Behavior of Stratospheric Chlorine Nitrate During a Recovery Phase of Polar Ozone Destruction in both Hemispheres

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Improved Limb Atmospheric Spectrometer (ILAS) monitored the profiles of ozone and ozone-related species, such as aerosols (or PSCs), nitric acid, nitrogen dioxide, nitrous oxide, methane, and water vapor on a regular basis, 14 times daily at high latitudes (57.1_N - 72.7_N and 64.3_S - 88.2_S) from November 1996 until the end of June 1997. In addition to those chemical species, the newest retrieval algorithm of the ILAS, version 6.0, succeeded to derive chlorine nitrate (ClONO2) profiles for both hemispheres. Nakajima et al. (paper in preparation) compared the ILAS-measured ClONO2 data with three balloon-borne measurements at around 69_N in March though May 1997, and concluded that the ClONO2 data have enough quality for scientific use in 15 to 35 km altitudes, though they may be about 30% lower values than the validation data. In spite of this systematic bias, the continuous data set of ClONO2 in the polar regions simultaneously observed with many chemical species is variable to understand chemical propagation during a polar ozone depletion event. The measurement period of ILAS covered a recovery phase after Antarctic ozone hole in 1996 and the whole period of severe Arctic ozone depletion in 1996/1997. The boreal winter of 1996/1997 is well known for long-lasting polar vortex resulting in frequent PSC appearance (Hayashida et al., JGR, 2000) and significant ozone loss over the Arctic (Terao et al., JGR, 2002). The time series of the ClONO2 in the Arctic lower stratosphere indicated outstanding enhancement (as high as about 2 ppbv at 475 K and 550 K isentropic surfaces) in late February and in March 1997. As well known, heterogeneous reactions on PSCs convert chlorine reservoir species into active chlorines that
destroy ozone. The active chlorines produce predominantly ClONO2 rather than HCl upon deactivation when a sufficient abundance of ozone is available. On the other hand, the behavior of ClONO2 in the recovery phase of the Antarctic ozone hole was quite different. At the 475 K surface, some ClONO2 mixing ratio data indicated extremely low values, corresponding to very low ozone (less than 1 ppmv) in November 1996 in the polar vortex. At the boundary region, ClONO2 mixing ratio was much higher, corresponding to higher ozone mixing ratio (2.5-3.5 ppmv). As examined by Prather and Jaffe [JGR, 1990], HCl are preferably formed under ozone-depleted conditions because of the shift of Cl/ClO and NO/NO2 ratios toward Cl and NO, resulting in suppression of ClONO2 formation and enhancement of HCl formation through the reaction of Cl and CH4. Under the assumption of photochemical equilibrium, [ClONO2]/[HCl] ratio has a linear correlation to [O3]/[CH4] (Sen et al., JGR, 1999; Voss et al., JGR, 2000). The analysis of [ClONO2]/[HCl] ratio combined with corresponding HCl/HALOE data shows a good linear correlation in May and June 1997 for the northern hemisphere. The linear correlation is also found even in the southern polar vortex in November 1996, which suggests quasi-photochemical equilibrium condition in the Antarctic late spring.