Ozone has been very, very good to me!

Richard S. Stolarski

NASA Goddard Space Flight Center, Emeritus

Research Professor, Johns Hopkins University

Ozone (baseball) has been very, very good to me! Thank you.

SNL, Nov 1978

Is that all?

Keep your eye on the ball!

Some Personal History

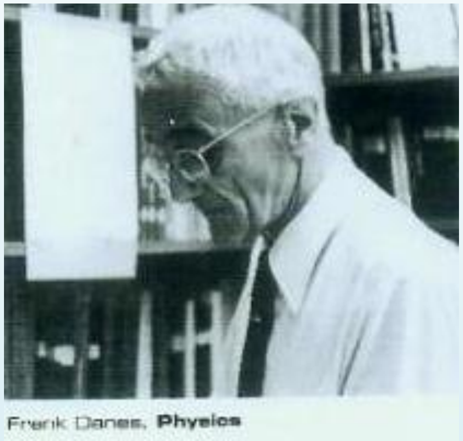
- Born, Ft. Lewis, WA, 1941
  - Mother, born Tacoma, WA
  - Father, born Pittsburgh, PA; career military
    - Grandparents all immigrants from Eastern Europe (1905-1910)
    - Maternal grandparents from Istria (Croatia)
    - Paternal grandparents from Poland

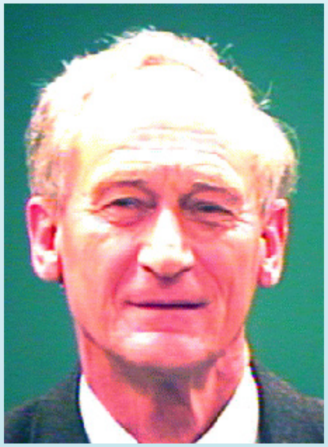
Obligatory Childhood Picture

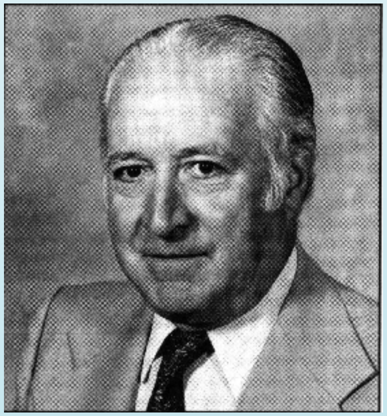
School and Career

- BS from University of Puget Sound, 1963, Physics and Math
- PhD in Physics from University of Florida, 1966
  - Thesis on the penetration of energetic particles into the atmosphere
- Postdoc at University of Michigan, 1967-1974
  - Worked on ionosphere and thermosphere problems
  - Managed to move into stratospheric research during SST debate
- Took NASA position at Manned Space Center (now JSC), 1974
  - Part of Environmental Effects Project Office
  - Office wrote environmental impact statement for the space shuttle
- Move to GSFC to newly-formed Stratospheric Physics and Chemistry Branch, 1976

Some people who had a major influence on my career

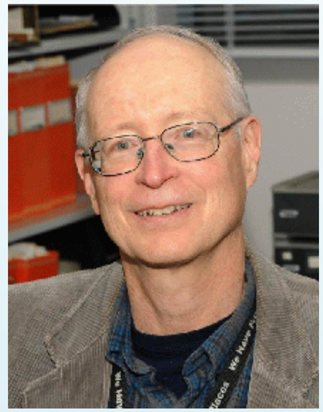






Some of the many colleagues who enabled me to have a good scientific career



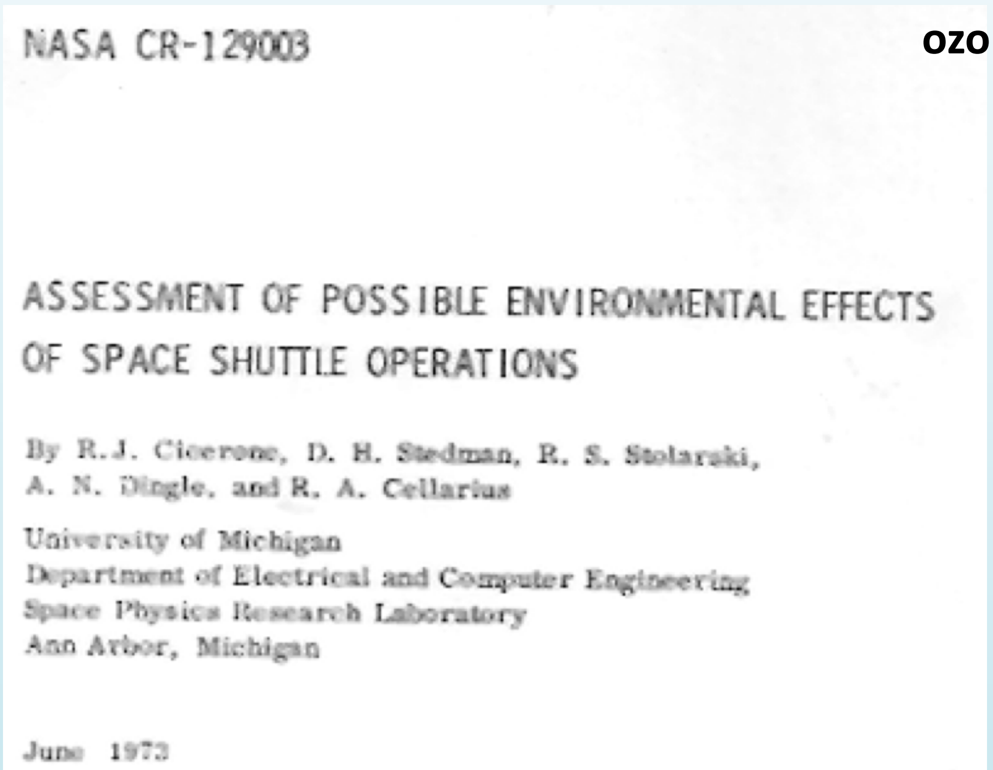







How did we get involved in chlorine chemistry in the atmosphere?

Stedman, circa 1972: “Chlorine destroys ozone; everybody knows that!”



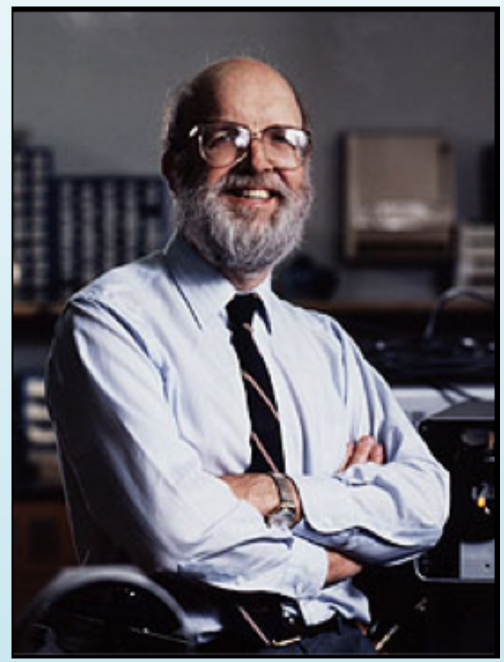
NASA CR-129003

ASSESSMENT OF POSSIBLE ENVIRONMENTAL EFFECTS OF SPACE SHUTTLE OPERATIONS

By R. J. Cicerone, D. H. Stedman, R. S. Stolarski, A. N. Dingle, and R. A. Cellarius

University of Michigan
Department of Electrical and Computer Engineering
Space Physics Research Laboratory
Ann Arbor, Michigan

June 1973



How did we come to know about the importance of CFCs?

Article

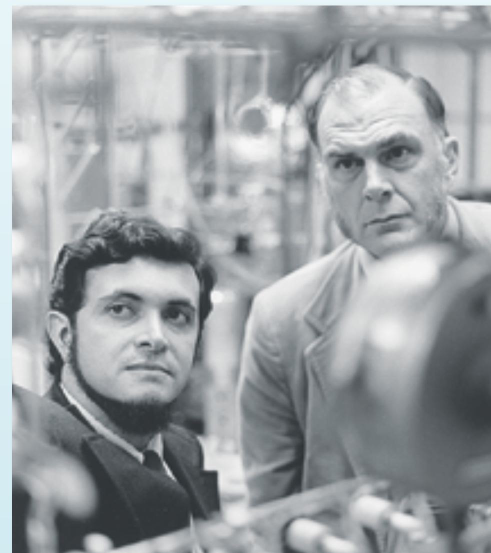
Nature **249**, 810-812 (28 June 1974) | doi:10.1038/249810a0

Stratospheric sink for chlorofluoromethanes: chlorine atomc-atalysed destruction of ozone

Mario J. Molina & F. S. Rowland

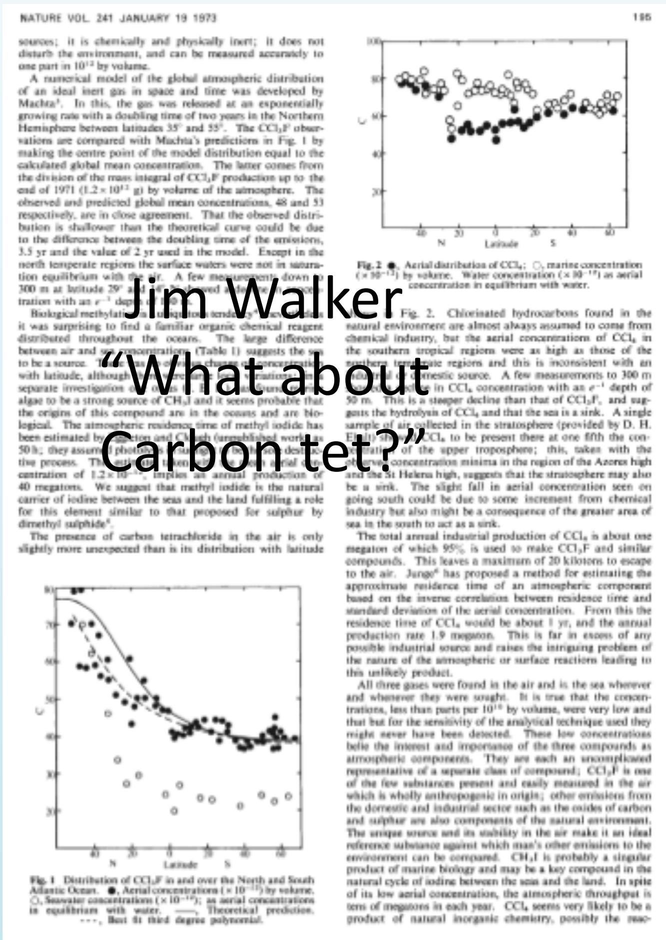
1. Department of Chemistry, University of California, Irvine, California 92664

Chlorofluoromethanes are being added to the environment in steadily increasing amounts. These compounds are chemically inert and may remain in the atmosphere for 40–150 years, and concentrations can be expected to reach 10 to 30 times present levels. Photodissociation of the Chlorofluoromethanes in the stratosphere produces significant amounts of chlorine atoms, and leads to the destruction of atmospheric ozone.



The information was out there for people like us to have put forward the fluorocarbon-ozone theory – but we didn't!

Lovelock et al. Nature, 1973



Jim Walker
"What about Carbon tet?"

Chuck Kolb (to me), November 1973 "Have you ever thought about Freons? They are inert in the troposphere, not soluble and don't absorb visible light. They will get up into the stratosphere where uv will produce chlorine."

Meanwhile, we wrote a paper for Canadian Journal of Chemistry in 1974

Stratospheric Chlorine: a Possible Sink for Ozone

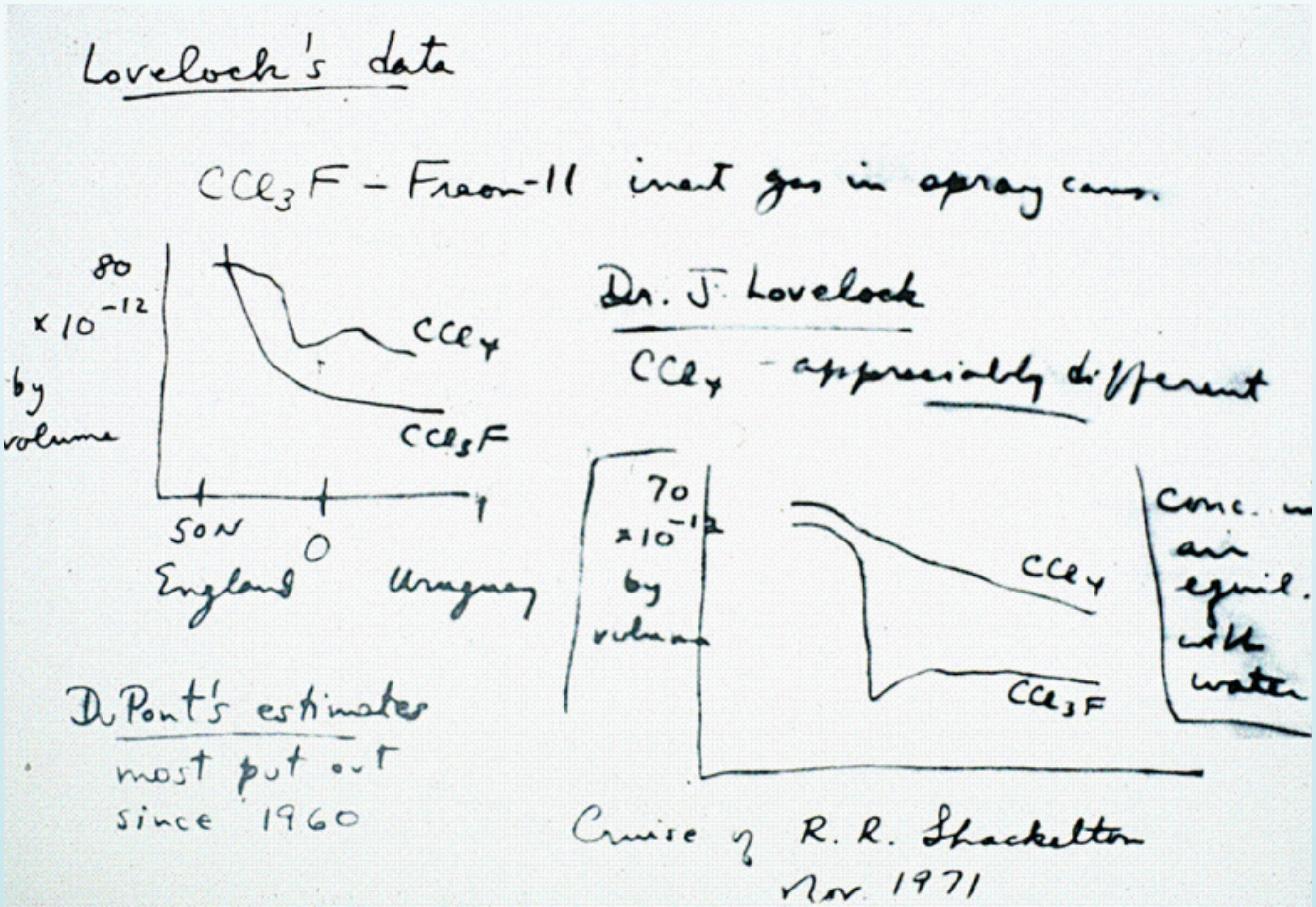
R. S. STOLARSKI AND R. J. CICERONE

Space Physics Research Laboratory, The University of Michigan, Ann Arbor, Michigan 48105

Received January 18, 1974

This study proposes that the oxides of chlorine, ClO_x, may constitute an important sink for stratospheric ozone. A photochemical scheme is devised which includes two catalytic cycles through which ClO_x destroys odd oxygen. The individual ClX constituents (HCl, Cl, ClO, and OClO) perform analogously to the respective constituents (HNO_3, NO, NO_2, and NO_3) in the NO_x catalytic cycles, but the ozone destruction efficiency is higher for ClO_x. Our photochemical scheme predicts that ClO is the dominant chlorine constituent in the lower and middle stratosphere and HCl dominates in the upper stratosphere. Sample calculations are performed for several ClX altitude profiles: an assumed 1 p.p.b. volume mixing ratio, a ground level source, and direct injection by volcanic explosions. Finally we discuss certain limitations of the present model: uncertainty in stratospheric OH concentrations, the possibility that ClOO exists, the need to couple ClO_x cycles with NO_x and HO_x cycles, and possible heterogeneous reactions.

Rowland heard a talk by Lovelock – his notes below were the basis for studies by his new post-doc, Mario Molina



Slide 1: Ft. Lauderdale, January 1972.

The Antarctic Ozone Hole

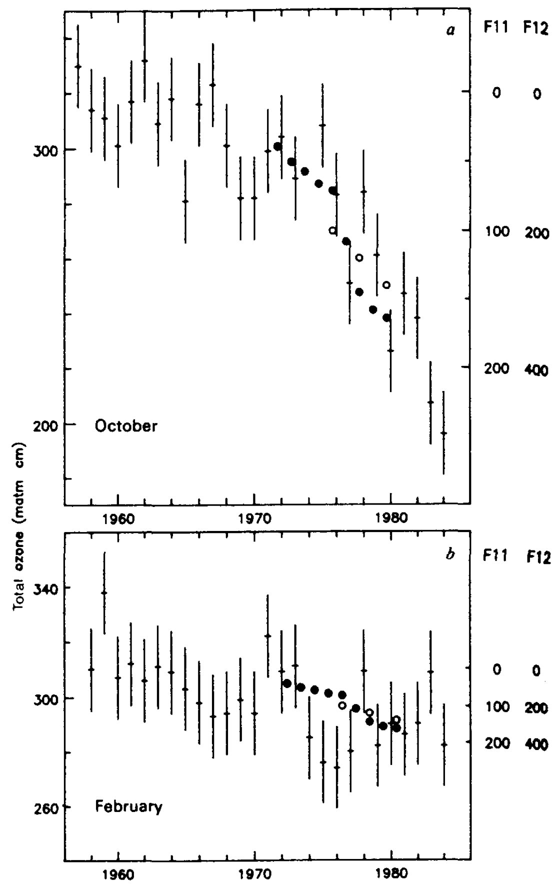
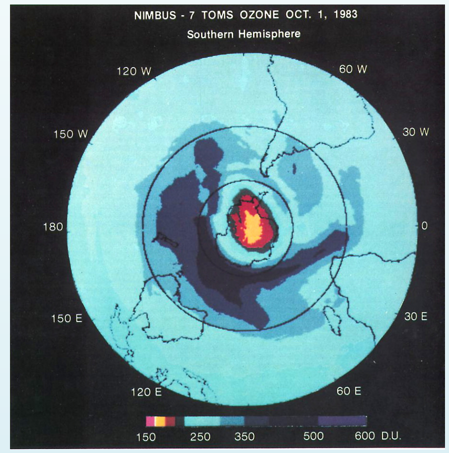
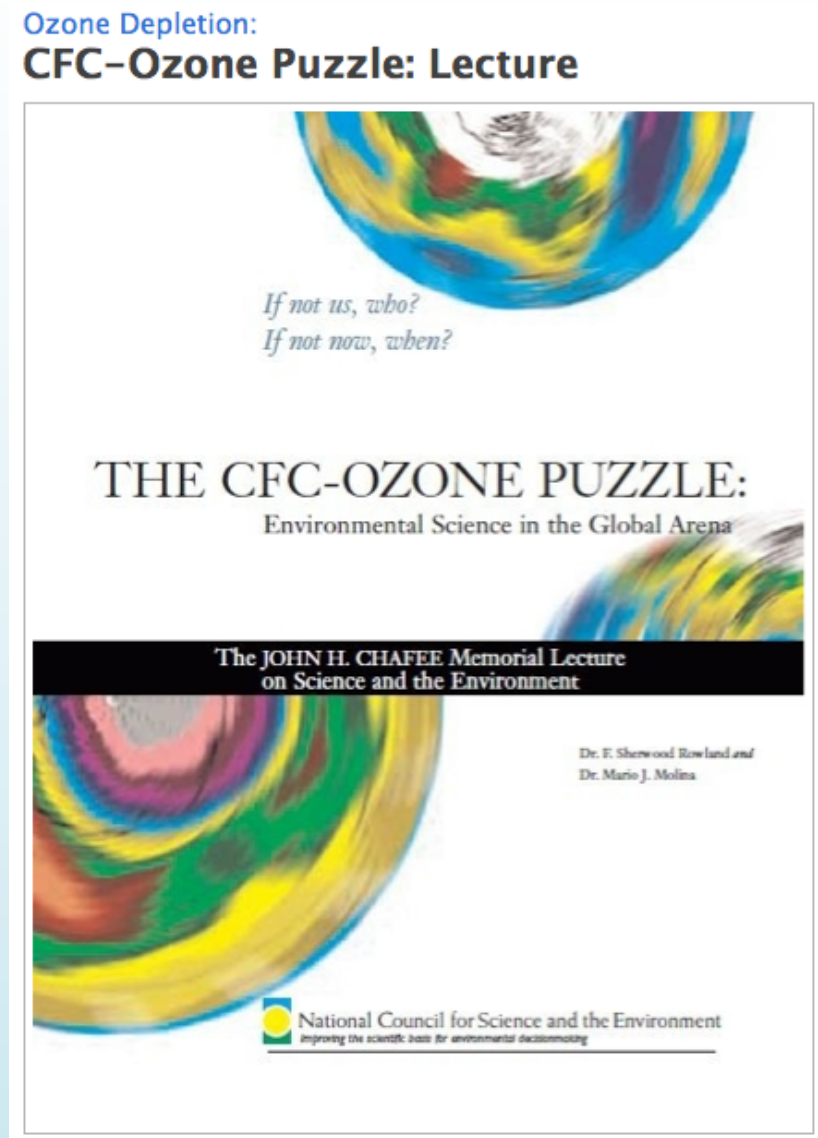


Fig. 2 Monthly means of total O_3 at Halley Bay, and Southern Hemisphere measurements of F-11 (●, p.p.t.v. (parts per thousand by volume) $CFCl_3$) and F-12 (○, p.p.t.v. CF_2Cl_2). *a*, October, 1957–84. *b*, February, 1958–84. Note that F-11 and F-12 amounts increase down the figure.



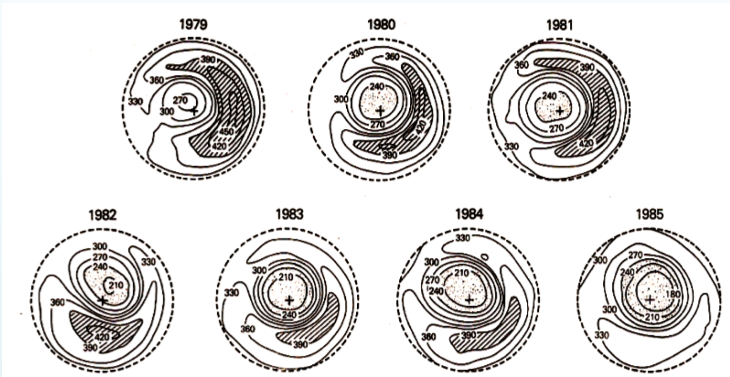
June 1985: Farman et al. paper published
July 1985: WMO/UNEP Ozone Assessment meeting
August 1985: Bhartia presentation at IAGA

Rowland: This observation by the British brought the NASA satellite people back to look at their data. Actually, they've gotten sort of a bad rap about it. What they did was program their data to reject, but notify, that some unusually low ozone values were being recorded. If you are getting unexpected low values when the instrument is working at the very limit of its detection, then you put that aside, saying, maybe there is something happening there that is real, but maybe it is an instrumental problem, and we'll have to go back and look at it carefully.



http://WWW.EOEARTH.ORG/ARTICLE/CFC-OZONE_PUZZLE:_LECTURE?TOPIC=49594

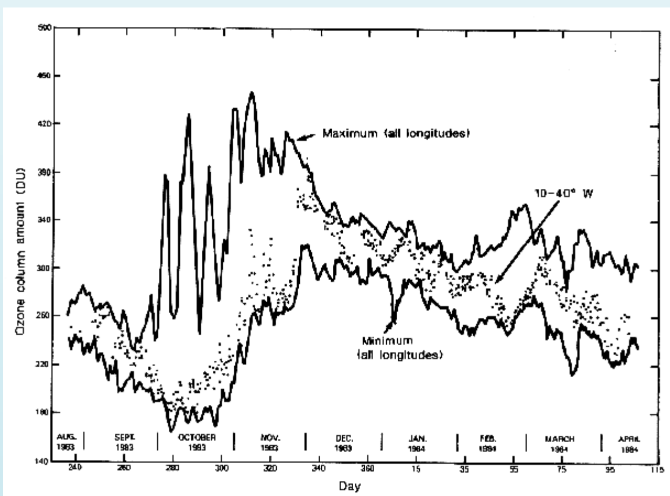
Figures from our 1986 Nature paper on the ozone hole

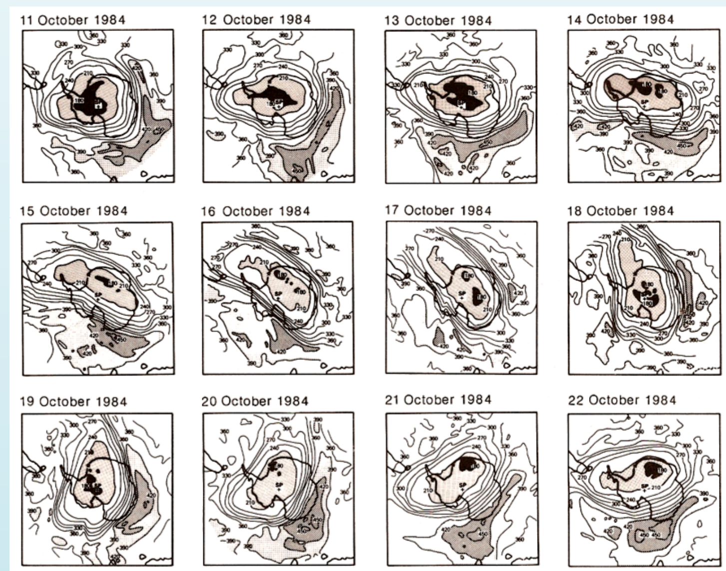


Nimbus 7 satellite measurements of the springtime Antarctic ozone decrease

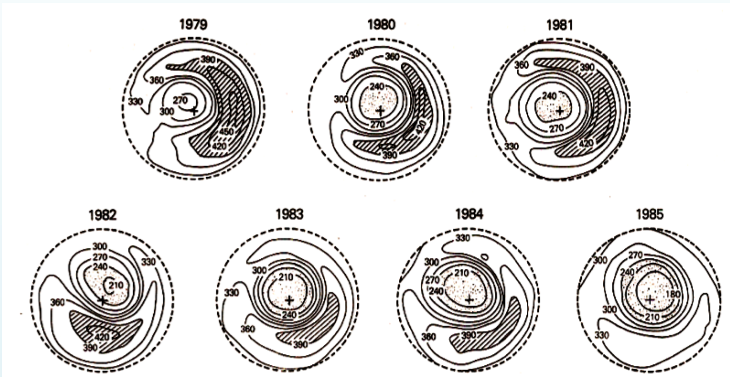
R. S. Stolarski, A. J. Krueger, M. R. Schoeberl, R. D. McPeters, P. A. Newman & J. C. Alpert*

NASA/Goddard Space Flight Center, Laboratory for Atmospheres, Greenbelt, Maryland 20771, USA





Figures from our 1986 Nature paper on the ozone hole



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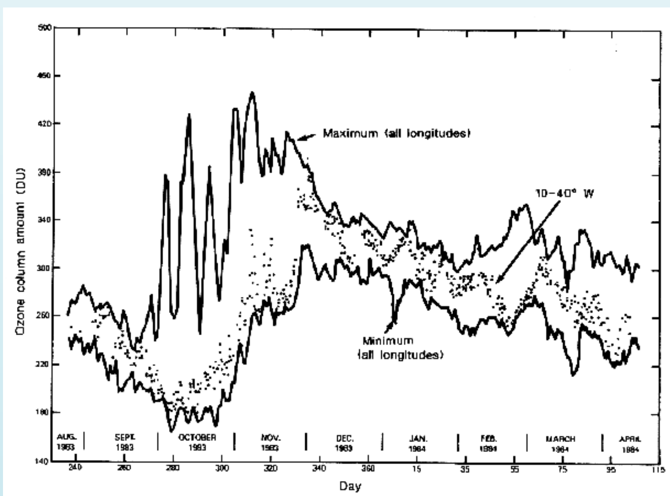
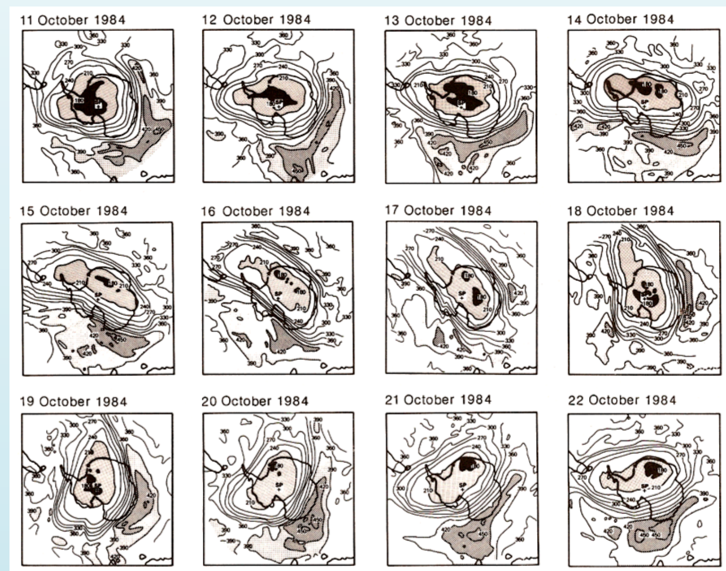




What about the theory of the ozone hole?

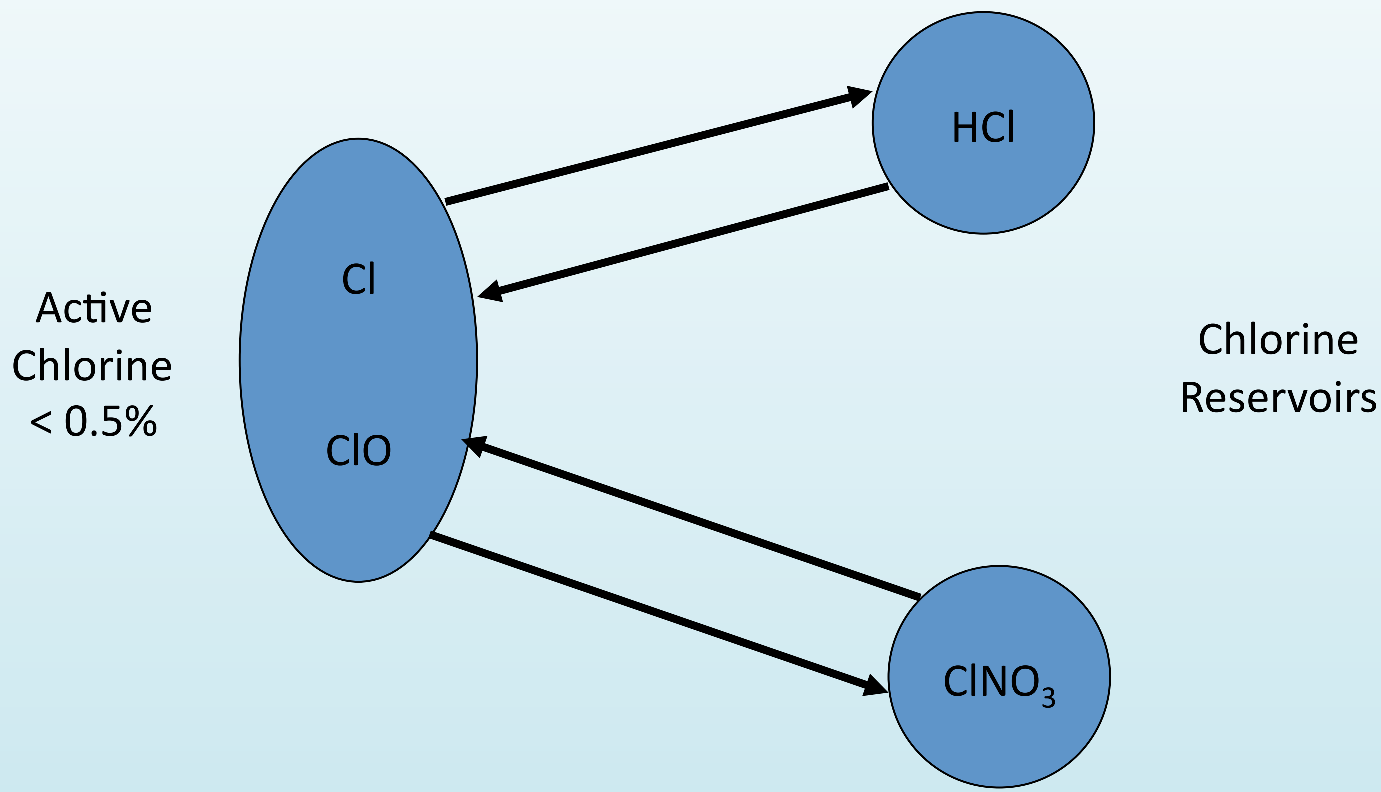
Feldafing Workshop, June 1984

The rationale for the scope of the present assessment report was based on information gained at a scientific workshop, entitled "Current Issues in Our Understanding of the Stratosphere and the Future of the Ozone Layer" which was held in Feldafing, Federal Republic of Germany in June 1984 with international participation, co-sponsored by NASA, FAA, WMO, and BMFT.

Sherry Rowland: "Missing Chemistry"

- **Don't need to worry about fast channels**
- **What if one reservoir molecule reacted with another, e.g. $ClONO_2$ and HCl?**
- **Molina (and Sato and Rowland independently) had been studying this reaction in the lab**
- **They found a small, but measureable rate coefficient**
- **Inserted into a model, the reaction implied that 30% of the global amount of ozone should have already disappeared**
- **Later reported that reaction was occurring on teflon walls of the experiment**

Chlorine Photochemistry (normal)



Polar Stratospheric Clouds (PSCs)

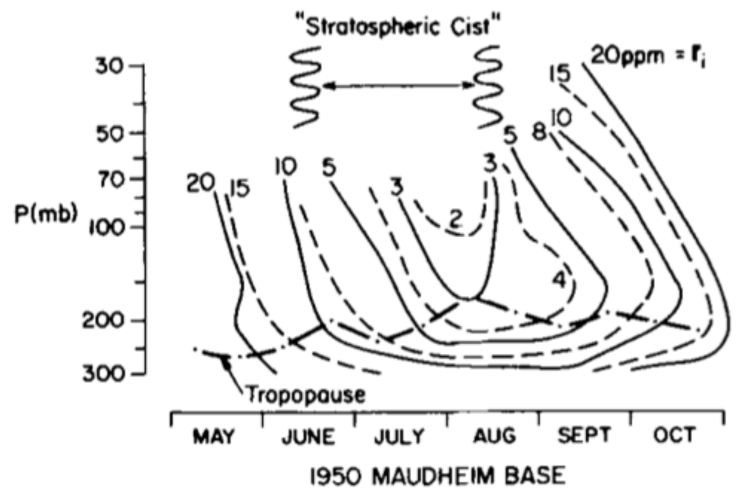


Fig. 1. Saturation mixing ratios of water vapor with respect to ice (in ppm, i.e. units of 10^{-6} g H_2O/g air) vs pressure for Maudheim base (70°03′ S, 10°56′ W) during the 1950 Antarctic winter. The curves extend up to the bursting level of the balloons. The temperature data used were half-monthly means. The position of the half-monthly mean tropopause is indicated. The approximate dates over which "Stratospheric Cist" were reported are also indicated.

Tellus (1977), *29*, 530–534

On the nature of persistent stratospheric clouds in the Antarctic

By JOHN L. STANFORD, *Physics Department, Iowa State University, Ames, Iowa 50011, U.S.A.*

(Manuscript received November 8, 1976; in final form January 26, 1977)

ABSTRACT

Thin, persistent very high clouds called "Stratospheric Cist" were reported at Maudheim base (71°03′ S, 10°56′ W) during the 1950–51 austral winters. The nature of these clouds, whether they are H_2O or dust, is of importance in the proposed sink for stratospheric moisture in the Antarctic winter.

Results are presented from analyses of unpublished daily cloud reports and daily upper air measurements from the Maudheim expedition. The results reveal strong evidence that the "Stratospheric Cist" were stratospheric ice clouds. Given this identification, an upper limit of 6–7 ppm can be assigned to the water vapor mixing ratio in the lower stratosphere for the 1950–51 Antarctic winters.

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 87, NO. C7, PAGES 5001–5008, JUNE 20, 1982

A Model of the Antarctic Sink for Stratospheric Water Vapor

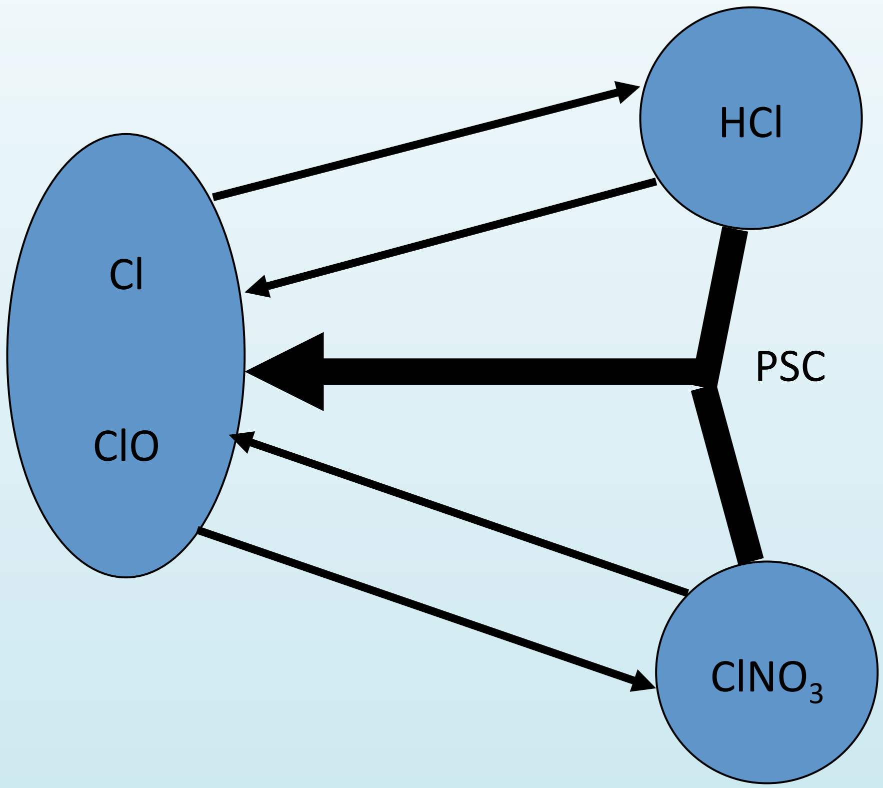
ANNE R. DOUGLASS[1] AND JOHN L. STANFORD

Department of Physics, Iowa State University, Ames, Iowa 50011

Model calculations have been performed to quantitatively estimate the amount of stratospheric H_2O removed by freeze out during the winter in the Antarctic region. The model uses realistic temperature and wind fields based on Hartmann's Nimbus 5 temperature grids. For the 1973 Antarctic winter stratosphere, initial water vapor mixing ratios of 2.7 and 3.0 ppmm lead to removal of 1.6 and 2.7 × 10^{10} kg H_2O from the lower stratosphere. This annual sink magnitude is small, representing about 2% of the total stratospheric H_2O burden and about 10% of the apparent sink identified by Ellsaesser (1974). However, a sink of this magnitude is close to the lower rate estimates of water produced in the stratosphere by methane oxidation. The sink is not large enough to balance additional, proposed water vapor sources.

Chlorine Photochemistry (polar)

Active
Chlorine
Increases
By 50 to 100x



PSC Reactions
Drain
Chlorine
Reservoirs

Digression to Atmospheric Models

- We build numerical models because we cannot reason through the complexities with a few simple equations.
- We use the models to work our way through complex interactions, especially when there are feedbacks.
- The problem is then to diagnose the model to work our way through the sequence of calculations to understand why the model came up with the result.
- Finally we need to ask what this means about how the atmosphere works.

The model is not the atmosphere:

It is a representation of the atmosphere that can be used as a tool to gain knowledge about how the atmosphere works.

Our computer capability as increased remarkably: What we used to struggle to do in two dimensions, we now can do in three dimensions.

The first model I worked with was an 8-level 1D model of a few chemical cycles that took 20 minutes on a Wang calculator to obtain a steady state.

One problem that I see is that when computer capability increases, we tend to expand the complexity of the model. I would argue for slowing the increase of complexity in favor of doing more expansive experimentation with simpler versions of the model to try to understand how it works.

The CCM Project

- **In about 2004, we got together with GMAO to create a chemistry-climate model**

- **Coupled our chemistry-transport model with the GMAO general circulation model**

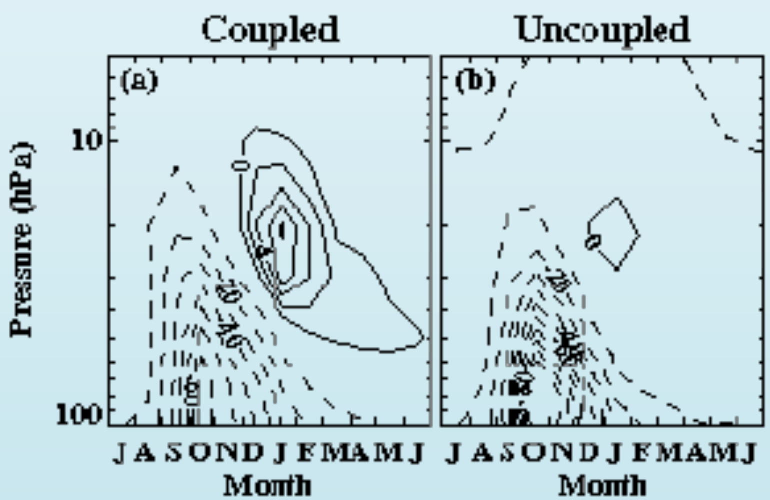
How do you test the coupling?

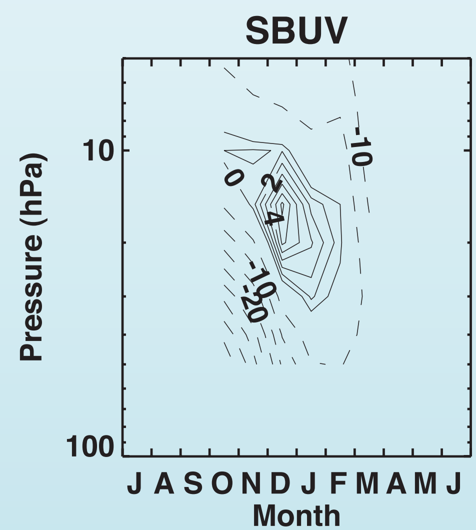
GEOPHYSICAL RESEARCH LETTERS, VOL. 33, L21805, doi:10.1029/2006GL026820, 2006

An ozone increase in the Antarctic summer stratosphere: A dynamical response to the ozone hole

R. S. Stolarski,[1] A. R. Douglass,[1] M. Gupta,[1,2] P. A. Newman,[1] S. Pawson,[3]
M. R. Schoeberl,[4] and J. E. Nielsen[5]

Received 5 May 2006; revised 31 August 2006; accepted 19 September 2006; published 7 November 2006



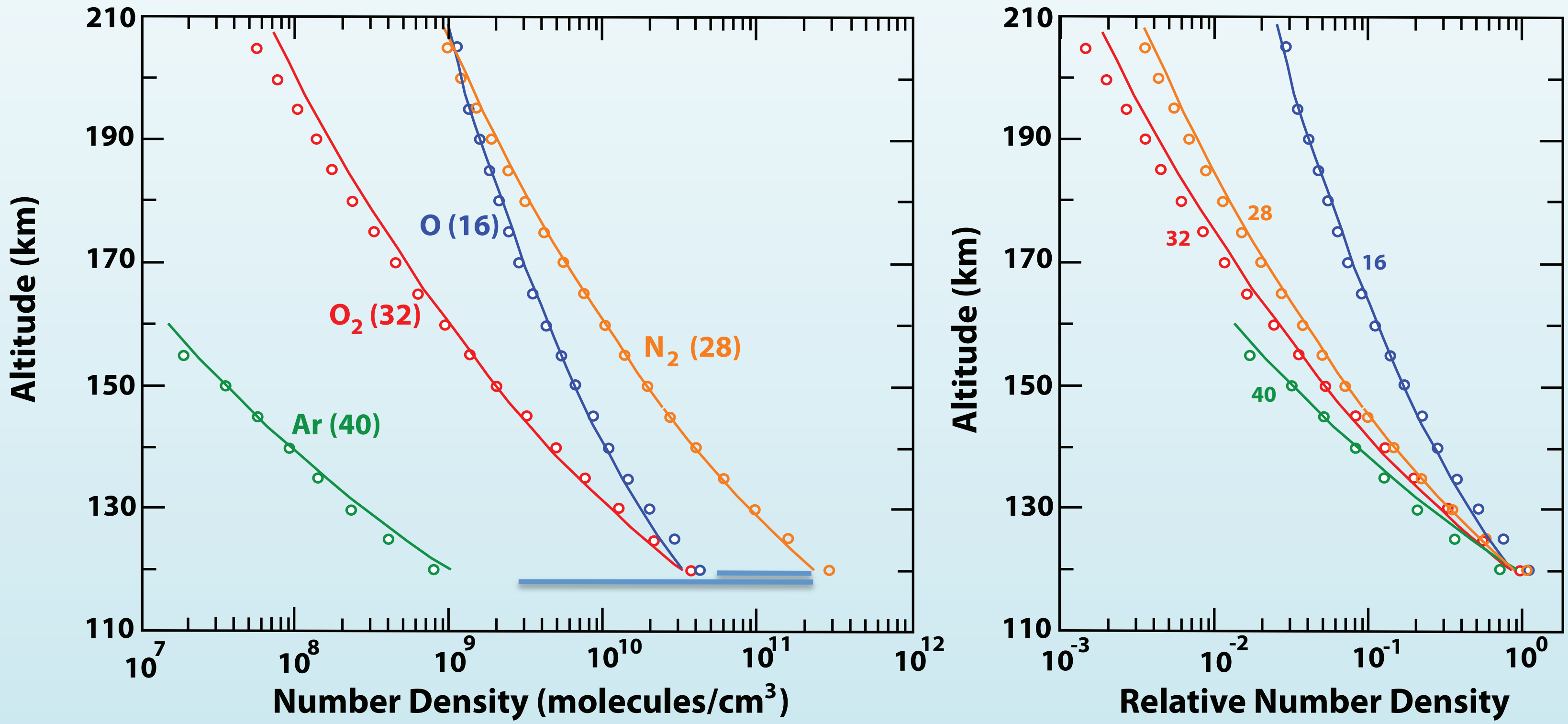


An aside on giving talks on hot issues such as the fluorocarbon-ozone issue was at one time

- **You are an expert on science: separate your science from any opinions on what should be done by society**
- **There are no dumb questions: Even if you have heard a question a thousand times, respect the questioner**
- **Be prepared: you know the standard questions so you should have a prepared approach to these**
- **If you don't know the answer say so.**

Question asked over and over when making presentations on fluorocarbon-ozone theory:

Fluorocarbons are heavier than air-how can they get to the stratosphere?



Diffusive Separation in the Thermosphere: a concept studied since the 1960s: graphs redrawn from Hedin and Nier (1965)

Final thoughts

- Details are important, but if you don't try to step back and see the big picture you can waste a lot of time on the wrong details
- Opportunities abound: will you recognize them? Will you follow up on them? Or will you just let them pass by?
- Ideas are cheap – Selection of good ones and follow through on them is what counts.

Remember:
Keep your eye on the ball