1. Introduction
   The simultaneous gas and aerosol retrieval (see below) is applied to the ILAS data for the first time, as the ILAS Version 7 data set. Here, the similar algorithm is applied both for the ILAS and ILAS-II data, as the ILAS Version 8 and the ILAS-II Version 3 data sets, respectively. Although the ILAS V7 data is validated (Oshchepkov et al., 2006), no technical report on the data quality of these two data sets has been published so far. Both of the data sets are based on the HITRAN 2004 database, so that these two data sets are adequate to any comparisons between ILAS and ILAS-II.

2. Simultaneous gas and aerosol retrieval (Oshchepkov et al., 2005, 2006) is applied for altitudes between 13 and 35 km. See Appendix 1 for details. It describes ILAS-II V3 but it is the same for ILAS V8.

3. For altitudes between 36 and 40 km, and below 12 km, a nongaseous contribution correction method (Yokota et al., 2002) is applied.

4. As a result of simultaneous retrieval, aerosol extinction coefficients for all of the 44 pixel elements can be deduced. Moreover, size distributions by particle volumes for assumed aerosol types can also be deduced. These subsets of the data are provided (ilas-request@nies.go.jp). See Appendix 2 for file descriptions. It describes ILAS-II V3 but it is the same for ILAS V8.

5. Positive constraints for both gas and aerosol parameters were introduced through a penalty function for spectral inversion. (Oshchepkov et al., 2005)

References:

Oshchepkov, S., Y. Sasano, T. Yokota, N. Uemura, H. Matsuda, Y. Itou, and H. Nakajima (2005), Simultaneous stratospheric gas and aerosol retrievals from broadband infrared occultation measurements, Applied Optics, 44, 22, pp. 4775-4784.

Oshchepkov, S., Y. Sasano, T. Yokota, H. Nakajima, N. Uemura, N. Saitoh, T. Sugita, and H. Matsuda (2006), ILAS data processing for stratospheric gas and aerosol retrievals with aerosol physical modeling: Methodology and validation of gas retrievals, J. Geophys. Res., 111, D02307, doi:10.1029/2005JD006543.

Yokota, T., H. Nakajima, T. Sugita, H. Tsubaki, Y. Itou, M. Kaji, M. Suzuki, H. Kanzawa, J. H. Park, and Y. Sasano (2002), Improved Limb Atmospheric Spectrometer (ILAS) data retrieval algorithm for Version 5.20 gas profile products, J. Geophys. Res., 107, D24, 8216, doi:10.1029/2001JD000628.

APPENDIX1: Version 3.0 data processing algorithm

The Version 3.0 software program has been used for retrieving gas and aerosol parameters as a research product from ILAS-II signals. The software has been developed on the basis of studies conducted by the NIES scientists.

A distinguishing feature of the Version 3.0 retrieval algorithm is simultaneous gas and aerosol retrievals using aerosol physical modeling. In brief, a basic concept for retrieval is similar as the previous Versions but volume particle size distributions for several aerosol/PSCs components are added as unknowns to be retrieved simultaneously with gas mixing ratios. A comprehensive description of the methodology is described elsewhere [*Oshchepkov et al*., 2005; *Sasano et al*., 2004a; *Oshchepkov et al*., 2006].

According to the methodology, the total aerosol extinction coefficient (AEC) spectrum in theoretical pseudo transmittance [*Yokota et al*., 2002] is presented through a linear combination of the AEC spectra assigned to the following aerosol and PSC's components: H_2SO_4/H_2O and HNO_3/H_2O binary liquid solutions, two modifications of Nitric Acid Trihydrate (α-NAT and β-NAT), Nitric Acid Dihydrate (NAD), and water ice. Applying the aerosol physical modeling, the AEC for each of these five components is presented through integral of the single particle cross section over the number particle size distributions. For further numerical solution, each integral is reduced by a trapezoidal approximation [*Twomey*, 1977] to a set of linear algebraic equation. The unknowns of the equations constitute volume particle size distributions at given size nodes (five for this case) equidistantly spaced on a logarithmic scale and the columns of the coefficients are reference aerosol spectra defined at the corresponding size nodes as a product of the linear quadrature [*Twomey*, 1977].

The reference aerosol spectra are calculated according to the Mie theory using spectral dependences of the refractive indices from [*Niedziela et al*., 1999] (H_2SO_4/H_2O at 75% of weight of sulfuric acid), [*Norman et al.*, 1999] (HNO_3/H_2O at 40% of weight of nitric acid), [*Richwine et al.* 1995,] (α-NAT), [*Toon et al.*, 1994] (β-NAT), [*Niedziela et al*., 1998] (NAD), and [*Clapp et al*., 1995] (water ice). These refractive indices excluding Toon's data for β-NAT concern laboratory aerosol spectrometry measurements.

The component selection at each tangent height from 13km to 35km km is not restricted by thermodynamic considerations.

Spectral inversion is based on the constrained weighed least square minimization between actual and theoretical ILAS-II IR signals to retrieve simultaneously gas mixing ratios and particle size distributions for the selected aerosol components. In the minimization procedure, positive constraints are imposed on both gas and aerosol parameters by a using penalty function.

A set of trace gases to retrieve is the same as that retrieved in Version 2.0 data processing algorithm. Primary retrieved particle volume size distributions are used to produce vertical profiles of volume density for the STS, NAT, NAD, water ice, and total aerosol/PSC. The total AEC for all 44 channels is also available at each tangent height. All retrieved gas and aerosol parameters are supported by accurate error bar estimations using covariance matrix of the retrievals and the transmittance residuals [*Yokota et al*., 2002; *Oshchepkov et al*., 2005].

Due to the occultation geometry, the retrievals concern aerosol characteristics averaged over pathlength for single atmospheric layer. In particular, because a length of PSCs does not necessarily cover the entire pathlength, the retrieved aerosol volume densities do not exceed those for the clouds. Another limitation concerns particle sizing. Namely, for particles smaller than 0.4 μm and for particles larger than 5 μm the available quantities to retrieve are only total particle volume and total particle cross section densities, respectively.

References

Clapp, M. L., R. E. Miller, and D. R. Worsnop (1995), Frequency-dependent optical constants of water ice obtained directly from aerosol extinction spectra, *J. Phys. Chem., 99*, 6317-6326.

Niedziela, R. F., R. E. Miller, and D. R. Worsnop (1998), Temperature- and frequency-dependent optical constants for nitric acid dihydrate from aerosol spectroscopy, *J. Phys. Chem., 102*, 6477-6484.

Niedziela R. F., M.L. Norman, C. L. DeForest, R. E., Miller, and D. R. Worsnop (1999), A temperature- and composition-dependent study of H_2SO_4 aerosol optical constants using Fourier transform and tunable diode laser infrared spectroscopy, *J. Phys. Chem*., *103*, 8030-8040.

Norman M. L., J. Qian, R. E. Miller, and D. R. Worsnop (1999), Infrared complex refractive indices of supercooled liquid HNO_3/H_2O aerosols, *J. Geophys. Res*., *104*(30), 30,571-30,584.

Oshchepkov, S., Y. Sasano, T. Yokota, N. Uemura, H. Matsuda, Y. Itou, and H. Nakajima (2005), Simultaneous stratospheric gas and aerosol retrievals from broadband infrared occultation measurements, *Appl. Opt.*, 44, 4775–4784.

Oshchepkov, S., Y. Sasano, T. Yokota, H. Nakajima, N. Uemura, N. Saitoh, T. Sugita, and H. Matsuda (2006), ILAS data processing for stratospheric gas and aerosol retrievals with aerosol physical modeling: Methodology and validation of gas retrievals, *J. Geophys. Res.*, 111, D02307, doi:10.1029/2005JD006543.

Richwine, L. J., M. L. Clapp, R. E. Miller, and D. R. Worsnop (1995), Complex refractive indices in the infrared of nitric acid trihydrate aerosols, *Geophys. Res. Lett.*, *22*, 2625-2628.

Sasano, Y., S. Oshchepkov, T. Yokota, and H. Nakajima (2005), Characterization of stratospheric liquid ternary solution from broadband infrared extinction measurements, *J. Geophys. Res.*, 110, D18212, doi:10.1029/2004JD005709.

Toon, O. B., M. A. Tolbert, B. J. Koehler, A. M., Middlebrook, and J. Jordan (1994), Infrared optical constants of H_2O ice, amorphous nitric acid solution, and nitric acid hydrates, *J. Geoph. Res.*, *99*(D12), 25,631-25,654.

Twomey, S., *Introduction to the mathematics of inversion in remote sensing and indirect measurements*, 243 pp., Elsevier Sci., New York, 1977.

Yokota, T., H. Nakajima, T. Sugita, H. Tsubaki, Y. Itou, M. Kaji, M. Suzuki, H. Kanzawa, J. H. Park, and Y. Sasano (2002), Improved Limb Atmospheric Spectrometer (ILAS) data retrieval algorithm for Version 5.20 gas profile products, *J. Geophys. Res.*, *107*(D24), 8216, doi:10.1029/2001JD000628.

APPENDIX2

File Descriptions for ILAS-II V3.0x Aerosol Data

1. Aerosol volume density file

Data format

(1) Record
One record for each altitude at 1 km interval

(2) Content of one record
Altitude, volume densities and their errors corresponding to the following nine aerosol types:
"Saw" = Sulfuric acid volume density
"Naw" = Nitric acid volume density
"alph_NAT" = alpha-NAT volume density
"beta_NAT" = beta-NAT volume density
"NAD" = NAD volume density
"ICE" = Ice volume density
"LTS" = Liquid Ternary Solution volume density (sum of Saw and Naw)
"T_NAT" = Total NAT volume density (sum of alph-NAT and beta-NAT)
"T_Aerosol" = Total volume density (sum of NAD, ICE, LTS, and T_NAT)

(3) Special Notes
Error values are marked with a minus sign, when iterations reach one hundred.

File example
1) 7 1001
2) Aerosol Volume Density (micron**3/cm**3)
3) Observation time (UTC,TH=20km point): 2003 07 15 23:47:01.799
4) Occultation event number: 20030715151
5) Latitude (deg, positive=north): -67.46
6) Longitude (deg, positive=east): 164.86
7) Start time of measurement: 2003 07 15 23:46:29.739
8) TH(km) Saw error Naw error alph_NAT error beta_NAT error NAD error ICE error LTS error T_NAT error T_Aerosol error
9) 23
10) SunSet
11) 13.00 4.729E-02 4.694E-02 -4.697E-04 1.017E-02 9.510E-02 4.188E-02 -3.206E-04 1.017E-02 9.462E-02 9.663E-02 -1.483E-04 1.017E-02 4.682E-02 4.756E-02 9.478E-02 4.209E-02 2.361E-01 6.493E-02

2. Aerosol extiction coefficient file

Data format

(1) Record
One record for each altitude at 1 km interval

(2) Content of one record
Altitude, aerosol extinction coefficients and their errors corresponding to the wavelength for the following 44 IR spectral elements and a visible spectral element.

(3) Special Notes
Error values are marked with a minus sign, when iterations reach one hundred.

File example
1) 7 1001
2) Aerosol Extinction Coefficient (/km)
3) Observation time (UTC,TH=20km point): 2003 07 15 23:47:01.799
4) Occultation event number: 20030715151
5) Latitude (deg,positive=north): -67.46
6) Longitude (deg,positive=east):   164.86
7) Start time of measurement: 2003 07 15 23:46:29.739

8) TH(km)	IR00	error	IR01	error	IR02	error	
IR03	error	IR04	error	IR05	error	IR06	error
IR07	error	IR08	error	IR09	error	IR10	error
IR11	error	IR12	error	IR13	error	IR14	error
IR15	error	IR16	error	IR17	error	IR18	error
IR19	error	IR20	error	IR21	error	IR22	error
IR23	error	IR24	error	IR25	error	IR26	error
IR27	error	IR28	error	IR29	error	IR30	error
IR31	error	IR32	error	IR33	error	IR34	error
IR35	error	IR36	error	IR37	error	IR38	error
IR39	error	IR40	error	IR41	error	IR42	error
IR43	error	Vis	error				

9) 23
10) SunSet

11) 13.00	9.861E-05	2.038E-05	9.072E-05	1.876E-05	9.272E-05	2.013E-05
9.975E-05	2.423E-05	1.319E-04	4.412E-05	2.569E-04	1.309E-04	2.167E-04
5.784E-05	2.222E-04	4.871E-05	2.091E-04	4.494E-05	1.827E-04	3.955E-05
1.782E-04	5.617E-05	2.054E-04	9.833E-05	2.090E-04	1.045E-04	1.804E-04
7.812E-05	1.418E-04	4.313E-05	1.280E-04	3.732E-05	1.283E-04	4.027E-05
1.307E-04	4.252E-05	1.352E-04	4.361E-05	1.360E-04	4.327E-05	1.307E-04
4.023E-05	1.183E-04	3.608E-05	1.054E-04	3.305E-05	9.516E-05	3.218E-05
9.327E-05	3.514E-05	9.559E-05	4.112E-05	1.022E-04	3.735E-05	1.226E-04
4.252E-05	7.997E-05	2.937E-05	6.746E-05	2.534E-05	6.083E-05	2.122E-05
5.796E-05	1.869E-05	5.453E-05	1.661E-05	5.188E-05	1.506E-05	4.940E-05
1.468E-05	4.871E-05	1.601E-05	5.199E-05	1.938E-05	5.626E-05	2.219E-05

5.627E-05	2.146E-05	5.411E-05	1.870E-05	5.378E-05	1.559E-05	5.137E-05
1.317E-05	5.113E-05	1.201E-05	5.231E-05	1.172E-05	9.294E-04	3.198E-04