Gravitational separation of the stratospheric air over Syowa, Antarctica and its connection with meteorological fields

Shigeyuki Ishidoya1 | Satoshi Sugawara2 | Yoichi Inai3 | Shinji Morimoto3 | Hideyuki Honda4 | Chusaku Ikeda4 | Gen Hashida5 | Toshinobu Machida7 | Yoshihiro Tomikawa5,6 | Sakae Toyoda8 | Daisuke Goto5,6 | Shuji Aoki3 | Takakiyo Nakazawa3

To examine gravitational separation of the stratospheric air over Antarctica in austral summer, we collected air samples from altitudes of 10 to 30 km over Syowa Station (69.0°S, 39.6°E) using balloon-borne cryogenic samplers for the period 1998–2013, and then analyzed them for $\delta^{15}$N of N$_2$, $\delta^{18}$O of O$_2$, $\delta$(Ar/N$_2$) and $\delta^{40}$Ar. The normalized mass ratio “$\delta$,” calculated using their measured values, decreases with increasing altitude, implying an upward enhancement of the gravitational separation effect. The observed stratospheric $\delta$ profiles are generally well reproduced by a two-dimensional atmospheric model, but the model tends to underestimate the observed $\delta$ values in the middle stratosphere above 26 km. We also observe interannual variation in the $\delta$ vertical profile in the middle stratosphere. A backward trajectory analysis suggests that this variation is attributable to different horizontal mixing of the stratospheric air over Antarctica from year to year.

KEYWORDS
Balloon-borne cryogenic air sampler, Gravitational separation of the atmosphere, Meteorological field, Stratosphere over Antarctica

1 | INTRODUCTION

Recent technical advances in the measurement of major atmospheric constituent gases have made it possible to observe their molecular diffusive separation. For example, Adachi et al. (2006) reported, from their observations above a desert surface, a thermal diffusive separation of Ar and N$_2$ during the nighttime when a strong surface temperature inversion appears. Ishidoya et al. (2006, 2008, 2013) and Sugawara et al. (2018) also found gravitational separation of...
the stratospheric air from equatorial to polar regions, based on their analyses of the Ar/N ratio and the isotopic ratios of N₂, O₂ and Ar. The magnitude of gravitational separation is determined by a balance between mass-independent atmospheric transport, that is, advection and eddy diffusion, and mass-dependent molecular diffusion in the atmosphere. The contribution of molecular diffusion to gravitational separation can be evaluated quantitatively by using molecular diffusion coefficients of related air molecules, which are determined by physical parameters such as molecular mass, temperature and number density of air molecules (Banks and Kockarts, 1973). Therefore, we can use the observed gravitational separation as an indicator of advection and eddy diffusion in the stratosphere, after correcting for the molecular diffusion effect.

In our previous studies, the average vertical gradient of gravitational separation of the stratospheric air was examined in terms of long-term changes in the atmospheric circulation (Ishidoya et al., 2013), with less attention on fluctuations found in each observed vertical profile. However, such fluctuations are clearly observed, for example, over Syowa (69.0°S, 39.6°E), Antarctica (Ishidoya et al., 2008). By inspecting a relationship between the vertical profiles of gravitational separation and the N₂O mixing ratio observed in the stratosphere, Ishidoya et al. (2008) suggested three different transport processes as a function of altitude. In this paper, we re-examine these transport processes in more detail by presenting all our data on gravitational separation taken so far in the stratosphere over Syowa, including those unpublished in Ishidoya et al. (2008), and discussing irregular fluctuations of gravitational separation profiles in terms of meteorological transport fields over the Antarctic region.

2 | METHOD

Stratospheric air samples over Syowa were collected at altitudes between 10 and 30 km using two kinds of cryogenic samplers (Honda et al., 1996; Morimoto et al., 2009) mounted on board large scientific balloons. Details of our air sampling and observation results of greenhouse gases have been described in Aoki et al. (2003), Morimoto et al. (2009) and Goto et al. (2017). In this study, the air samples collected on January 3, 1998, December 26, 2003, January 5, 2004, January 4, 2008, December 31, 2012 and January 10, 2013 were analyzed for Δ₁⁵N of N₂ and Δ₁⁸O of O₂ using a mass spectrometer (Finnigan, Palo Alto, CA, MAT-252; Ishidoya et al., 2003) with the respective reproducibility of ±12 and ±26 per meg. The mass spectrometric method was also applied to the air sampled in 2012 and 2013 to determine Δ₁⁵N of N₂, Δ₁⁸O of O₂, δ(At/N₂) and δ⁴⁰Ar with the reproducibility of ±5, ±7, ±35 and ±22 per meg, respectively (Thermo Scientific, Waltham, MA, Delta-V; Ishidoya et al., 2013). Here, Δ₁⁵N is defined by

\[
\delta^{15}N = \left[ \frac{(15N^{14}N/14N^{14}N)_{sa}}{(15N^{14}N/14N^{14}N)_{ref}} - 1 \right] \times 10^6 \text{ (per meg) } (1)
\]

where subscripts “sa” and “ref” denote the sample and the reference, respectively. By replacing ¹⁵N and ¹⁴N by ¹⁸O and ¹⁶O (Ar and N₂, or ⁴⁰Ar and ³⁶Ar), respectively, δ¹³⁸O (δ(At/N₂) or δ⁴⁰Ar) is defined. In this study, δ¹⁵N of N₂, δ¹⁸O of O₂, δ(At/N₂) and δ⁴⁰Ar of the stratospheric air samples are given as deviations from annual averages of the corresponding values observed continuously at the surface in Tsukuba (36.3°N, 140.7°E) in 2013 (Ishidoya and Murayama, 2014). The respective variables of the stratospheric and surface air were determined against our primary standard air, which was prepared by drying natural air at the dew point temperature lower than −80°C and then stored in a 48-L aluminum high-pressure cylinder in 2011 (Ishidoya and Murayama, 2014).

3 | RESULTS AND DISCUSSION

Figure 1a shows vertical profiles of Δ₁⁵N of N₂, Δ₁⁸O of O₂, δ(At/N₂) and δ⁴⁰Ar observed in the stratosphere over Syowa on December 31, 2012 and January 10, 2013. Based on our understanding of gravitational separation, we should expect to see all these variables to decrease with increasing altitude (Ishidoya et al., 2013). Indeed, as seen in Figure 1a, all the variables generally decrease in value going upward. To confirm that such vertical profiles are formed by gravitational separation, we plot δ¹⁸O/2, δ(At/N₂)/12 and δ⁴⁰Ar/4 against Δ₁⁵N in Figure 1b. A linear regression analysis gives slope values of 1.89 ± 0.04, 11.1 ± 0.5 and 4.2 ± 0.2 (±1σ) per meg/per meg for δ¹⁸O/Δ₁⁵N, δ(At/N₂)/Δ₁⁵N and δ⁴⁰Ar/Δ₁⁵N, respectively. These slope values are close to 2, 12 and 4 expected from gravitational separation, which are proportional to the difference between the mass numbers of related molecules, rather than the corresponding ratios of 1.55 ± 0.02, 16.2 ± 0.1 and 2.75 ± 0.05 calculated based on thermally diffusive fractionations of air molecules (Ishidoya et al., 2013; Ishidoya et al., 2014). Therefore, the gravitational separation effect of major components of air is also observable in the stratosphere over Antarctica, as well as over Japan (Ishidoya et al., 2013) and the equator (Sugawara et al., 2018).

Using the values of Δ₁⁵N, Δ₁⁸O and δ(At/N₂) measured in 2012 and 2013, we define a new “δ” as an indicator of the degree of gravitational separation:

\[
\delta = \frac{1}{3} [\delta^{15}N + \delta^{18}O/2 + \delta(At/N₂)/12] \quad (2)
\]

Because the measurement precision is relatively worse for δ⁴⁰Ar than for δ¹⁵N, δ¹⁸O/2 and δ(At/N₂)/12, we omit inclusion of δ⁴⁰Ar/4 from the calculation of δ. For the data before 2008, δ is defined as:
By defining δ in this way, the mass number differences of related molecules are normalized, and smaller δ indicates stronger gravitational separation effect (Ishidoya et al., 2013). Figure 2 shows the calculated vertical profiles of δ over Syowa. The average vertical profile of δ over Sanriku (39.1°N, 141.8°E), Japan, observed on June 8, 1995, May 31, 1999, August 28, 2000, May 30, 2001, September 4, 2002, September 6, 2004, June 3, 2006 and June 4, 2007 (Ishidoya et al., 2008, 2013), and that over Biak (1.1°S, 136.1°E), Indonesia observed on February 22, 24, 26, and 28, 2015 (Sugawara et al., 2018) are also shown in the figure. As seen from the figure, δ decreases with increasing altitude, and an average difference of δ between the tropopause and the middle stratosphere (altitudes of 25–30 km) is approximately 50 per meg. This difference is similar in magnitude to those observed over Sanriku at northern mid-latitudes, but much larger than 11 per meg found in the equatorial region over Biak. On the other hand, the difference in δ between the tropopause and the stratosphere below 25 km is larger over Syowa than over northern mid-latitudes by about 10 to 20 per meg (Ishidoya et al., 2008). Based on the simulation results using a 2-dimensional model of the middle atmosphere (SOCRATES) developed by the National Center for Atmospheric Research (e.g., Huang et al., 1998), Ishidoya et al. (2013) and Sugawara et al. (2018) reported that the gravitational separation effect at the same altitude is generally enhanced going poleward. Our observed latitudinal differences in gravitational separation are consistent with the model-simulation results.

The δ values calculated using the SOCRATES model for each season under the same conditions as the control run of Ishidoya et al. (2013) are shown in Figure 2 for comparison with the observational results. The δ values observed over Syowa are reproduced relatively well by the model, while some observed δ values above 26 km are clearly higher than the model results. It is also seen from Figure 2 that the vertical distribution of δ above 25 km fluctuates noticeably from year to year and exceeds significantly the seasonality of δ simulated by the SOCRATES model. On the other hand, the vertical δ gradients observed in the lower-to-middle stratosphere over Japan are reproduced relatively well by the model (see fig. 4a in Ishidoya et al. (2013) and fig. 1a in

\[
\delta = \frac{1}{2} \left( \delta^{15}N + \delta^{18}O/2 \right) 
\]
Ishidoya et al. (2008)), although year-to-year fluctuations are apparent in the observed vertical profiles to some extent. The year-to-year fluctuation, defined as \(\pm 1\sigma\) deviation of the observed \(\delta\) values from their least-squares regression line, is \(\pm 9\) per meg for 10 to 25 km and \(\pm 21\) per meg for 25 to 30 km. These facts suggest that the SOCRATES model fails to incorporate some atmospheric transport processes necessary for reproducing the summertime vertical profile of \(\delta\) above 25 km over Syowa.

In order to understand the large year-to-year \(\delta\) variability observed above 25 km over Syowa, we investigate it in terms of interannual variation in the stratospheric meteorological fields in the Southern Polar region. Typically, the Antarctic seasons are characterized especially by the development of the polar vortex in the winter, its breakdown in the spring, and leading to a dominance of high pressure in the summer. We examined seasonal variations in the vertical velocity of residual mean circulation and geopotential heights before each respective observation date, and 30-day backward trajectories for individual observations to interpret the year-to-year variation in \(\delta\) over Syowa.

Figure 3 shows vertical velocities of the residual mean circulation at the standard isobaric surfaces of 10, 20 and 30 hPa at 69°S observed from January of 1997, 2003 and 2012 to March of the following year. These vertical velocities were calculated using the European Centre for Medium-Range Weather Forecasts reanalysis meteorological data (ERA-Interim) (Andrews et al., 1987; Dee et al., 2011). The ERA-Interim meteorological data are also used for the discussion made below. It is clearly seen in Figure 3 that the descending of air gradually strengthens from the austral summer to winter, due to a residual mean circulation driven primarily by Rossby waves in the austral winter. It is also obvious that the descending velocity decreases suddenly to around zero in November, in association with the destruction of the polar vortex. The destruction timing of the polar vortex is characterized by the stratospheric final warming (SFW) event in the Southern Hemisphere (e.g., Hardiman et al., 2010). Hirano et al. (2016) reported that the SFW event occurred earliest in 2012–2013 and latest in 1997–1998 during the period covered by this study. Their finding is consistent with our result that the change in wind direction from descending to ascending is found earlier in 2012–2013 than in 1997–1998 and 2003–2004, as seen in Figure 3. This suggests that the year-to-year differences in the \(\delta\) value observed over Syowa are related to year-dependent destruction timings of the polar vortex. We also examined time series of residual mean meridional wind, but any further insight was not obtained.

Figure 4 shows temporal changes in the geopotential height distribution at 10- and 20-hPa surfaces over the Southern Polar region in December 1997, 2003 and 2012. The spatial patterns of the geopotential height field are nearly concentric at 10 and 20 hPa. However, a closer inspection shows that the isopleths are skewed especially at 20 hPa, crossing those at 10 hPa in early December of 1997 and 2003. The skewed isopleths imply that the geostrophic wind has a meridional component, and the crossed isopleths indicate the existence of wind shear by which vertical mixing of air could be enhanced (Appenzeller and Holton, 1997) for 10–20 days before the observation. In 2012, the isopleths of the geopotential height are concentric at 10 and 20 hPa, but no crossing of isopleths is observed, indicating that air masses are transported zonally and the vertical air mixing is less likely to occur. These results suggest that the higher \(\delta\) values (weaker gravitational separation) observed above 25 km over Syowa during the period December 2012 to January 2013 are associated with meridional transport and/or vertical mixing that are different from those observed in January 1998, December 2003 and January 2004. On the other hand, the \(\delta\) values observed at the highest altitudes in 2003 and 2004 are comparable to those above 25 km in 2012 and 2013. These high values would be due to weak meridional air transport to be discussed later.

In order to examine atmospheric transport processes in the middle stratosphere over Syowa, we calculated 30-day backward trajectories for individual observations using 3-D ERA-Interim wind field. Following Inai et al. (2013), the starting points of trajectories were set every 0.1° latitude × 0.1° longitude in a circular area with a radius of 1° latitude/ longitude centered at Syowa Station. Air parcels were released from all grid points at an altitude and time coinciding with each air sampling. Horizontal distributions of the
30-day backward trajectories are shown in Figure 5. It is seen from this figure that the trajectory patterns for the period of December 2012 to January 2013 are compact and concentric, consistent with the geopotential heights shown in Figure 4. The trajectories above 29 km in December 2003 and 28 km in January 2004 also show concentric distribution. On the other hand, the trajectories for January 1998, as well as those below 29 km in December 2003 and 27 km in January 2004, show significant divergence, with a number of trajectories passing over higher latitude regions than the observation locations. By comparing the trajectories with the vertical profiles of $\delta$ shown in Figure 2, it is possible to conclude that the stratospheric air that passed over higher latitudes within the last 30 days prior to the observation time tends to show lower $\delta$ values.

The backward trajectory analysis suggests that the large fluctuation in $\delta$ observed in the middle stratosphere above 25 km over Syowa is likely caused by the year-to-year variation in the horizontal mixing of air. On the other hand, our observations show that the $\delta$ values obtained in the lower stratosphere below 25 km over Syowa in different years are relatively similar to each other, as seen in Figure 2. This is consistent with the result of numerical simulations using the SOCRATES model that gravitational separation is less sensitive to changes in the horizontal mixing in the lower stratosphere than in the middle stratosphere (Sugawara et al., 2018). It is also found from Figures 2 and 5 that the middle stratospheric $\delta$ values calculated using the SOCRATES model agree with the values observed for the air that passed over higher latitudes within the last 30 days prior to the observation time, but the model underestimates all the observed $\delta$ values above 28 km except for the value at 28.5 km in January 2004. This underestimation suggests that the summertime transport of middle stratospheric air from the mid-latitudes to the polar region is not well represented in the SOCRATES model.
It should be noted that the period of 30 days employed in the present trajectory calculations is much longer than the period of 8 days used by Friedrich et al. (2017) in which uncertainties in the trajectory were examined based on the ERA-Interim and other reanalysis meteorological data. In this regard, it is evident from Figure 5 that the low δ value at 28.5 km in January 2004 is not associated with the air mass transport from higher latitudes, for which uncertainties in the trajectory analysis may be partly responsible. In order to better understand the relationship of spatiotemporal changes in gravitational separation with regional atmospheric transport in more detail, further studies will be required for the evaluation of the uncertainties in the 30-day trajectories.

Possible contributions of air masses with lower δ from higher latitudes to the observed fluctuations of δ over Syowa may also be examined in terms of stratospheric CO2 and SF6 ages (e.g., Waugh and Hall, 2002) derived from CO2 and SF6 mixing ratios of the air samples used for the δ15N and δ18O analyses (our unpublished data). The air age (CO2 and SF6 ages) is defined as the time elapsed between the intrusion of an air mass into the stratosphere from the upper equatorial troposphere and reaching the observation site. Because SF6 is decomposed by ultraviolet absorption and reactions with electrons in the mesosphere while CO2 remains stable, any air mass observed in the polar stratosphere with the SF6 age older than the CO2 age, it is likely that the mesospheric air with low SF6 mixing ratios descended into the stratosphere through the polar vortex (e.g., Ray et al., 2017). By defining the difference in the air age as Δage (Δage = SF6 age – CO2 age), we calculated the average Δage to be 1.5 ± 0.5 years for the stratospheric air samples collected above 24 km over Syowa in January 1998, December 2003 and January 2004. On the other hand, the air samples collected in December 2012 and January 2013 yielded an average Δage of 0.7 ± 0.5 years. The comparison of the calculated Δage with the corresponding δ indicates that air masses with large Δage values tend to have low δ values, suggesting a detectable influence of the mesospheric air in the stratosphere at polar latitudes even in the summer. The correspondence of strong gravitational separation to large Δage was also reported by Sugawara et al. (2018), based on their observation data in the equatorial stratosphere. However, the average Δage for the air samples

**FIGURE 5** Horizontal distributions of the 30-day backward trajectories of air parcels released for individual observations over Syowa, Antarctica. Colors denote the elapsed days from the starting point (purple) to the end point (red) of each trajectory.
collected above 29 km in December 2003 and January 2004 (1.9 ± 0.3 years) is slightly larger than 1.5 ± 0.5 years, which deviates from the inverse relationship between $\Delta_{age}$ and $\delta$ value. Further observations are needed to examine this discrepancy.

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ORCID

Shigeyuki Ishidoya https://orcid.org/0000-0001-7448-2899

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