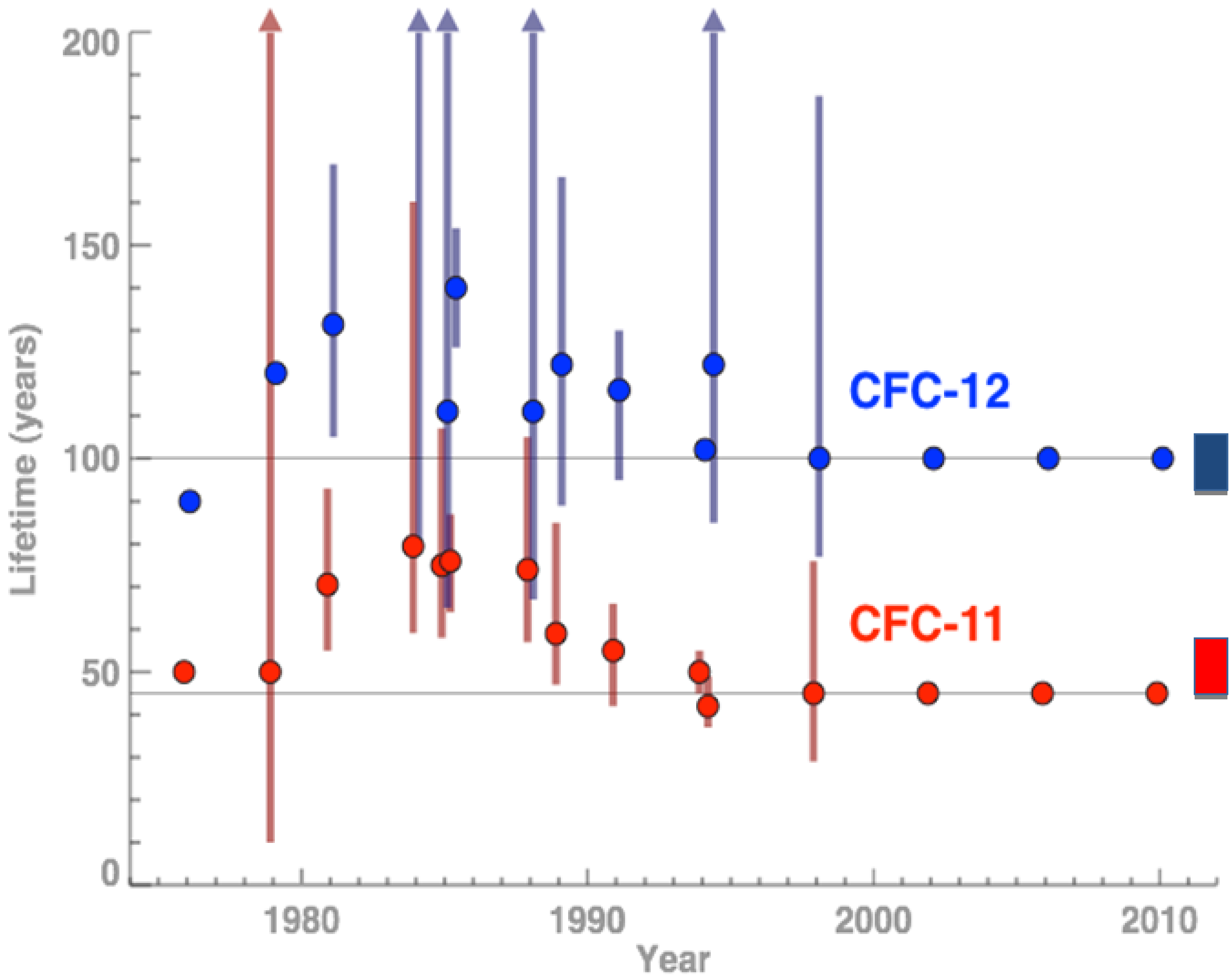
Run	Period	Length	Input	Notes
TS2000	Time slice 2000	~20 years	MBC	For timeslice runs MBC will be in equilibrium.
TS2100	Time slice 2100	~20 years	MBC	Compare how lifetimes change wrt 2000.
“REF-C1”	Transient 1960-2010	50+ years	MBC	Like CCMVal REF-B1

Results first draft
5 models (four 3-D and one 2-D) ➔ 8-10 models for the second draft.

- ➔ CFC-11: 45 years
- ➔ CFC-12: 100 years
- ➔ CFC-113 : 85 years
- ➔ CCl4: 35 years

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

History of lifetimes in previous ozone assessments



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

Conclusion

- First in-depth evaluation of ODS lifetimes since 15 years.
- Discrepancies getting more obvious with less emissions
- Different appproaches give a range of lifetimes but with a reasonable overall consistency
- Uncertainties of lifetimes have been reduced
- Lifetimes could be proposed to be changed, but it is not yet clear if this change will be significant.

Lifetimes of ozone-depleting substances in perspective of time

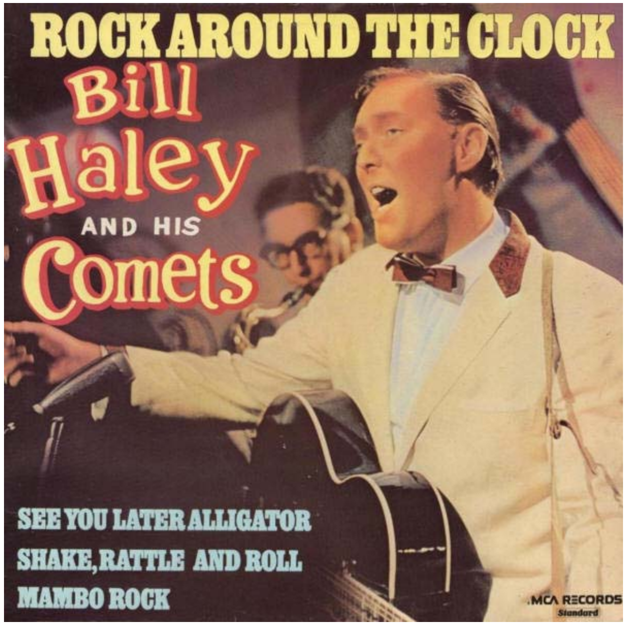
Lifetime of CFC-11: 45 years

44 years ago (1968) first night of Musical HAIR at Broadway

Welcome sulphur dioxide,
Hello carbon monoxide
The air, the air is everywhere
Breathe deep, while you sleep,
breathe deep

Lifetime of CFC-11: 57 years

57 years ago (1955) number 3 of the charts



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