Interactive comment on "Long term changes in the upper stratospheric ozone at Syowa, Antarctica" by K. Miyagawa et al.

The history of depletion of stratospheric ozone over Antarctica in the austral spring is well known and well documented from the time of the discovery [*Farman et al*, 1985]. The rapid increase in this depletion during the 1990s [*Solomon*, 1999] is now being followed by a leveling off of the annual depletion [*Salby et al.*, 2011]. The cause of this depletion again is well known; excess stratosphere chlorine from man-made chlorofluorocarbons (CFCs) entering into chemical reactions at various heights and temperatures [*Molina and Roland,* 1974; *Solomon et al.*, 1986]. The first stage of the atmospheric response to the implementation of the 1987 Montreal Protocol and subsequent revisions, a lessening in the ozone reduction has been noted in the Northern mid-latitudes [*Newchurch et al*., 2003]. The second stage response, a statistically significant ozone increase has not yet been detected [*Solomon et al.*, 2005; *WMO*, 2011]. Over Antarctica, no stage of recovery however has yet to be detected [*WMO*, 2011; and references therein].

The Dobson ozone spectrophotometer measurements provide not only estimates of total ozone column above the surface, but also vertical profile information. The Umkehr technique has been used since the 1930s [*Götz et al*., 1934] to derive the vertical ozone profile from zenith sky measurements. The algorithm is described elsewhere [*Petropavlovskikh et al.*, 2009]. We investigate the information in the data from Syowa ground-based station (69.0S, 39.6E) which has been collecting ozone profiles from Dobson Umkehr measurements since 1977 [*Miyagawa et al.*, 2009a], for long-term changes in the upper and middle stratospheric ozone. The re-evaluated Umkehr data-sets from the Syowa Japanese ground-based Dobson station [*Miyagawa et al*., 2009b] are used for this study (Section 2), with some further data analysis, and more data.

The primary investigation is upper stratosphere where the variability is controlled by chemical reactions influenced by the anthropogenic ozone-depleting substances (ODS) and should be the place for detection of ozone recovery [*Newchurch et al.,* 2003]. The high inter-annual variability in the Antarctica region and the changes in the atmosphere from other factors since 1977 must be well quantified to be able to detect the signal due to ODS. We will be using a number of well-known proxies (Table 1) to remove the effects of both dynamic and long term variables, as well as the EESC record, using various age-of-air scenarios. The ODS concentrations are often represented by the time series of the EESC (equivalent effective stratospheric chlorine) concentration that are based on the known budgets of the anthropogenic and natural sources of CFCs, halogens and bromines, understanding of the life-time of these species in the atmosphere, and estimates of the transport/mixing of ODS from the tropical troposphere to stratosphere and from tropics to high latitudes by a meridional transport and vertical mixing (Brewer Dobson Circulation, or BDC). The ODS concentrations over Polar region (or EESC curves) are defined by the "age of the transported air" [*WMO*, 2011].

We complement and verify the analysis of ground-based measurements with the recently released SBUV Merged Ozone Dataset Version 8.6, Overpass data.

1. Introduction

Even at the current stage of enhanced control of CFCs production, the balloon, satellite and ground-based ozone observations in the Antarctic region show high year-to-year variability in the ozone hole region, and reveal a delay in ozone recovery as compared to the mid-low latitude trends. The ozone variability in the upper stratosphere is primarily controlled by chemical reactions influenced by the anthropogenic ozone-depleting substances (ODS), as compared to the dynamically driven troposphere, and thus is expected to provide information for early detection of ozone recovery [*Newchurch et al*., 2003]. However, continuous increase of greenhouse gases in the atmosphere and enhanced cooling of the

stratosphere can affect recovery of the ozone layer [*Waugh et al.*, 2009; *Li et al*., 2009; *Oman et al.*, 2010 and references therein]. While the ODS levels have been significantly decreasing after reaching a maximum in 1998, stratospheric temperature has been decreasing in response to the increase in concentrations of greenhouse gas chemicals such as water vapor, CO_2 and methane. Cooling of the upper stratosphere reduces ozone loss rates (mainly through the $O + O_3 = 2O_2$ chemical cycle), and therefore results in faster ozone recovery [*Rosenfield et al.*, 2002]. Lower temperatures due to climate change in the polar lower stratosphere increase heterogeneous conversion of chlorine and bromine reservoir species to more active forms, leading to a decrease in ozone. In this paper we discuss the long-term trend of polar upper stratospheric ozone in relation to the ODS concentration changes over the Antarctica. The ODS concentrations are often represented by the time series of the EESC (equivalent effective stratospheric chlorine) concentration that are based on the known budgets of the anthropogenic and natural sources of CFCs, halogens and bromines, understanding of the life-time of these species in the atmosphere, and estimates of the transport/mixing of ODS from the tropical troposphere to stratosphere and from tropics to high latitudes by a meridional transport and vertical mixing (Brewer Dobson Circulation, or BDC). The ODS concentrations over Polar region (or EESC curves) are defined by the "age of the transported air" [*WMO*, 2011].

In addition to the chemical reaction mechanism, stratospheric ozone is radiatively coupled to atmospheric dynamics, and can be affected by meridional transport (BDC), or stratospheric temperature changes related to heat transport through wave activity and to variations in the polar vortex [*Jiang et al.*, 2008; *Hitchman and Rogal*, 2010]; ozone depletion has been shown to affect the strength of the polar vortex [*Thompson and Solomon*, 2002; *Perlwitz et al*., 2008, *Fogt et al.*, 2009; *Li et al.*, 2010; *Oman et al.*, 2010, *Son et al.*, 2010]. While the SH has weaker planetary wave activity than the NH, the change in the BDC contributes strongly to ozone variability over the South Pole region. A relationship between theAntarctic stratospheric ozone depletion, strengthening of the polar circulation in the SH stratosphere and the Southern Annular Mode (SAM) index has been found in observations and discussed in many papers [*Kushner et al.*, 2001*; Thompson and Solomon*, 2002*; Thompson and Solomon*, 2005*; Arblaster and Meehl*, 2006; *Jiang et al.*, 2008; *Fogt et al.*, 2009*; Wang et al.*, 2011]. The intra-annual ozone-SAM link has also been identified in coupled chemistry-climate models [*Perlwitz et al.*, 2008; *Son et al.*, 2008; and *Fogt et al*., 2009; *Thompson et al*., 2011]. The largest variability in the Antarctic stratosphere associated with the SAM signal is observed in the September through December period. A positive trend in SAM is also related to a stronger planetary wave activity in stratosphere and prolonged westerly anomalies [*Li et al.*, 2010]. At the same time, Antarctic polar ozone long-term depletion has influenced the austral summer tropospheric circulation (SAM) in the Southern hemisphere [*Perlwitz et al.*, 2008]**.** Correlation of the SAM index and Syowa ozone data interannual variability is addressed in this paper.

The El Nino Southern Oscillation (ENSO) [*Diaz and Markgraf*, 1992] is the largest climatic signal observed in many meteorological data sets, not just over the equator, but also in remote areas. ENSO is found to be responsible for the generation of a type of Rossby wave that travels from the Equator towards the Poles, and are responsible for coupling between the stratosphere and troposphere over Antarctica [*Hoskins and Karoly*, 1981; *Randel*, 1987, 1988; *Perlwitz and Harnik*, 2004; *Turner*, 1994; *Hu and Fu*, 2009; *Ialongo et al*., 2011]. The ENSO signal at sea level pressure and at high latitudes is amplified when it is in phase with SAM [*Fogt and Bromwich*, 2006], for example, during the 1990s. The ENSO signal for the time period between 1958 and 2001 is highly anti-correlated with SAM at the surface, but has weaker correlation (negative) at 50 and 30 hPa atmospheric levels [*Haigh and Roscoe, 2006*]. It shows high correlation during austral summer, when 25 % of the SAM inter-annual variability is linearly related to ENSO [*L'Heureux et al., 2006*]. Therefore, the relationship between ozone variability and ENSO or SAM signal is highly non-linear, where the SAM's decadal changes enable shifts in climate regimes [*Yuan and Yonekura*, 2011]. The change in sea level pressure affects the westerlies, which therefore create a stronger polar vortex, and thus affect polar ozone [*Hu and Fu*, 2009].

Impact of the 11-year solar cycle and the stratospheric equatorial Quasi-Biennial Oscillation (QBO) on the Southern Annular Mode (SAM) in late winter/spring is discussed in the paper by *Kuroda and Yamazaki* [2010]. Under an Easterly QBO mode, the critical latitude shifts more toward the Pole, and

planetary waves propagate to the stratosphere, which strengthens residual BDC and reduces ozone in the lower stratosphere. Similar results are also found by *Haigh and Roscoe* [2006], when SAM variability is analyzed in relation to the compound of solar and QBO signals as compared to using these signals separately. Moreover, significant stratospheric warming over the high latitudes in the Southern Hemisphere has been derived from Microwave Sounding Unit observations [*Hu and Fu,* 2009]. Warming trends are related to the increases of the stratospheric eddy-heat fluxes during austral late summer and fall, which are caused by the increase of the wave propagation into stratosphere and adiabatic heating. *Hu and Fu* [2009] found close correlation with Sea-Surface Temperatures, which are influenced by the anthropogenic increase in the green-house gases. Future stratospheric warming would influence ozone recovery in the upper stratosphere [*Waugh et al*., 2009; *Stolarski et al.,* 2010]. Moreover, analysis of the coupling between stratospheric ozone variability and climate changes that are affecting dynamics and meteorology of the atmosphere and oceans in the Southern Hemisphere are of great importance for future ozone level predictions.

Since both QBO and ENSO influence polar stratospheric temperatures and wave activity [*Hamilton*, 1998], and therefore ozone, it is difficult to separate the signals. Using an equatorial zonal wind data archive for the 1958-2008 periods, *Taguchi* [2010] found ENSO-related changes in the phase and amplitude of the QBO signal that are related to the wave activity at the Equatorial region. However, there are modeling studies that find stratospheric polar temperature changes between El Nino (EN) and La Nina (LN) periods even when no QBO signal is included in the model [*Sassi et al.*, 2004, *Garcia-Herrera et al*., 2006; *L'Heuraux and Thompson*, 2006]. Clear differences in general circulation and total ozone distribution in the Southern hemisphere are found between LN and EN years [*Garfinkel and Hartmann*, 2007*; Hitchman and Rogal,* 2010]. *Hitchman and Rogal* [2010] showed that in months of August-September-October there is a strong relation between UTLS anticyclone near the tip of South Africa (SA), wave activity, and reduction of ozone distribution that is driven by ENSO events. Syowa station is located along the longitude that intersects the location of the South African High anticyclone, and therefore is affected by the variability in regional meridional circulation and by ENSO variability, i.e. strong anticyclone forms near SA during EN phase, and stronger transport of ozone-poor air-masses to high latitude stratosphere occurs, thus affecting year-to year variability in stratospheric ozone over Syowa station.

Global warming (climate change) also affects stratospheric ozone layer depletion through sensitivity of the chemical reactions to the atmospheric temperature change. When the concentration of greenhouse gases increases in the atmosphere, the temperature of the troposphere rises (greenhouse effect) [*Turner et al.*, 2006]. At the same time more of the outgoing Earth infrared radiation becomes trapped in the troposphere instead of reaching the stratosphere. Therefore, this missing IR radiation does not balance out stratospheric cooling due to emission from the increase in stratospheric water vapor and carbon dioxide gases. The change of the heat balance reduces temperature in the stratosphere [*Schindell*, 2001]. This effect is especially large in the polar lower stratosphere, and it strengthens the occurrence of the PSCs in winter. Therefore, it strengthens the Antarctic ozone-hole chemistry. Chemical processes on the surface of the PSC clouds release active chlorine and create conditions for rapid ozone destruction in both Antarctic and the Arctic region when the sun returns [*Solomon et al*., 1986; *Solomon*, 1999]. Conversely, the temperature decline of the upper stratosphere leads to the increase in ozone (O_3), via an increase in the rate of the binding reaction of an oxygen molecule (O_2) and an oxygen atom (O). On the other hand, water vapor in stratosphere converts to hydrogen oxide (OH) molecules that destroy ozone, while OH also reacts with CFCs that create Chlorine compounds that also destroy ozone. Additionally, a change in the transport pattern known as the BDC tends to increase water vapor in the stratosphere, which also affects ozone layer [*Garcia and Randel*, 2008]. It is important to distinguish the influence that climate change has on ozone layer depletion from the effects of ODS. While stratospheric ozone concentrations will recover with the reduction in ODS the impact of the stratospheric ozone change on the temperature trend will be altered by the effects of the increase in the greenhouse gases [*Stolarski et al.*, 2010].

Continuous monitoring by high quality ground-based observations is very important for detecting long-term changes in the ozone global distribution. The mechanisms that affect ozone on different time scales

need to be investigated. The Dobson ozone spectrophotometer measurements provide not only estimates of total ozone column above the surface, but also vertical profile information. The Umkehr technique has been used since the 1930s [*Götz et al*., 1934] to derive the vertical ozone profile from zenith sky measurements. The algorithm is described elsewhere [*Petropavlovskikh et al.*, 2009]. The re-evaluated Umkehr datasets from the Syowa Japanese ground-based Dobson station [*Miyagawa et al*., 2009b] are used for this study (Section 2). A further data refinement is described below.

The paper by *Miyagawa et al.* [2009a] describes trends determined in the austral spring-summer season from ozone profiles measurements at Syowa station using techniques similar to *Reinsel et al.* [2002]. In this paper we are using the same data set (plus 4 more years), similar statistical methods, and investigating the contributions from other atmospheric parameters (Section 3). We compare the ozone trend derived from Umkehr measurements at Syowa station with the trend derived from the homogenized Solar Backscatter UV (SBUV) instrument satellite data that is restricted to the station over-pass (see details in sub-section 3.4). We investigate ozone trends in the upper stratosphere with respect to changes in temperature while also accounting for the changes in ODS. Moreover, we find that long-term changes in ozone at high latitudes and in the upper stratosphere cannot be fully explained by the ODS related chemistry. Therefore, we include additional explanatory parameters in the statistical model and study their correlation with the long-term changes in the Umkehr ozone profile record over Syowa.