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L. G. HANSCOM FIELD, BEDFORD, MASSACHUSETTS

Neutral Composition Measurements of the Mesosphere and Lower Thermosphere

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Abstract

A recently developed rocket-borne mass spectrometer which employs a LN_2 cryosorption pump has successfully obtained the first direct measurements of the major and minor neutral constituents of the mesosphere and lower thermosphere. The mass analysis is performed using an rf quadrupole spectrometer with an electron impact ion source. Measurements were obtained from two flights, and the preliminary results from one of these are presented. The constituents measured include N_2, O_2, Ar, O, CO_2, O_3, and several other minor constituents. Measurements were obtained from 70 to 120 km. Atomic oxygen measurements were obtained between 85 km and apogee on both up-leg and down-leg of the flight. The atomic oxygen measurements were obtained at lower altitudes by switching the ion source potentials on alternate spectral scans to ionize with 20-eV electrons. The lower ionizing energy reduces that portion of the signal at 16 amu due to dissociative ionization of O_2. Number density profiles of individual constituents were obtained by normalizing the relative abundances to a density model.

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Neutral Composition Measurements of the Mesosphere and Lower Thermosphere

1. INTRODUCTION

The study of the physical and chemical processes occurring between 60 and 120 km is important because of the strong influence of these processes on the behavior of the neutral atmosphere and the lower ionosphere, and yet this is one of the least explored regions. The mesosphere and lower part of the thermosphere, as this region is referred to, lies above the altitude range of balloon measurements and below the region where satellites can be orbited. Thus, rocket-borne experiments are used to perform in situ measurements of parameters of interest. Over the past several years, a new instrument has been developed to study the atmospheric composition in this region. The objective of the experiment is to measure both the major and the minor species which exist in concentrations down to one part in 10^8 (or about $10^4/cm^3$). Rocket measurements of the composition in this region are complicated by several factors. First, the atmospheric pressure is sufficiently high so that any conventional mass analyzer must be pumped to eliminate collisional scattering. Second, sampling of the lower part of the region (below about 95 km for a vehicle with an apogee of 120 km) will be influenced by a shock formed around the instrument. Third, the measured gas sample will be obtained under continuum, transitional, and free molecular flow conditions over the 60- to 120-km range.

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The instrument which has been developed consists basically of an rf quadrupole mass spectrometer with a cylindrically-symmetric electron impact ion source and a liquid nitrogen cryosorption pump. The instrument has been studied and tested under laboratory conditions for instrument stability and linearity. These tests also investigated the cryosorption pump characteristics under static conditions and the dynamic conditions which simulate the pressure response expected in flight. Tests have been performed in wind tunnels to study the effects associated with the shock at lower altitudes. The wind tunnel results have shown that measurements behind the shock can be related experimentally and theoretically to the ambient species density. The experiment has been flown successfully twice, and the preliminary results from one of those flights are discussed. The first instrument was launched on 7 March 1970 during the solar eclipse program. The second flight, which is discussed in this paper, took place on 20 November 1970 and was part of a coordinated rocket program designated as ALADDIN I (Atmospheric Layering And Density Distribution of Ions and Neutrals). The vehicle was launched from Eglin Air Force Base at 17:23 CST at a solar zenith angle of 98^{o}.

2. DESCRIPTION OF THE EXPERIMENT

The instrument is shown in Figure 1 as an artist's conceptual view and in Figure 2 as a schematic drawing of the cross section. The experiment is referred to as NACS (Neutral Atmospheric Composition Spectrometer). The quadrupole analyzer utilizes rods of 0.381 cm diameter and 7.6 cm length. The ion source is mounted directly below the entrance orifice plate which also serves as the anode for the ion source. The filament and accelerator electrodes are cylindrically symmetrical. The objective of this design is to provide a high density of ionizing electrons in the region where the gas sample effuses through the orifice. This configuration enhances the signal for the incoming sample compared to the background signal

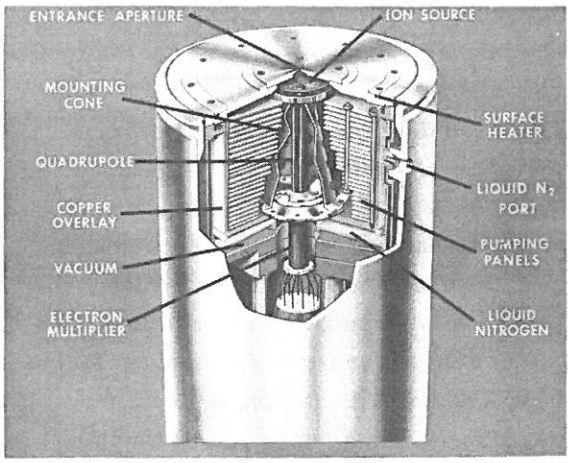

Figure 1. The NACS Instrument Showing the Quadrupole Mass Spectrometer and Cryosorption Pump Configuration

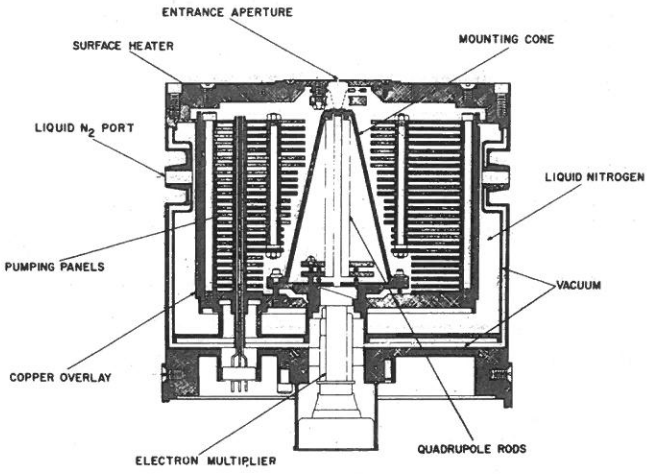

Figure 2. Cut-away View and Details of the Experiment (for reference, the instrument diameter is 6 in.)

from unpumped gases in the instrument and/or from particles reflected in the ion source region. Since several species of interest are chemically reactive, reflected particles may not give a true measure of the atmospheric species. Another advantage of this type of source is that its symmetry minimizes effects associated with the vehicle motion. Disadvantages lie in the fact that background species can contaminate the measurement (at higher altitudes), and the hot filament can distort the species concentrations when the gas sample flow is small. For example, with a very small gas flow the molecular oxygen signal may be lowered because of reactions with the hot filament (this effective pumping of O_2 has also been reported by Gross and von Zahn, 1971). The electron ionizing energy is switched between two values on alternate mass spectra. This capability allows a better measurement of some of the species (particularly atomic oxygen) because the fraction of molecules which are dissociatively ionized in the ion source is considerably less for 20-eV ionizing electrons than for 100-eV ionizing electrons (less than 0.5% for 20-eV electrons compared to about 10% for 100-eV electrons).

The cryosorption pump removes the incoming gas flux and maintains a low background pressure in the region of the analyzer. The cryosorption pump was made by bonding a 5-Å molecular sieve onto aluminum plates which have been cross-mill grooved. These plates are sandwiched into a dewar which is chilled with liquid nitrogen. High pumping speeds are obtained for atmospheric species (about 100 liters/sec for nitrogen) except for minor species such as H, He, and Ne. The gas capacity of the pump is many times greater than the gas load presented in a single rocket flight. The holding time after removal of liquid nitrogen is considerably longer than the typical 6-min rocket flight.

The neutral species were sampled through a 0.125 cm diameter orifice. The ions which are formed by electron impact ionization of the gas effusing through the orifice are drawn by electric fields into the quadrupole mass analyzer. The pressurized electronics package below the quadrupole provides the ac and dc voltages to the quadrupole rods. The ac voltage, at a fixed frequency of about 4.5 MHz, is

controlled by a dc voltage sweep such that the dc/ac voltage ratio is maintained at 0.224 to within 1% over the mass range. The ions focused through the quadrupole are detected by a secondary emission multiplier with a gain of about 10^4. The output of the electron multiplier is measured with a 5-decade logarithmic electrometer.

Figure 3 shows a sample of the flight record. The lower trace is a monitor of the ac voltage which provides a linear mass calibration for the instrument. The upper trace shows samples of the spectra obtained. The mass range is swept from 3 to 90 amu in about 1 sec. The sweep is followed by a short step wherein the dc voltage is switched to ground potential, and all masses greater than 50 amu are transmitted (the strong focusing mode). Some of the mass identifications are: 16 (O and contributions from dissociatively ionized O_2 and H_2O), 18 (H_2O), 28 (N_2), 32 (O_2), 40 (Ar), 44 (CO_2), 14 (dissociatively ionized N_2), and 34 ($^{16}O^{18}O$ isotopes of oxygen). Most of the current at high mass numbers is due to background (mainly diffusion pump oil). The electrometer calibration pulses at 10^{-8} and 10^{-10} amp occur at the beginning of each spectrum scan. The high minimum background current ($\sim 10^{-10}$ amp) is due to uv radiation (with possibly some contribution from metastable species) produced by electron collisions with the incoming gas sample in the ion source. The magnitude of this background is proportional to the pressure in the ion source region. In the present instrument configuration (multiplier in line of sight with the ion source), this background signal limits the effective dynamic range of the instrument to concentrations of about one ppm. The spectrum on the left was made with 20.3-eV ionizing electrons, and the one on the right was made with 38.1-eV electrons. The energy indicated is the total voltage between the filament and the anode; therefore, the effective

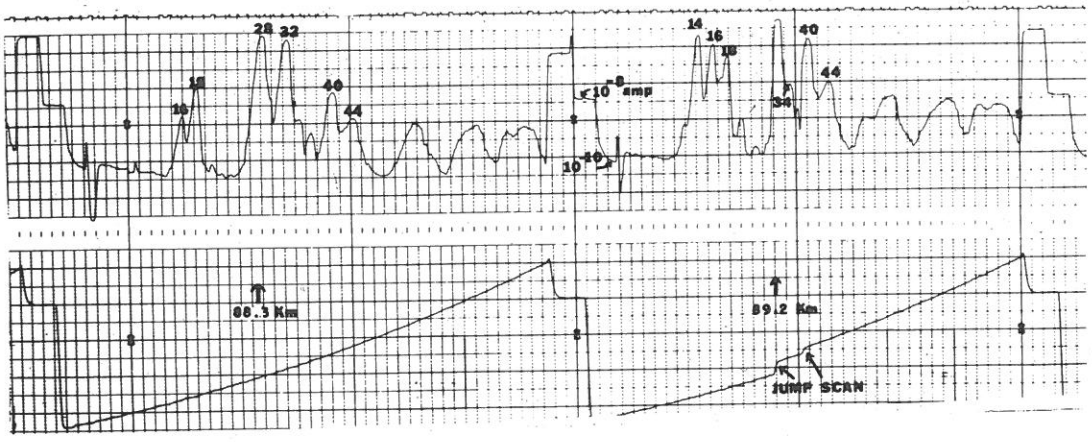

Figure 3. Strip Chart Record Showing Two Flight Spectra with Different Electron Ionization Energies. The upper trace is the output from a 5-decade logarithmic electrometer, and the lower trace is a monitor of the rf voltage which provides a mass calibration

ionizing energy may be slightly less than these values. The electron energy is alternated between these two values on each spectrum. During those portions of the flight where the currents for N_2, O_2, and Ar would have been far off scale, a jump in the mass scan is used to prevent an electrometer saturation problem.

3. FLIGHT DATA AND RESULTS

The payload which included a gyro for attitude information, a tracking beacon, telemetry system, and other support instrumentation was launched to an altitude of 121 km by a Nike-Iroquois vehicle at 17:23 CST on 20 November 1970. The flight was made in coordination with other measurements which included measurements of density, temperature, horizontal and vertical wind components, and turbulence. Following burn-out of the second stage rocket motor, the NACS vehicle developed a very large coning angle. The spin frequency of the vehicle was about one rps, and the coning period was about nine sec. Figure 4 shows the variation of the angle of attack of the instrument axis with the velocity vector. Since the mass scan period was about one sec, the maximum angular change over a mass spectrum amounted to about 40 deg. The data, in its preliminary form, presented here have not been reduced using the aspect information. A large part of the modulation caused by the vehicle motion is effectively removed from the data presented in this paper since only current ratios of the masses of interest with molecular nitrogen are used.

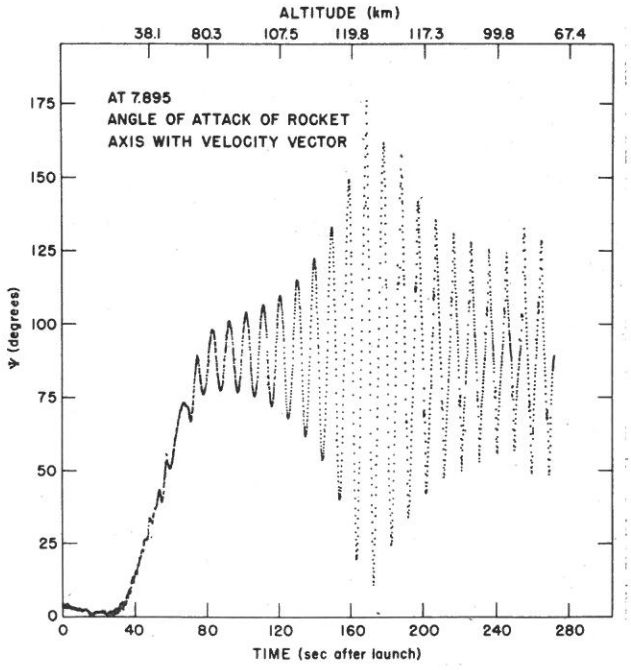

Figure 4. The Vehicle Motion from the Reduced Gyro Data. The large coning angle is not typical of rocket vehicle motion and resulted from spin-pitch coupling

The results presented were obtained by the following procedure. The current for each mass of interest was divided by the N_2 current in that spectrum. When the nitrogen mass peak was off scale, its value was obtained by taking the proper ratio of the 14-amu peak which comes from dissociative ionization of N_2. The current ratio was then divided by the ratio of ionization cross sections of the mass of interest and molecular nitrogen. The laboratory calibrations had shown that the only measurable mass discrimination in this instrument was due to the differences in ionization cross sections.

Ionization cross sections used for most of the species were those determined by Rapp and Englander-Golden (1965). The atomic oxygen cross section used was that determined by Fite and Brackmann (1959), and the ionization cross section of ozone was assumed to be the same as that of molecular oxygen (see comparison of Herron and Schiff, 1956). The only 20-eV data presented in this paper are for atomic oxygen. Since no ionization cross section measurements are available for atomic oxygen in the 20-eV range, the atomic oxygen data shown for this case were normalized at altitudes above 105 km to the profile obtained in the higher energy case. Figures 5 and 6 show current ratios for several of the constituents of interest. In Figure 5, the O_2/N_2 and the Ar/N_2 ratios are shown for both the low- and high-eV case. Ascent and descent data between 85 and 121 km are shown. However, the useful data extends below 70 km. Much of the scatter in the data points will be removed when corrections for the vehicle attitude are made. Some of the modulation in the ratios above 110 km is due to the fact that a significant portion of the argon and oxygen signals were due to the slower pumping of argon and oxygen compared to nitrogen. The modulation caused by the vehicle motion will aid in removing the signal due to the background gases. Thus, it is expected that the O_2 and Ar signal near apogee will be reduced somewhat in the final analysis. Figure 6 shows the 16-amu signal ratio to the O_2 signal for both electron energies on ascent and descent. The reason for using the lower ionizing energy is evident in this figure. The amount of 16-amu signal from dissociative

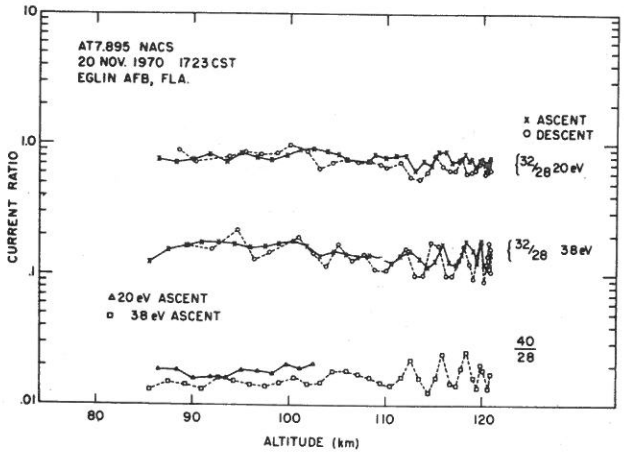

Figure 5. The Ratio of the Molecular Oxygen and Argon Signals to Molecular Nitrogen Signal as a Function of Altitude for Two Ionization Energies

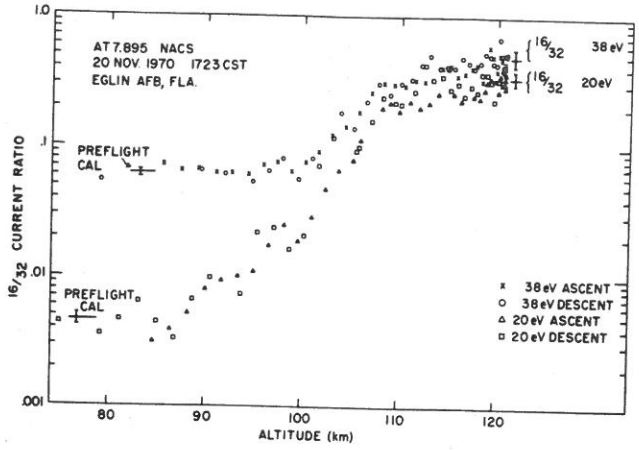

Figure 6. The Ratio of Atomic Oxygen to Molecular Oxygen Signal as a Function of Altitude for Two Ionizing Energies. The lower ionizing energy provides data to a lower altitude before the 16-amu contribution from dissociative ionization of molecular oxygen becomes predominant

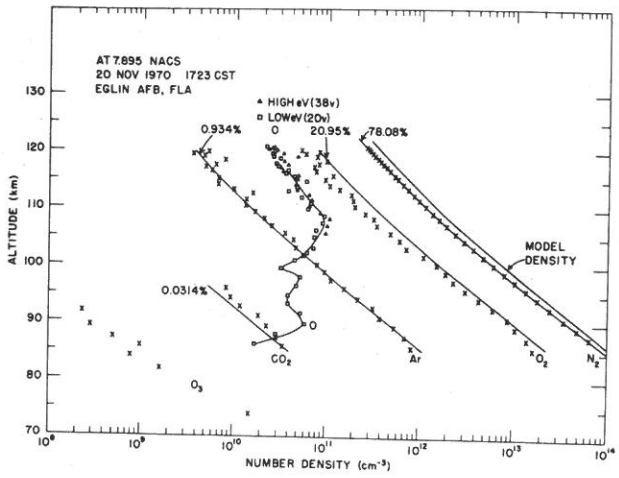

Figure 7. The Number Densities for Several of the Species Resulting From a Preliminary Treatment of the Data

ionization of molecular oxygen is shown as a pre-flight calibration level. The amount of dissociative ionization is considerably less in the low-ev case and therefore allows for lower altitude measurements.

In Figure 7, the altitude profiles for several of the constituents are shown from this analysis of the data. A model density profile from U. S. Standard Atmosphere Supplements, 1966 was assumed, and densities for each of the constituents were inferred based on the model. The nitrogen profile was taken to be 78% of the model density, and the current ratio divided by the cross section ratio was used to obtain a profile for each of the constituents. The solid lines shown for N_2, O_2, Ar, and CO_2 represent the ground level mixing ratio of those constituents. The solid line shown for the O measurement is only to aid the eye in following the points. The O_2 profile follows the ground level mixing ratio to an altitude of about 100 km, and then its atmospheric percentage drops as expected due to solar dissociation. The apparently localized increases in the O_2 profile near 112, 116, 118, and 120 km actually correspond to times when the instrument was facing away from the velocity vector. The apparent increases are due to the fact that the nitrogen pumping speed was higher than that of O_2, and the background O_2 signal contributed a measurable amount to the incoming gas sample

when the instrument was pointed into the wake of the vehicle. The argon profile shows a similar effect at high altitudes. Both the O_2 and Ar profiles will be decreased somewhat above 110 km in the final analysis because of the contribution from background gases. The CO_2 profile was not plotted above 95 km because of a significant background contribution. Each of these three profiles show reasonably good agreement with the ground level mixing ratio curves below 100 km. The atomic oxygen profile was obtained by removing the contribution to the 16-amu peak from dissociative ionization of both O_2 and H_2O and then using its current ratic divided by the ionization cross section ratio in the same manner as for the other constituents. This procedure provides an O profile between 105 km and apogee for the high ionizing energy case. The low ionizing energy data were normalized by a single multiplicative factor in the 105- to 120-km region and provides an extension of the O profile to lower altitudes. The profile shows irregularities which are significant and indicate a more complex variation in the O profile than might have been supposed. Large modifications of model-generated O profiles have been produced with models which include atmospheric dynamics (Keneshea et al, to be published). The absolute value of the O profile densities may well be questioned. However, studies are being carried out in an effort to gain some insight into the problems of recombination of such a reactive specie. The arrangement of the entrance geometry is such that a large fraction of the ions are formed before the incoming gas sample has had the opportunity to strike an instrument surface. At altitudes below 90 km where a shock is fairly well developed around the front of the instrument, several collisions with the surface (some of which result in recombination) can occur before the gas is sampled through the entrance orifice.

All of the profiles in Figure 7 (except for ozone) are those obtained on vehicle ascent. The down-leg O_3 densities were obtained in the same way as the other profiles using the assumption that the O_3 cross section is the same as that of O_2. The O_3 profile obtained is higher by factors between 2 and 10 than several other ozone profiles obtained by optical techniques (Evans and Llewellyn, 1970). The optical measurements of the ozone profile are typically representative of ozone profiles with solar zenith angles near 90^o. The profile shown in Figure 7 corresponds to a solar zenith angle of about 99^o when the ozone densities would be expected to increase in the early evening.

Figure 8 shows a spectrum of the down-leg data in the vicinity of 70 km for the case of low energy electrons. The main reason for showing this spectrum is to point out the fact that significant high mass peaks are observed. Some, or possibly all, of these peaks at high mass numbers may result from ion molecule reactions. However, it is worthwhile at this time to note them as follows:

Altitude Range (km)	Mass Number (amu)
60 - 76	56
60 - 85	60
60 - 76	68 ± 1
73 - 84	77 ± 1

Results incorporating the vehicle attitude data will be reported later. Future flights of this experiment will study the properties and variations of the mesosphere and lower thermosphere.

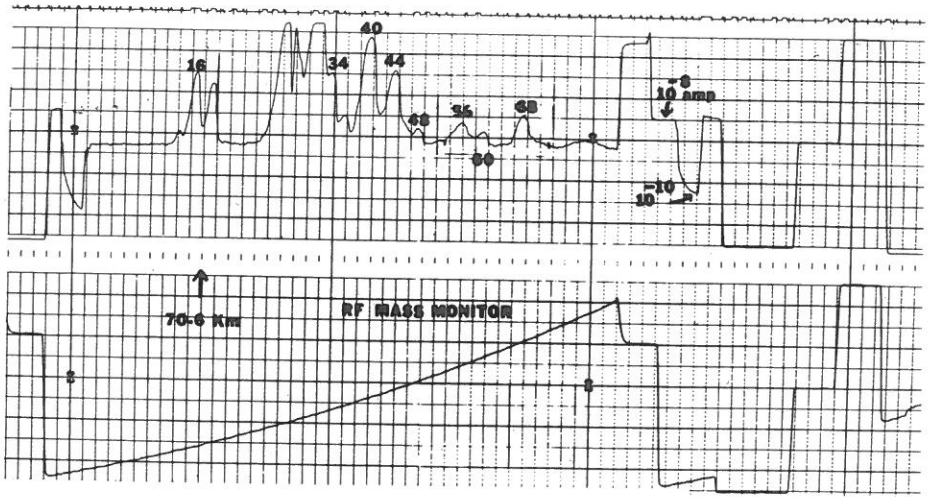

Figure 8. A Low Altitude Spectrum of the Low Ionizing Energy Mode Showing Examples of High Mass Peaks at 56, 60, and 68 amu

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13. ABSTRACT

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