N92-14514

A. **Title of Research Task**

RADIATIVE FLUX MEASUREMENTS IN THE STRATOSPHERE

B. **Investigators and Institution**

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C. **Abstract of Research Objectives**

The objective of this research is to determine how the stratospheric tropospheric exchange of water vapor is affected by the interaction of solar (visible) and planetary (infared) radiation with tropical cumulonimbus anvils. This research involves field measurements from the ER-2 aircraft as well as radiative transfer modelling to determine heating/cooling rates and profiles that directly affect the enchange between the troposphere and the stratosphere.

D. **Summary of Progress and Results**

The data sets obtained during the 1987 Stratospheric - Tropospheric Exchange Project (STEP) deployment in Australia includes:

a) Infrared intensities at 6.5 and µm. Field of view: ~8° half angle. Measured radiation intensities were referenced to a liquid Nitrogen cooled black body.

b) Broad spectral bandpass IR fluxes (3 to 4O µm)

c) Broad spectral bandpass (0.3 - 0.6 µm) solar fluxes

We have used the above data sets to determine cloud optical depths, brightness, temperatures and heating rates. The IR intensities at 10.5 µm are being used for satellite - aircraft intercomparison studies.

E. Journal Publications

Ackerman T. P., Liou, K. N., Valero F. P. J. and Pfister, L., "Heating Rates in Tropical Anvils". Jour. Atm. Sci 45, 1606 (1988).

Liou, K. N., S. C. Ou, F. P. J. Valero and T. P. Ackerman, "Remote Sounding of the Cirrus Cloud Temperature and Optical Depth using ER-2, 6.5 and 10.5 μm Radiometers". Submitted Jour. Atm. Sci (1989).

Valero F. P. J., T. P. Ackerman, W. J. Y. Gore. "Optical Depths and Brightness Temperatures of Tropical Anvils, a Statistical Study". Submitted, Jour. Atm. Sci (1989).

Hammer P. D., F. P. J. Valero and S. Kinne; "Comparative Study of Infrared Radiance Measurements by an ER-2 based Radiometer and the LANDSAT 5 Thematic Mapper (TM6)". Summitted, Monthly Weather Review (1989).

A. TITLE: Use of radon and cosmogenic radionuclides in the study of stratosphere-troposphere exchange processes

B. PRINCIPAL INVESTIGATOR: Dr. Mark A. Kritz, Atmospheric Sciences Research Center, State University of New York, Albany, NY 12222

C. RESEARCH OBJECTIVES: To develop and implement, in the context of NASA's Stratosphere-Troposphere Exchange Project (STEP), means of using radon (Rn-222) and cosmogenic radionuclide (Be-7 and P-32) measurements to identify and quantify the exchange of air between the troposphere and the stratosphere. STEP goals include the determination of where, when, how, and at what rate tropospheric air (with its burden of chemical trace constituents) enters the stratosphere, and the identification of the process or processes responsible for the observed extreme dryness of stratospheric air. Because Rn-222, Be-7 and P-32 are subject to the same transport processes as other, chemically reactive atmospheric trace constituents, but have no chemical sources or sinks, the measurement of these and other natural radionuclides has proved to be a valuable component of many field experiments.

D. SUMMARY OF PROGRESS AND RESULTS: The natural cosmogenic radionuclides Be-7 and P-32 (with half-lives of 53 and 14 days, respectively) are produced in the atmosphere (primarily in the stratosphere) by interactions between cosmic rays and nitrogen, oxygen and argon.

The Spring 1984 and Spring 1986 STEP flight series studied transports and troposphere-to-stratosphere exchanges associated with cyclogenesis and tropopause folding in the vicinity of mid-latitude upper tropospheric jets. One of the goals of the mission was to determine whether the entrainment of upper level tropospheric air in conjunction with these processes plays a role in maintaining the vertical gradients of many trace constituents in the lower stratosphere. An analysis of Be-7 measurements made in zones of active mixing near tropopause folds, and in an undisturbed region far from the jet supports this hypothesis, and is presented in publication 1.

In contrast to Be-7 and P-32, radon originates in the earth's crust, where it is produced by the decay of the trace quantites of Ra-226 found in rocks and soils. As a result of its surface-level source, the absence of chemical sources or sinks, and its short (3.8 day) radioactive half-life, radon concentrations are typically large in the continental boundary layer, decrease with height in the troposphere, and are vanishingly small in the stratosphere. Thus the presence of any significant concentration of radon in the stratosphere would be indicative of a recent intrusion of tropospheric air, and the simultaneous observation of high radon and low total water concentrations would be unambiguous evidence of a dehydration process accompanying this exchange.

The decay of radon leads to the formation (via a number of short-lived intermediates) of Pb-210, which has a 22 year radioactive half life. The extensive series of measurements made by the Department of Energy in the High Altitude Sampling Program (HASP) and Project Airstream revealed that there is a large standing crop of Pb-210 in the stratosphere. The observed Pb-210 concentrations were relatively invariant with altitude and latitude, as would be expected from the length of its radioactive half life (22 years) relative to stratospheric residence times. Since the only source of Pb-210 is the decay of radon, it follows that this stratospheric Pb-210 can be accounted for only by a), the transport into the stratosphere of tropospheric Pb-210; or b), the decay, in the stratosphere, of radon entering from the troposphere. As discussed in papers 2 and 4 (see list of publications), there are several lines of evidence suggesting that the transport of significant quantities of tropospheric Pb-210 to the stratosphere is unlikely, leaving the transport of tropospheric radon as the only likely source for the stratospheric Pb-210.

If this reasoning is correct, it follows that the dominant troposphere-to-stratosphere exchange process (or processes) must carry with them enough radon to account for the 30 x 10^4 atom/SCM mean Pb-210 concentration observed in the stratosphere. This would imply a mean radon activity in tropospheric air entering the stratosphere of approximately 17 pCi/SCM.

Radon concentrations in the undisturbed upper troposphere are typically on the order of 1 pCi/SCM, about an order of magnitude less than that needed to account for the observed stratospheric Pb-210 abundances. Radon concentrations in the continental boundary layer, on the other hand, are typically on the order of 100 pCi/SCM. Thus the rapid convective ascent of lower tropospheric air in cumulonimbus cells, an exchange mechanism receiving close scrutiny in STEP, would appear to have the potential of bringing the requisite quantity of radon to the stratosphere.

During the STEP field campaign in Darwin, Australia the ER-2 aircraft was directed to regions where meteorological analysis indicated the possibility of active troposphere-to-stratosphere exchange. Elevated radon concentrations were observed in these regions, confirming the occurence of recent or ongoing intrusions of tropospheric air into the stratosphere. Low radon concentrations were found, as expected, in regions where meteorological analyses indicated an absence of recent troposphere-to-stratosphere exchanges.

While a through analysis considering the entire set of meteorological observations and trace constituent measurements must be made before any final conclusions are drawn, preliminary support for the importance of cumulonimbus convection as an exchange mechanism was provided by the radon measurements made during the Darwin field campaign, during which radon concentrations on the order of 20 pCi/SCM were observed in the lower stratosphere downwind of high cumulonimbus turrets.

E. PUBLICATIONS:

1) Airmass origins and troposphere-to-stratosphere exchange associated with mid-latitude cyclogenesis and tropopause folding inferred from Be-7 measurements, by M. A. Kritz, E. F. Danielsen and H. B. Selkirk, in press, Journal of Geophysical Research.

2) A box model for the exchange of ^{222}Rn and ^{210}Pb between troposphere and stratosphere, by G. Lambert, J. Le Roulley and M. A. Kritz, in press, Tellus.

3) Dehydration of the upper tropical troposphere by gravity wave uplift: A case study from STEP, by H. B. Selkirk, L. Pfister, K. Kelly, M. A. Kritz and K. R. Chan, submitted to Geophysical Research Letters.

4) Constraints on troposphere-to-stratosphere exchange set by ^{222}Rn measurements made during NASA's STEP experiments, by M. A. Kritz, S. W. Rosner and E. F. Danielsen, to be submitted to the Journal of Geophysical Research.

A. ER-2 Polar Ozone Studies

B. D.W. Fahey, K.K. Kelly, M.H. Proffitt, A. F. Tuck
Aeronomy Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO 80303

C. Research Objectives

Polar ozone depletion has been addressed by NASA in both the Airborne Antarctic Ozone Experiment(AAOE) and the Airborne Arctic Stratospheric Expedition(AASE). Both missions involved the NASA ER-2 high altitude aircraft instrumented to measure chemical constituents and dynamical variables in the lower stratosphere. The contribution of the NOAA Aeronomy Laboratory has been two fold. First, Dr. Adrian Tuck, acting as Mission Scientist, has played a principal role in the planning and execution of each mission. Second, the total water, ozone, and reactive nitrogen measurements on the ER-2 were provided by instruments that have Laboratory members as Principal Investigators. More than ten flights of the ER-2 were made at latitudes above 60° over each pole in the late winter and early spring. The Arctic mission in early 1989 followed the close of the Antarctic mission by some fifteen months. This interim period was primarily dedicated to data reduction and interpretation and included several group meetings to present preliminary results. The effort has resulted in the publication of a number of peer-reviewed manuscripts in the Journal of Geophysical Research. A similar effort is now underway with the results from the Arctic mission.

D. Summary of Progress and Results

Under the direction of the Project Scientist and with the cooperation of a wide range of participants, both polar missions were a distinct logistic success. Each mission included at least ten flights of both the NASA ER-2 and DC-8 aircraft. As described by Tuck et al.[1989], mission planning required a variety of inputs including consultation with pilots, winds and temperature forecasts aloft and on the surface, satellite and radio sonde data, and atmospheric data from previous flights.

The synoptic and chemical evolution of the Antarctic vortex in 1987 is reconstructed by Tuck[1989] using the aircraft data and satellite and radio sonde data throughout the vortex in the late winter/early spring season. The evidence suggests that cooling, driven by synoptic scale forcing events, produces polar stratospheric clouds that intervene in the homogeneous gas phase chemical balance. The balance is shifted by the heterogeneous production of reactive chlorine and by the removal of total water and reactive nitrogen. In addition, the effects of this perturbed chemistry are transmitted to lower latitudes by the combination of horizontal mixing, diabatic descent, and advective transfer.

The latitude boundary of the chemically perturbed region on each Antarctic flight was noted, in part, by elevated levels of the ClO radical[Proffitt et al., 1989a]. Direct evidence for the removal of both total water and reactive odd nitrogen inside the boundary was obtained by the ER-2 instruments on nearly all flights[Fahey et al., 1989a, Kelly et al., 1989]. The removal is thought to occur by sedimentation of background aerosol particles that grow to sufficient size through condensation of nitric acid and total water. On one flight, ice crystals large enough to have appreciable sedimentation velocities were observed in a region of depleted gas-phase reactive nitrogen. The effects of chemical depletion on ozone were clearly observed from the measurements inside the vortex over the course of the 6 week mission[Proffitt et al., 1989b]. The total water vertical profile measurements outside the vortex provide evidence for the transport of dehydrated air out of the vortex throughout the mission. Evidence for mixing across the vortex boundary was noted in the ClO and ozone measurements near the boundary[Murphy et al., 1989a].

Measurements of nitrous oxide, total water, and reactive nitrogen with knowledge of potential temperature and potential vorticity were combined to show poleward movement of air near the vortex with on-going diabatic cooling[Proffitt et al., 1989c]. Cooling rates of 1.75K per day were estimated. The data suggest predominantly advective flow of ozone rich air across the boundary and into the ozone hole above 425K potential temperature and accompanying outflow to lower latitudes below this potential temperature.

Measurements of reactive nitrogen and aerosol particles on several flights, combined with temperature, pressure, and laboratory vapor pressure data, were shown to be consistent with the condensation of nitric acid trihydrate on pre-existing sulfate aerosol, several degrees above the local frost point[Fahey et al., 1989b]. The geographical distribution of clouds in the Antarctic as observed with satellite extinction measurements has been analyzed by Watterson and Tuck[1989] to show an asymmetry between the location of dense clouds and temperature mimima. The asymmetry is consistent with the restriction of cloud formation in large regions of the vortex because of removal of condensable material and with resupply of condensable material from lower latitudes over a narrow longitude range.

E. Publications

Fahey, D.W., D.M. Murphy, C.S. Eubank, K. Kelly, M.H. Proffitt, G.V. Ferry, M.K.W. Ko, M. Loewenstein, K.R. Chan, Measurements of nitric oxide and total reactive odd-nitrogen in the Antarctic stratosphere: Observations and chemical implications, J. Geophys. Res., in press, 1989a.

Fahey, D.W., K.K. Kelly, G.V. Ferry, L.R. Poole, J.C. Wilson, D.M. Murphy, M. Loewenstein, K.R. Chan, In-situ measurements of total reactive nitrogen, total water, and aerosol in Polar Stratospheric Clouds in the Antarctic stratosphere, J. Geophys. Res. 94, 11299-11315, 1989b.

Kelly, K., A.F. Tuck, D.M. Murphy, M.H. Proffitt, D.W. Fahey, R.L. Jones, D.S. McKenna, M. Loewenstein, J.R. Podolske, S.E. Strahan, G.V. Ferry, K.R. Chan, J.F. Vedder, G.L. Gregory, W.D. Hypes, M.P. McCormick, E.V. Browell, L.E. Heidt, Dehydration in the lower Antarctic stratosphere during late winter and early spring, 1987, J. Geophys. Res. 94, 11317-11357, 1989.

Murphy, D.M., A.F. Tuck, K.K. Kelly, K.R. Chan, M. Loewenstein, J.R. Podolske, M.H. Proffitt, S.E. Strahan, Indicators of transport and vertical motion from correlations between in-situ measurements in the Airborne Antarctic Ozone Experiment, J. Geophys. Res. 94, 11669-11685, 1989a.

Murphy, D.M., Time offsets and power spectra of the ER-2 data set from the 1987 Airborne Antarctic Ozone Experiment, J. Geophys. Res., in press, 1989b.

Proffitt, M.H., J.A. Powell, A.F. Tuck, D.W. Fahey, K.K. Kelly, K.R. Chan, A chemical definition of the boundary of the Antarctic ozone hole, J. Geophys. Res. 94, 11437-11448, 1989a.

Proffitt, M.H., M.J. Steinkamp, J.A. Powell, R.J. McLaughlin, O.A. Mills, A.L. Schmeltekopf, T.L. Thompson, A.F. Tuck, T.Tyler, R.H. Winkler, K.R.Chan, In-situ ozone measurements within the 1987 Antarctic Ozone Hole from a high altitude ER-2 aircraft, J. Geophys. Res., in press, 1989b.

Proffitt, M.H., K.K. Kelly, J.A. Powell, M.R. Schoeberl, B.L. Gary, M. Loewenstein, J.R. Podolske, S.E. Strahan, K.R. Chan, Evidence for diabatic cooling and poleward transport within and around the 1987 Antarctic Ozone Hole, J. Geophys. Res., in press, 1989c.

Tuck, A.F., R.T. Watson, E.P. Condon, J.J. Margitan, O.B. Toon, The planning and execution of ER-2 and DC-8 aircraft flights over Antarctica, August and September, 1987, J. Geophys. Res. 94, 11181-11222, 1989.

Tuck, A.F., Synoptic and chemical evolution of the Antarctic vortex in late winter and early spring, 1987, J. Geophys. Res. 94, 11687-11737, 1989.

Watterson, I.G. and A.F. Tuck, A comparison of the longitudinal distributions of polar stratospheric clouds and temperatures for the 1987 Antarctic spring, J. Geophys. Res., in press, 1989.

RESEARCH SUMMARY 1988-1989

A. COUNTING CONDENSATION NUCLEI IN THE LAST YEAR OF THE STRATOSPHERE-TROPOSPHERE EXCHANGE PROJECT
COOPERATIVE AGREEMENT NCC 2-470

B. James Charles Wilson, Principal Investigator
University of Denver

C. Research Objectives for 1988-1989

1. Finish a paper describing the condensation nuclei results for the Stratosphere-Troposphere Exchange Project (STEP) 1984 missions.
2. Do preliminary analysis on the STEP 1986-1987 data
3. Do preliminary analysis on Airborne Arctic Stratospheric Experiment (AASE) data from the ER-2 Condensation Nucleus Counter (ER-2 CNC) and the NASA owned Passive Cavity Aerosol Spectrometer (PCAS).
4. Report on the performance of the inlet of the PCAS.

D. Progress and Results

1. The STEP 1984 paper has been submitted for publication.
2. STEP 1986-1987 data have been reviewed and the 1987 data have been circulated to investigators.
3. The AASE data are being analyzed and preliminary results have been reported at AASE Science Team meetings.
4. The performance of the PCAS inlet is being analyzed and partial results have been reported.

E. Journal Publications

Wilson, J. C,,W. T. Lai, S. D. Smith, Measurements of Condensation Nuclei Above the Jet Stream: Evidence for Cross Jet Transport by Waves and New Particle Formation at High Altitudes, Journal of Geophysical Research, submitted 1989.

RESEARCH SUMMARY 1988-1989

A. COUNTING CONDENSATION NUCLEI IN THE ANTARCTIC OZONE MISSION
RESEARCH GRANT NAG 2-458

B. James Charles Wilson, Principal Investigator
University of Denver

C. Research Objectives for 1988-1989

1. Analyze data taken with the ER-2 Condensation Nucleus Counter (ER2-CNC) and other sensors in the Airborne Antarctic Ozone Experiment (AAOE) and publish results.
2. Suggest improvements to the NASA owned Active Cavity Aerosol Spectrometer (ASAS-X) so that it would perform reliably and quantitatively in the Airborne Arctic Stratospheric Experiment (AASE) so that accurate measurements of the background sulfate aerosol would be made in AASE.
3. Improve the sample inlet of the NASA owned Passive Cavity Aerosol Spectrometer (PCAS) so that sampling efficiency would be quantitatively known.
4. Develop field calibration techniques for the PCAS and implement them in the Airborne Arctic Stratospheric Experiment.
5. Operate the ER-2 CNC in the Airborne Arctic Stratospheric Experiment.

D. Progress and Results

1. The AAOE data were analyzed. Polar stratospheric cloud episodes were identified and described. They were classified according to whether or not the size distributions could be explained by existing theory. It was concluded that the data did not permit the denitrification mechanism to be determined. It was determined that sequential, rapid freezing of nitric acid trihydrate and then water such that most or all nuclei grew probably does not account for the denitrification of the polar vortex.

A strong correlation was observed between CN and N_2O. The correlation was positive in the region where the troposphere is the source of CN and was negative in the region of new particle formation. New particle formation was seen to occur throughout the chemically perturbed region and just outside of it.

2. It was recommended that the ASAS-X be replaced with the PCAS. This recommendation was accepted.
3. The sample inlet of the PCAS was modified and instrumented. The sampling efficiency was determined for each point in flight which resulted in a quantitative description of the sulfate aerosol encountered in AASE.
4. The PCAS was calibrated in the field during AASE. The calibrations were used in the development of a calibration which accounts for the refractive index of sulfate aerosol. The field calibrations provide important quality assurance for the PCAS data.

5. The ER-2 CNC was operated in AASE. Data were taken and provided to the project archive for 18 science and ferry flights.

E. Publications

Wilson, J.C., et al., Observations of Condensation Nuclei in the Airborne Antarctic Ozone Experiment: Implications for New Particle Formation and Polar Stratospheric Cloud Formation, *Journal of Geophysical Research*, in press, 1989.

Fahey, D. W. et al. (including J.C. Wilson),In-Situ Measurements of Total Reactive Nitrogen, Total Water and Aerosol in a Polar Stratospheric Cloud in the Antarctic, *Journal of Geophysical Research*, in press, 1989.