Can Ozone Assimilation Constrain Inorganic Chlorine in the Stratosphere?

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SPARC Data Assimilation Workshop
21-23 June 2010
Exeter, UK

One of the potential of 4D-Var chemistry data assimilation is to use observed species to constrain directly coupled unobserved species.

- Past studies found that only few observed species convey to constrain unobserved modeled species using a simple stratospheric (Fisher and Lary, 1995) or tropospheric (Elbern et al., 1997) DA system
- Using the CRISTA stratospheric data, Errera and Fonteyn (2001) showed that observations of $ClONO_2$ were able to constrain unobserved modeled HCl
- Chai et al. (2006) showed that assimilating NO_y aircraft data with a 4D-Var system improves the analyses of O_3, HNO_3, PAN and RNO3

However, these experiments were based on short term datasets.

- In order to support SPARC CCMVal, BIRA-IASB and DLR, within the PROMOTE project (www.gse-promote.org), are contributing to set a record of 3D fields of ozone and related trace gases using chemistry data assimilation of past observations

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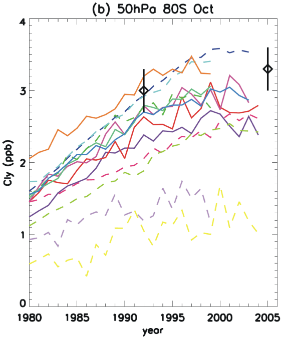
- One of the issue of SPARC CCMVal is the lack of information on the total inorganic chlorines (Cl_y) at South Pole



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- In this talk, we will discuss:
  1. How the ozone observations are able to constrain unobserved inorganic chlorine species in the stratosphere represented in the model
  2. How these inorganic chlorine analyses agree with independent observations

1. The link between O_3 and Cl_y
2. Influence of O_3 observations on Cl_y species
3. Case study using UARS MLS O_3 and the BASCOE system

- Interaction between O_3 and chlorines is carried out through O_3 destroying catalytic cylces, e.g.:

$$Cl + O_3 \rightarrow ClO + O_2 \quad (1)$$

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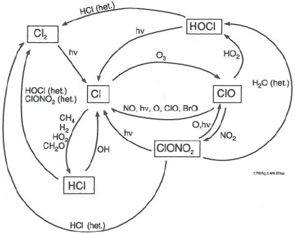

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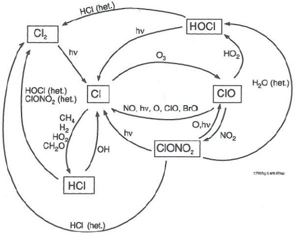
- Cl is linked to the other chlorine species
- Two regions/seasons where O_3 destruction by Cl is significant
  1. the upper stratosphere
  2. the Winter/Spring polar lower stratosphere



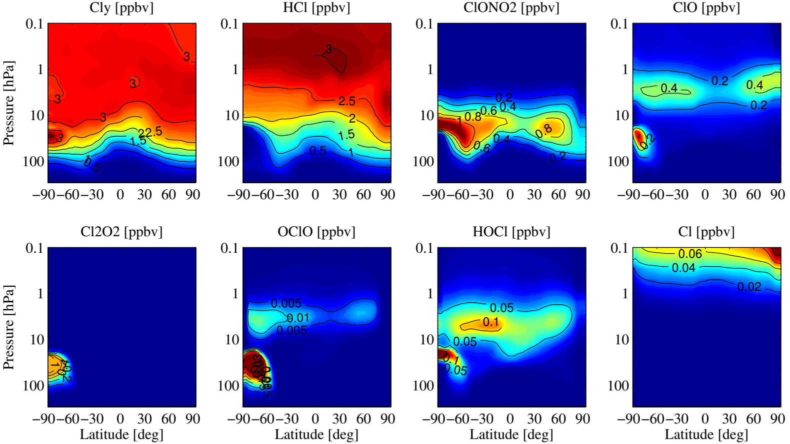
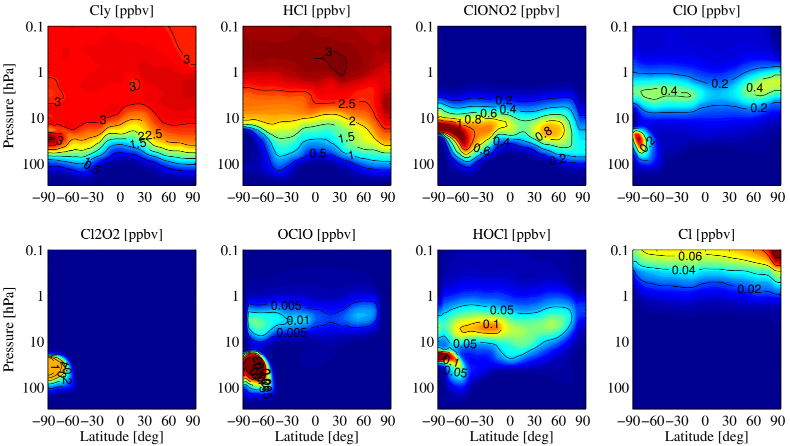


Figure: Zonal average of total inorganic chlorines (Cl_y) and the most abundant members of the family, taken from BASCOE on 15-Jan-1994.

- In the upper stratophere, $[Cl_y] \approx [HCl]$



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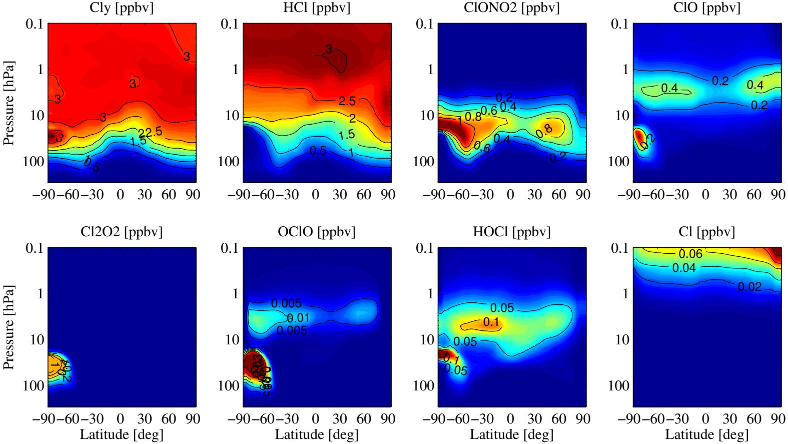
- In the Winter/Spring polar lower stratosphere, $[Cl_y] \approx [ClO] + [Cl_2O_2]$



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Influence functions are used to estimate the constraint of O_3 on Cl_y.
The influence functions are defined as (Fisher and Lary, 1995):

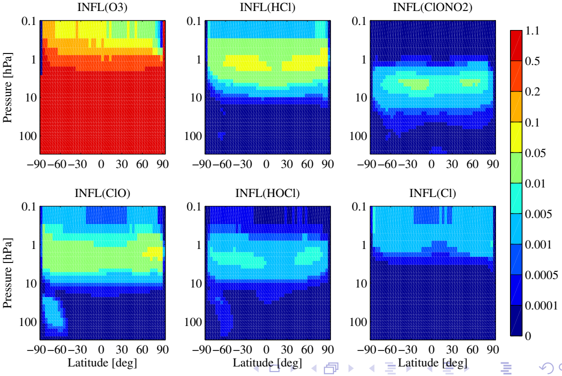
$$\gamma(Species\ j \rightarrow i, Time\ step\ n \rightarrow m) = \frac{(\nabla_{\mathbf{x}_m} J_m^{obs})_i (\mathbf{x}_m)_i}{(\nabla_{\mathbf{x}_n} J_n^{obs})_j (\mathbf{x}_n)_j} \qquad (3)$$

Where:

- i, j denote the species index
- m, n denote the time step, $n \geq m$
- $\mathbf{x}$ denotes the volume mixing ratio (vmr)
- $(\nabla_{\mathbf{x}_m} J_m^{obs})_i$ denotes the gradient of the cost function J with respect to the vmr for species i and time step m

By definition, $\gamma = 1$ for i=j and m=n.

- Influence calculated at every BASCOE grid point. Advection is OFF
- Influence of ${O_3}^{obs}$ at 12UT on VMR^{model} at 0UT, on 15-Sep-1994



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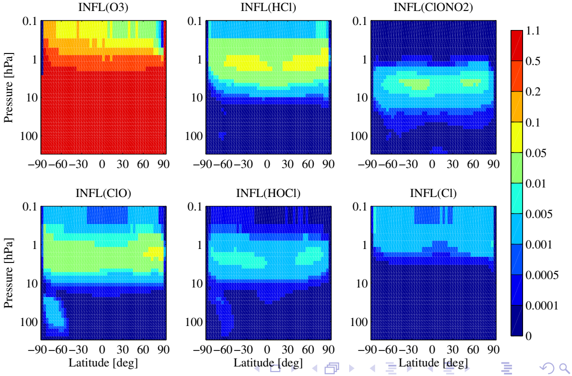

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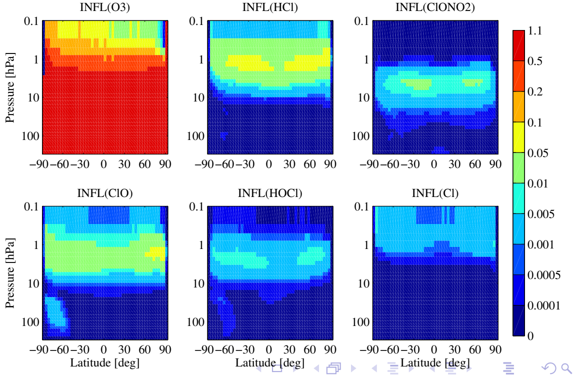

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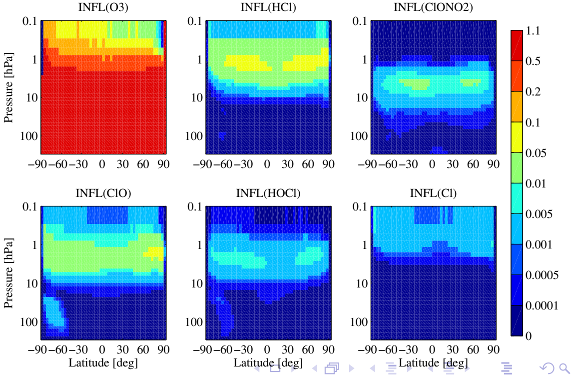

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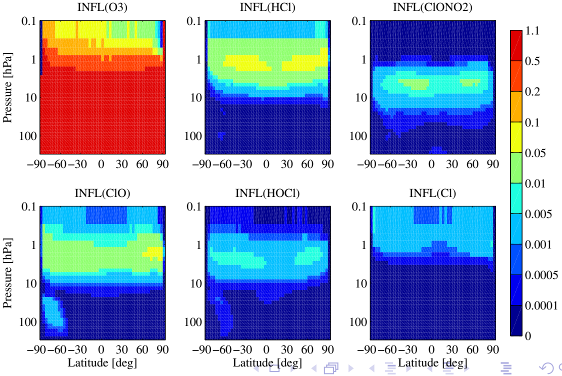
- How long will be the spin-up?



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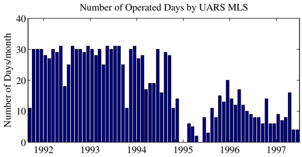

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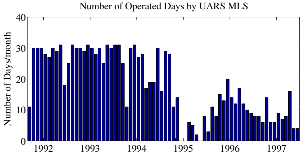

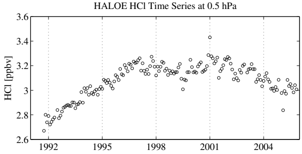

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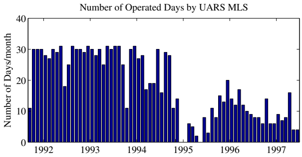
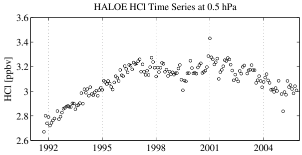
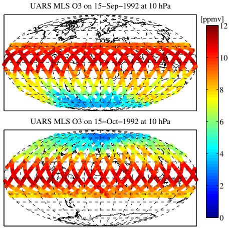
- No assimilation after Nov. 1994 because the number of operated days is too small
- This period occurs during the increase of stratospheric HCl, before the impact of the Montreal Protocol
- UARS performs a yaw maneuver every 36 days.
    - In practice, no MLS data from mid September to mid October at South Pole for 1992-1994 $\Rightarrow$ Influence of O_3 on ClO_x will be very limited







The BASCOE (Belgian Assimilation System for Chemical ObsErvations, Errera et al., 2008) uses the 4D-Var method with a 3D-CTM

- The CTM advects 57 stratopheric species that interact through 200 chemical reactions
- The effect of PSC microphysics are calculated by a simple parameterization
- Surface emissions of organic chlorines is NOT modeled
- In this study, the CTM is run at $5°$ long $\times$ $3.75°$ lat $\times$ 37 vertical levels (surface to 0.1hPa) and driven by the ECMWF ERA-Interim reanalyses
- Only O_3 is assimilated where 10% of data are dropped for a posteriori verifications
- The **B** matrix is set diagonal with a variance set to 50% of the first guess in order to give a strong weight to the observations
- First guess on day 1 taken for SOCRATES 2D model
- A free model run (no assimilation) initialized by SOCRATES was done to assess the benefit of DA

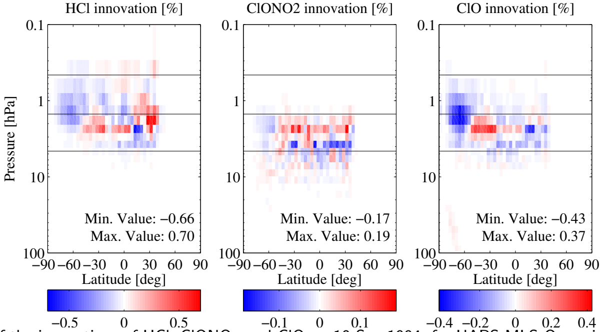
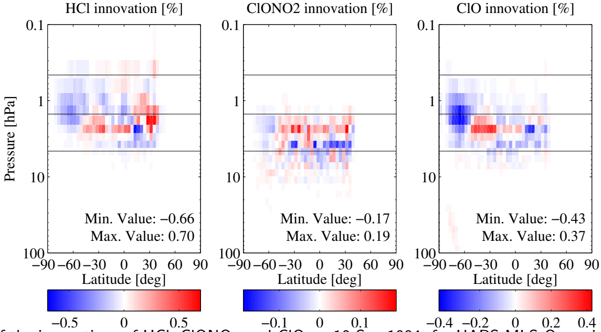


Figure: Zonal mean of the innovations of HCl, $ClONO_2$ and ClO on 10-Sep-1994, for UARS MLS O_3 assimilated by BASCOE

- The innovation is maximum for HCl arround 2hPa



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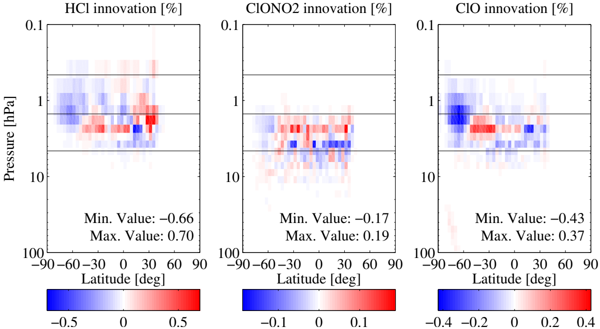
- The innovation is maximum for HCl arround 2hPa
- The innovation appears to be very small for ClO in the Antarctic polar vortex



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- A very good argement is found between MLS O_3 and BASCOE: BASCOE is able to reproduce MLS dropped data within the MLS error bars

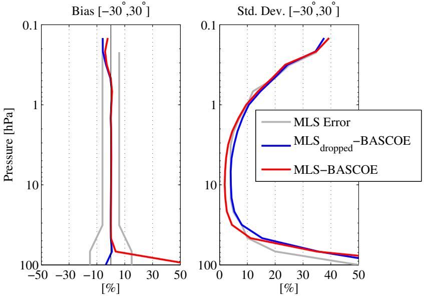


Figure: Bias and standard deviation of mean(MLS-BASCOE) for O_3 using all MLS data (red) and only the 10% of dropped MLS data (blue) between October 1991 and October 1994. Gray line represent the MLS accuracy (left figure) and precision (right figure).

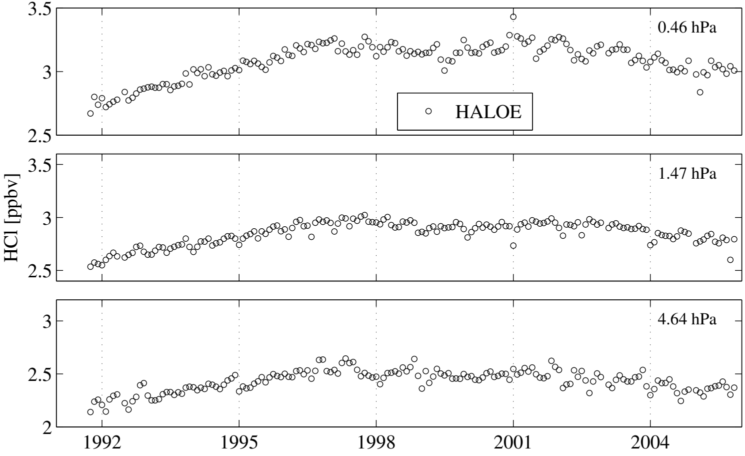
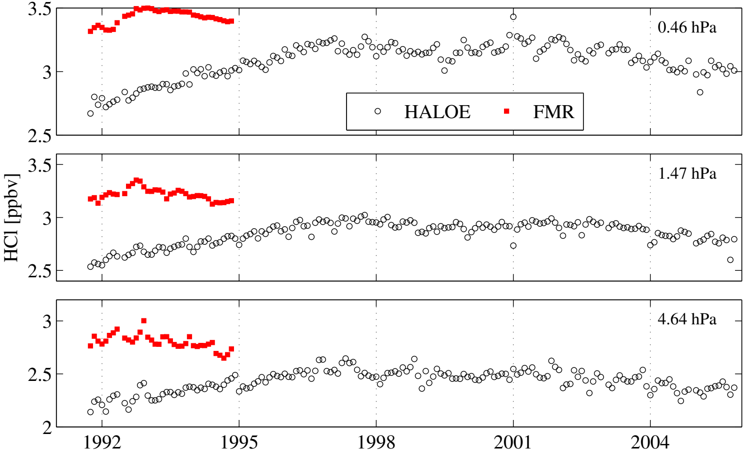


Figure: Time series of tropical (between -30°S and 30°N) monthly averaged HCl from HALOE, the analyses, and the control run at 0.46 hPa (a), 1.47 hPa (b) and 4.61 hPa (c).



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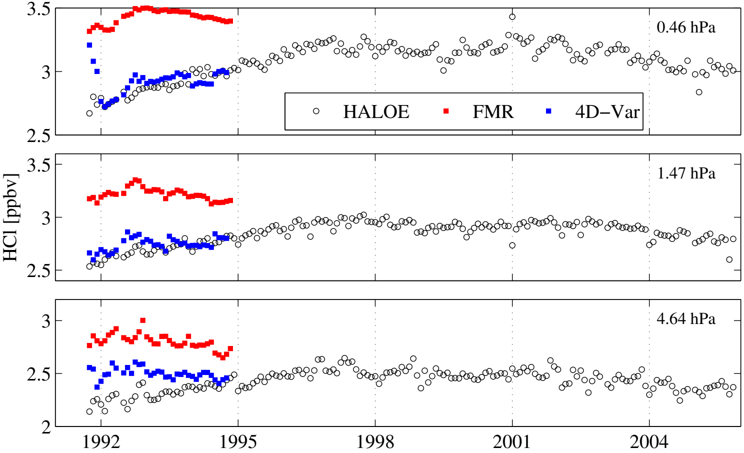

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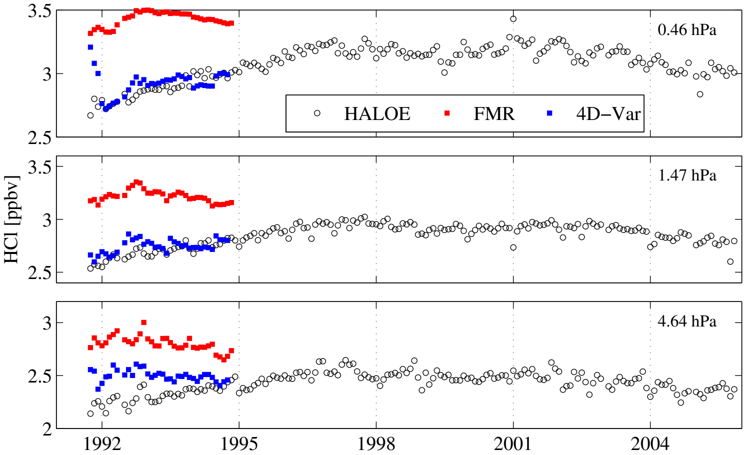
1. The difference between the analyses and FMR clearly shows the influence of O_3 observations on modeled HCl



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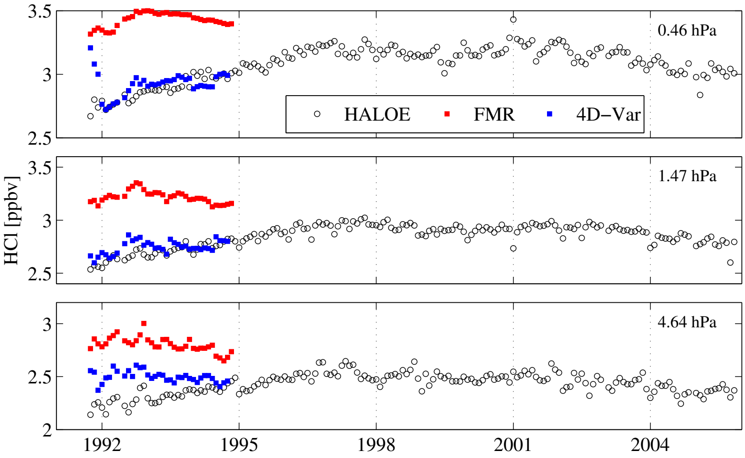
2. The effect of the spin-up at 0.46 and 4.6 hPa (4 and 3 months, respect.) reflects the relatively weak constraint of O_3 on HCl



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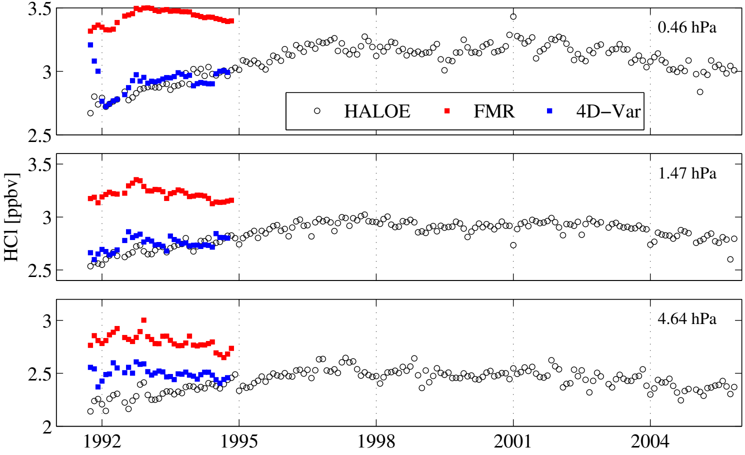
3. The analysed and observed trends are in good agrement at 0.46 and 1.47 hPa



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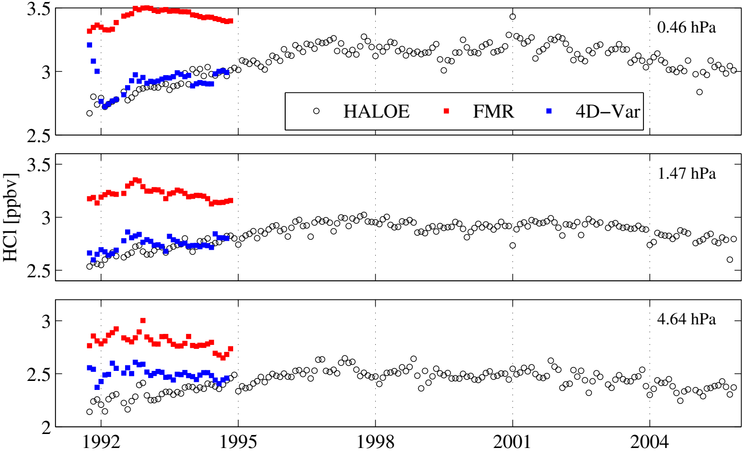
4. This is possible because HCl vary slowly regarding the contraint by O_3 data



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- Differences with upper stratosphere conditions
  1. Here, the production/loss cycle of Cl_y is achieved in few months ($><$ many years in the upper stratosphere)
  2. As the influence functions are relatively small here, the spin-up period is expect to be much longer thant the Cl_y cycle.
- UARS MLS O_3 assimilation shows no significant differences against the FMR (not shown)
- Aura MLS O_3 assimilation - no yaw maneuver - between April-November 2005 shows that (not shown):
  1. analyses are closer to Aura MLS indepentend observations HCl and ClO than the FMR ...
  2. ... but the analyses still too far to the observations
- Modeling the contraint of O_3 on Cl_y in the **B** matrix might improve this issue

1. Stratospheric chemical scheme (and their adjoint) allows O_3 observations to contrain Cl_y in the upper stratosphere
2. HCl analyses from the assimilation of UARS MLS O_3 by BASCOE over three years allow to reproduce the HALOE HCl trend
3. The constraint of O_3 data on modeled HCl appears to be relatively weak:
   - This might be increased by modeling this constraint in the **B** matrix
4. An alternative of the influence function is necessary to derive *a priori* the time of the spin-up
5. Two potential applications:
   - Estimation of the upper stratosphere HCl trend using assimilation of SBUV from 1978 to 1991 (no HCl data)
   - Estimation of the Winter/Spring polar stratospheric Cl_y using assimilation of ozonesondes

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