C. R. Geoscience 350 (2018) 341–346

Contents lists available at ScienceDirect

Comptes Rendus Geoscience

www.sciencedirect.com

External Geophysics, Climate (Aeronomy and Meteorology)

Two decades of polar ozone research via airborne science investigations: Addressing a NASA mandate in atmospheric composition

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ARTICLE INFO

Article history:
Received 4 December 2017
Accepted after revision 2 July 2018
Available online 9 October 2018

Handled by Paul A. Newman

Keywords:
Airborne science
Polar ozone depletion
Stratospheric ozone

ABSTRACT

The comprehensive investigation of polar ozone photochemistry and dynamics has required data obtained from as full a complement of available platforms as possible (ground-based, balloon, aircraft, and satellites). Perhaps the most detailed process studies have been conducted using measurements from aircraft, taking advantage of their targeting capabilities coupled with the potential for enabling measurements at high spatial and temporal resolution. The US National Aeronautics and Space Administration (NASA) conducted the first airborne science investigation of polar ozone in an effort to establish the causes of the recurring seasonal depletion of the Earth's stratospheric ozone layer over Antarctica that was identified in the mid-1980s. Subsequent airborne studies in the polar regions of both hemispheres benefitted from extensive successful collaborations among international scientists and the integration of the aircraft measurements with those obtained using ground-based, balloon-borne, and satellite instruments. This article provides an historical perspective of NASA's utilization of its airborne assets to advance our understanding of the chemical and physical processes that control the abundance of stratospheric ozone in both the Antarctic and Arctic.

1. Introduction

The interest in polar ozone chemistry predates these field measurement activities by many decades, beginning with the early work of S. Chapman in the late 1920s and continuing with studies by D. Bates and M. Nicolet (1950s), J. Hampson (1960s), P. Crutzen, H. Johnston, R. Stolarski and R. Cicerone, J. Lovelock, and M. Molina and F.S. Rowland (1970s). These "ozone pioneers" advanced the identification and understanding of the photochemical reaction cycles that controlled the natural distribution of stratospheric ozone and ultimately led to concerns about its possible catalytic destruction initiated by chemicals (e.g., chlorofluorocarbons – CFCs) introduced into the atmosphere via human activities. In response to this growing international attention, in 1975 the US Congress directed NASA via its FY1976 authorization bill

> "To conduct a comprehensive program of research, technology, and monitoring of the phenomena of the upper atmosphere."

This mandate to perform research on the depletion of the Earth's ozone layer soon led to the establishment of NASA's Upper Atmosphere Research Program (UARP).

However, NASA's airborne science research actually predates the UARP by more than five years, originating with the use of two Lockheed high-altitude U-2C aircraft beginning in 1971 for collecting Earth Resource data, primarily via multispectral photography. By 1979 NASA

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https://doi.org/10.1016/j.crte.2018.07.006
1631-0713/

had implemented a Climate Research Program under which an Aerosol Climatic Effects (ACE) special study was established to coordinate and build on much of the theoretical and experimental research being conducted within the NASA UARP and to complement the stratospheric aerosol observations provided by the Nimbus-G and SAGE research satellites. This coordination enabled a timely sequence of five flights of a specially instrumented U-2C aircraft during which measurements of gaseous constituents of the Mount St. Helens volcanic plume were obtained in the stratosphere (between 19 May and 17 June 1980).

2. Early high-altitude studies

The agency replaced the two U-2C's with two high-altitude airborne science aircraft (labeled ER-2's because of their civilian role in Earth Resource research) in 1981 and in 1989. The ER-2 has played an important role in NASA's Earth Science research primarily because of its ability to fly into the lower stratosphere at subsonic speeds, thereby enabling direct stratospheric sampling as well as virtual satellite simulation measurements. UARP's first dedicated airborne science mission was not conducted until more than a decade later, when it instrumented a U-2 and a (replacement) ER-2 for the Stratospheric-Troposphere Exchange Project (STEP) conducted primarily from Darwin Australia over the 4-year period 1984–1987. STEP was undertaken to investigate the transfer of mass, trace gases, and aerosols from the troposphere to the stratosphere and within the lower stratosphere, and to explain the observed extreme dryness of the stratosphere.

Tropospheric airborne campaigns: NASA also has a long history of tropospheric airborne studies within its Global Tropospheric Experiment (GTE), which was established as a contribution to the Global Tropospheric Chemistry Program (GTCP) recommended for initiation in 1984 by the US National Academy of Sciences in recognition of the central role of tropospheric chemistry in global change. NASA's GTE began utilizing large, extensively instrumented aircraft as primary research tools supplemented by ground-based measurements and later incorporated satellite data and model analyses into the field experiments. A first task of GTE was to foster development of new technologies and experimental techniques. These were evaluated through a series of rigorous intercomparisons under the Chemical Instrumentation Test and Evaluation (CITE) project. The initial GTE field expeditions (the Atmospheric Boundary Layer Experiment (ABLE) projects) were designed to probe the interactions between the biosphere and the atmosphere:

- CITE-1 11/83 Hawaii;
- CITE-1 4/84 Pacific-CA coast;
- ABLE-1 6/84 Barbados.

3. Field investigations of the Antarctic Ozone Hole

The 1985 discovery of the Antarctic Ozone Hole provided a strong impetus for more focused studies of the polar stratosphere. Specifically, the various scientific hypotheses that had been proposed about dynamical vs. chemical causes: lofting of low ozone air – the dynamics theory; nitrogen chemistry –the solar theory; heterogeneous halogen chemistry – the CFC theory drove the initial research formulation and measurement foci for ground-based and balloon measurements during the National Ozone Expeditions (NOZE I and II in 1986 and 1987, respectively). These campaigns preceded the airborne studies and were sponsored by the US National Science Foundation (NSF), NASA, the US National Oceanic and Atmospheric Administration (NOAA), the Chemical Manufacturer's Association (CMA), the US Navy and ITT Antarctic Services. Measurements were made using ozone and aerosol sondes together with ground-based microwave emission, solar IR, and visible absorption instruments. The data obtained provided the initial evidence for the role of chlorine chemistry involving heterogeneous reactions on the surfaces of ice crystals and helped to prioritize the soon to be conducted airborne measurements.

In 1987, UARP conducted the Airborne Arctic Ozone Experiment (AAOE), the first of its many field campaigns using instrumented aircraft to study polar ozone. The plans for AAOE were formulated in 1986 (following NOZE I) using instruments that had been flown in STEP plus others specifically built for the ER-2. Both the ER-2 (with a payload of instruments for in situ measurements) and the NASA DC-8 "flying laboratory" (with a payload of instruments for both in situ and remote-sensing measurements) were deployed to Punta Arenas, Chile in the Austral Spring of 1987. The spatial and temporal distributions of a large number of relatively short-lived chemical constituents that participate in the chemical reactions that affect the abundance of ozone were measured from both the ER-2 and DC-8. The temporal and spatial distributions of long-lived chemical tracers and dynamical variables were also measured in order to understand atmospheric motions. The data from these measurements and their interpretation have been published in numerous articles in the August 1989 and November 1989 Special Issues of the *Journal of Geophysical Research*. To summarize briefly, the weight of observational evidence strongly suggested that both chemistry and meteorology were responsible for the sizeable ozone perturbations observed and that meteorology unique to the Antarctic polar vortex set up the special conditions required for chlorine catalyzed ozone destruction. The observations of a developing anti-correlation between ozone and ClO concentrations during AAOE became known as the "smoking gun" that identified the chemistry responsible for the Antarctic Ozone Hole. There was now conclusive evidence for the ozone-destroying role of active chlorine from CFCs following its release from reservoir compounds via heterogeneous reactions on the surfaces of ice crystals. Specific details about these heterogeneous processes became available soon thereafter via research tasks supported by NASA's UARP. This rapid response was possible because of a robust and extensive UARP component in laboratory photochemistry and kinetics. Thus, several investigators were able to immediately refocus

their gas phase kinetics studies to studies of heterogeneous reactions.

4. First studies of the Arctic polar stratosphere

The attendant implications of the role of man-made CFCs in Antarctic ozone depletion raised concerns about the possibilities that similar ozone losses were possible in the Northern Hemisphere. Thus, it became a UARP priority to plan and conduct a follow-on field measurement campaign in the Arctic region as part of its responsibility to study the production and loss mechanisms for ozone in the polar stratosphere and to assess man's growing influence on the environment. Additional impetus for such a campaign was provided by the 1988 Report of the International Ozone Trends Panel, which documented that the largest decreases in total ozone were occurring in the Northern Hemisphere during January-February at latitudes near the edge of the Arctic vortex (World Meteorological Organization Global Ozone Research and Monitoring Project – Report No. 18, Geneva, Switzerland, 1988). The resulting Airborne Arctic Stratospheric Expedition (AASE) was coordinated by NASA with co-sponsorship from NOAA, NSF, and CMA. The campaign utilized the ER-2 and DC-8 based at Sola Airfield near Stavanger, Norway from January 1 to February 15, 1989. The wealth of data obtained indicated that the chemical composition of the Arctic polar stratosphere was highly perturbed, with chlorine monoxide and bromine monoxide concentrations comparable to those observed over Antarctica in 1987. Such high abundances could not be reproduced using models that did not include chemical reactions occurring on the surfaces of Polar Stratospheric Clouds (PSC's), thereby increasing confidence in the scientific understanding of PSC-induced, chlorine catalyzed, ozone depletion. It should be noted that such PSC-induced enhancements of reactive halogen had not been included in the assessment models initially used for the Montreal Protocol on Substances that Deplete the Ozone Layer. At the end of the AASE mission, the vortex air was primed for ozone destruction, with abundances of ClO radicals as high as 800 pptv and the potential for significant ozone loss to occur until the normal partitioning of chlorine species was reestablished.

Although this first Arctic campaign (AASE I) was similar to AAOE in many respects, it had been performed in the middle of winter rather than in late winter/early spring. Thus, a decision was made to conduct a second campaign over a more extended period of time and covering more of the Arctic region. The second Airborne Arctic Stratospheric Expedition (AASE II) used instruments on NASA's ER-2 and DC-8 aircraft (initially based in Fairbanks, AK and later in Bangor, ME) to examine the ozone-related chemistry and air motions of the lower stratosphere from August 1991 through late March 1992. Global maps of total ozone from the Total Ozone Mapping Spectrometer (TOMS) on board the Nimbus 7 satellite complemented the airborne data. As seen during AASE I, the Arctic stratosphere was chemically primed for ozone loss by mid-January. In fact, the measured concentrations of ClO of 1.5 ppbv were higher than any observed in previous aircraft missions in either the Arctic or Antarctic. However, an early warming of that region occurred in late January preventing the continued conversion of chlorine between its reservoir (unreactive) and reactive forms and, thus, precluding major ozone depletion. Ozone loss in the Arctic winter stratosphere of 1991/1992, while significant, was far less than the "Ozone Hole" levels seen over Antarctica. The extensive aircraft measurements during AASE II, combined with satellite and meteorological data, isolated two key variables that help to determine the amount of zone destroyed in any given year:

- the total amount of chlorine and bromine present in the stratosphere;
- the timing and vertical extent over which temperatures remain below the polar stratospheric cloud (PSC) threshold ($-78\ ^{\circ}C$).

The mission also revealed strong evidence for the influence of volcanic sulfate aerosols on stratospheric chemistry (particularly outside of the polar vortex). The observed influence of such aerosols on enhancing the abundances of ClO and BrO provided evidence linking the mid-latitude ozone losses discerned by satellite measurements over the previous decade to reactions involving chlorine and bromine free radicals. The results from AASE I and AASE II increased concerns about the vulnerability of the Northern Hemisphere ozone shield to depletion by man-made halogen and the US Congress voted 96-0 to accelerate the ban on CFCs.

5. Intensive airborne campaign period of the mid-1990s

In the years immediately following the completion of AASE II, NASA's UARP engaged in an intensive period of planning and conducting airborne science campaigns addressing a broad spectrum of objectives. The emphasis on stratospheric ozone depletion by halogen-containing chemicals became broadened to include studies of the atmospheric effects of aviation via a partnership with the Atmospheric Effects of Stratospheric Aircraft Program and the Atmospheric Effects of Aviation Project within NASA's High Speed Research Program. More comprehensive investigations of the global scale transport of atmospheric gases and particles were also included in the expanded science objectives. These airborne campaigns included the following:

- *Stratospheric Photochemistry, Aerosols and Dynamics Expedition (SPADE)* – This ER-2 mission was based in Moffet Field, CA in late 1992 and early 1993. Its primary focus was to study the chemical processes by which ozone could potentially be affected at altitudes that are most strongly influenced by stratospheric aviation;
- *Airborne Southern Hemisphere Ozone Experiment/Measurements for Assessing the Effects of Stratospheric Aircraft (ASHOE/MAESA)* – This was primarily an ER-2 mission conducted in four phases through the Antarctic winter of 1994 from Christchurch, NZ. A detailed discussion appears later in this article;

- *Stratospheric Tracers of Atmospheric Transport (STRAT)* – The primary goal of this campaign was to investigate the morphology of long-lived atmospheric tracers and dynamical quantities as functions of altitude, latitude, and season. The ER-2 was flow from Moffet Field, CA and Barbers Point Naval Air Station, HI in three phases from May1995 to February1996. The ER-2 measurements were augmented by the Observations from the Middle Stratosphere (OMS) high-altitude balloon component, which operated from July 1996 to December 1996 from Lynn Lake (Canada), Ft. Sumner (NM), and Juazeiro do Norte (Brazil);
- *Tropical Ozone Transport Experiment (TOTE)/Vortex Ozone Transport Experiment (VOTE)* – 17 flights of the NASA DC-8 covered latitudes from the tropics to the Arctic (December 1995 through February 1996). The primary mission objective was to examine the production and dispersal of small-scale filaments and streamers from both the polar vortex and tropical regions to better characterize filament transport. Some flights were coordinated with STRAT ER-2 flights during its Hawaii segment. Despite the new and extensive data collected during TOTE/VOTE, updated modeling studies were still not able to cannot account for the amount of ozone lost during a typical Arctic winter. In particular, the discrepancy pointed to a poor understanding of the balance of ozone production, loss and transport in the lower stratosphere in the early winter period when chlorine chemistry is not very active. This issue would motivate an extended return to the Arctic vortex a few years later;
- *Photochemistry of Ozone Loss in the Arctic Region In Summer (POLARIS)* – This campaign utilized the NASA ER-2 together with balloon platforms based in Fairbanks, AK during three deployment periods between mid-April and late September 1997. The mission focus was to provide an improved understanding of the seasonal behavior of polar stratospheric ozone as it changed from very high concentrations in spring to very low concentrations in autumn. A detailed discussion appears later in this article.

More comprehensive details about all of the airborne campaigns mentioned in this article can be found on the "Missions" section of the web page for NASA's Earth Science Project Office (https://espo.nasa.gov/content/ESPO_Missions_0).

The 1994 ASHOE/MAESA campaign mentioned above provided a unique opportunity to examine the causes of ozone loss in the Southern Hemisphere lower stratosphere and to investigate how the loss is related to polar, mid-latitude, and tropical processes. The measurements also provided information about stratospheric photochemistry and transport for assessing the potential environmental effects of stratospheric aircraft. Although most of the ER-2 measurements were conducted in the southern hemisphere from 70°S into the tropics, latitudes extending northward to upper Canada were also explored in conjunction with observations from the ground, balloons, and satellites. These combined observations have led to an improved understanding of the processes that lead to the Antarctic Ozone Hole and to ozone loss at mid-latitudes (e.g., the formation of PSCs, the conversion of chlorine from its reservoir forms to its ozone-destroying reactive form on cold, sulfate aerosols, and the return of chlorine to the inactive forms after all ozone is destroyed inside the Antarctic Ozone Hole). The transport mechanisms that redistribute the effects of these chemical processes were also better elucidated. For example, observations of variations of long-lived trace gases in the lower stratosphere shed new light on the transport of air both in the stratosphere and between the stratosphere and troposphere. Such transport of material is a central issue in assessing the atmospheric effects of emissions from aircraft flying in the lower stratosphere, particularly at mid- and low-latitudes.

The 1990s culminated with a unique airborne science campaign in the Arctic whose motivation extended beyond the need for better quantification of late winter/early spring polar ozone loss. The 1997 POLARIS ER-2 campaign was formulated to provide better understanding of the distinct seasonal cycle observed for polar stratospheric ozone. This cycle was not due to halogen catalyzed ozone destruction, but rather to the increased role of NO_x reaction cycles operating during the periods of prolonged solar illumination that occur in high latitude summer. Measurements of select species within the reactive nitrogen (NO_y), halogen (Cl_y), and hydrogen (HO_x) reservoirs; aerosols, and other long-lived species were used to evaluate the effectiveness of the respective catalytic reaction cycles for destroying ozone over a range of altitudes and latitudes throughout the campaign. The unique high latitude dataset on summer ozone changes in the lower stratosphere resulted in an improved understanding and prediction of photochemical partitioning and ozone loss rates over all seasons. Over the next two years, the analysis and modeling of data from more than 10 years of airborne science investigations of the polar atmosphere enabled a detailed assessment and an improved understanding of the combined effects of atmospheric chemistry and dynamics on stratospheric ozone and of existing uncertainties in projecting ozone recovery in response to the Montreal Protocol.

6. International airborne studies of polar ozone return to the Arctic

With this information in hand, plans had crystalized by the end of 1999 for mounting the largest field measurement campaign ever conducted to measure ozone amounts and changes in the Arctic upper atmosphere during the winter/spring. The NASA sponsored SAGE III Ozone Loss and Validation Experiment (SOLVE) would be staged jointly with the European Commission sponsored Third European Stratospheric Experiment on Ozone (THESEO 2000). This collaborative venture would involve scientists from the US, Europe, Russia, and Japan in conducting measurements of ozone and other atmospheric gases. The unprecedented international pooling of observational, modeling, and intellectual resources was aimed at providing a better understanding of the possibilities of continuing ozone loss and of expected ozone recovery over

the subsequent decades. Several instrumented aircraft were based at the "Arena Arctica" in Kiruna Sweden (the NASA ER-2 and DC-8, the German DLR Falcon, the French ARAT, and a French Lear Jet). In addition, heavy-lift balloons were launched from the Esrange balloon and rocket facility, while small balloons and ground-based instruments contributed measurements from numerous Arctic locations. Satellite measurements completed the broad spectrum of research capabilities. In fact, in addition to the detailed process scale science objectives, a key mission goal had been to acquire correlative measurements for validating the Stratospheric Aerosol and Gas Experiment (SAGE) III instrument aboard the Russian Meteor-3M satellite, and to use the SAGE III measurements of the vertical structure of aerosols, ozone, water vapor and other trace gases in the Arctic upper troposphere and stratosphere to help in quantitatively assessing high latitude ozone loss. To further facilitate a comprehensive examination of the processes that control ozone amounts at mid to high latitudes throughout the Arctic winter, the campaign was conducted over three intensive measurement phases centered on early December 1999, late January 2000, and early March 2000.

The complexity of the SOLVE/THESEO-2000 scientific planning was matched by the logistical complexities associated with conducting a stratospheric field mission involving more than 350 scientists, technicians and support workers in a remote Arctic location. However, in addition to the existence of a unique hangar facility at the Kiruna Airport this site was chosen because of its close proximity to the edge of the polar vortex throughout most of the winter period and well inside of the vortex on many days in January. In addition, having the deployment base located near the climatological average coldest region in the Arctic facilitated the study of chlorine activation on polar stratospheric clouds (PSCs) and other cold aerosol particles. This proximity provided an opportunity for an unprecedented achievement in airborne science when the NASA ER-2 completed its first science flight through Russian airspace on January 27, 2000. The NASA DC-8 flying laboratory flew through Russian airspace at the same time, making atmospheric measurements in conjunction with the ER-2.

The hard work that went into the planning and implementation of SOLVE/THESEO was rewarded with outstanding scientific success due to both the high level of international cooperation on all fronts and to some fortuitous meteorological conditions. Temperatures that were below normal in the polar lower stratosphere over the course of the 1999–2000 Arctic winter resulted in extensive PSC formation. Large particles containing nitric acid trihydrate were observed for the first time under conditions that demonstrated that denitrification could occur without the formation of ice particles. Heterogeneous chemical reactions on the surfaces of the PSC particles produced high levels of reactive chlorine within the polar vortex by early January. This reactive chlorine catalytically destroyed about 60% of the ozone in a layer near 20 km between late January and mid-March 2000. The determination of ozone loss by rate-limiting radical reactions up to 90°N latitude led to improved understanding of key photochemical parameters and of the evolution of ozone-destroying forms of chlorine. These, in turn, enabled better predictions of future ozone levels as chlorine levels declined in response to the provisions of the Montreal Protocol. Unfortunately, because of a prolonged delay in the launch of the SAGE III satellite instrument, direct intercomparison activities could not be conducted during the SOLVE campaign and the various process scale science objectives became the primary focus of the mission. Nevertheless, SOLVE/THESEO data were used in conjunction with measurements from several other satellite instruments to provide a level of indirect validation of SAGE III.

An opportunity for direct validation of the SAGE III instrument soon arose when NASA scheduled the SOLVE-II mission at the Arena Arctica in close collaboration with the European Commission sponsored VINTERSOL (Validation of International Satellites and study of Ozone Loss)/EUPLEX (EU Polar Stratospheric Cloud and Lee Wave Experiment) campaign during the Arctic winter of 2002–2003. The SOLVE-II mission component utilized the DC-8 aircraft and balloon measurements to acquire coincident correlative data with SAGE III as well as with the atmospheric chemistry instruments onboard the POAM and ENVISAT satellite missions, thereby enhancing comparison and science activities utilizing these data sets. The VINTERSOL/EUPLEX mission component added airborne measurements from the DLR Falcon and the high-altitude Russian Geophysica and balloon measurements by the French National Centre for Space Studies (CNES) team. The joint mission strategy benefitted extensively from the SOLVE/THESEO experience. As a result, the measurements from the international array of aircraft and balloon instruments have led to a more detailed understanding of the chemical and dynamical processes associated with stratospheric ozone loss in the Arctic and at Northern Hemisphere mid-latitudes.

7. Retrospective

Two decades of progressive achievements in polar ozone research via airborne science have demonstrated the high value of international cooperation in all aspects of mission formulation, planning, and implementation as well as in the analysis and interpretation of the data. Equally important, however, has been the critical partnership that continued to grow between science and operations. Without this partnership, the ability to provide the scientific underpinning of the Montreal Protocol and its Amendments and Adjustments would not have been possible. Airborne field campaigns and fundamental laboratory investigations, together with space-based missions and ground networks, have become international measurement resources and have enhanced the interactivity among the four research elements required to address ozone depletion and recovery. The interplay and feedback between each of these elements (measurement, verification, analysis, and description) provide a solid foundation for international assessments that are key to the development of the improved prognostic capabilities

needed to evaluate the potential impacts of changing atmospheric composition due to chemistry/climate coupling. The success of the Montreal Protocol serves as a clear justification for the enormous investments that have been made internationally in laboratory research on atmospheric photochemical processes as well as in planning and implementing scientific research and measurement campaigns.