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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term observation of mass-independent oxygen isotope anomaly instratospheric CO_2" *by* S. Kawagucci et al.

S. Kawagucci et al.

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We are grateful for fruitful referee comments on our discussion paper from Prof. K. Boering. Her helpful comments make our manuscript improved. We have carefully studied the comments and revised the manuscript accordingly.

(1) Referee comment (p. S8112 to p. S8114, line 19) As referee pointed out, the interpretation for a linear correlation between D17O and N2O mixing ratio as [controlled by transport alone] in discussion paper is mistaken. We delete the interpretation. In revised version manuscript, we interpreted the correlation as [the correlation between D17O and N2O would result from comparable time scales of the reaction (R1-R2) and N2O destruction reactions, which are dominated (˜90%) by photolysis and partly (˜10%) contributed by reactions to O(1D).] as discussed by Boering et al. (2004).

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(2) Referee comment (p. S8114, line 20 to p. S8115, line 5) We vastly change the discussion about a slope on d18O-d17O plot due to a remove of the interpretation by dataset division based on N2O mixing ratio. Details of new discussion will be presented in revised manuscript. Here, we describe brief conclusion of the discussion: [Observed d18O and d17O over Sanriku and Kiruna in this study showed a good linear correlation (r2$>$0.95) yielding a slope of 1.64+/-0.05 (1-sigma) that was quite consistent with one observed in middle-latitude lower/middle stratosphere (Lammerzahl et al., 2002) although clearly different from one observed in upper stratosphere/lower mesosphere (Thiemens et al., 1995b).]. [the difference of the slopes between lower/middle stratospheric CO2 (Lammerzahl et al., 2002; this study) and upper stratospheric/lower mesospheric CO2 (Thiemens et al., 1995b) does not mean a gradual slope change with altitude change, but just slope difference between each study.]. [the existence of gradual slope decline with increasing altitude is still open question. Observation of triple oxygen isotopes in CO2 continuously through middle atmosphere and/or intensively in upper stratosphere/lower mesosphere will help to clarify the slope changing.].

(3) Referee comment (p. S8115, line 6 to p. S8116, line 3) As pointed out by referee, the term [year-to-year variation] was ambiguous. Instead of [year-to-year variation] in discussion paper, we use [long-term trend] and [interannual variation] in revised paper. We classify the symbols in all Figures with respect to sampling year in order to clarify the interannual variation on sight. By these improvements in discussion and presentation, interannual variations are revealed in both the vertical profile of D17O and the D17O-N2O plot although no long-term trends have been detected. The interannual variations can be explained by a variation of chemical time scales between N2O photolysis and CO2 and O(1D) reaction or that of relative time scales between chemical and physical processes.

(4) Referee comment (p. S8116, line 4 to p. S8116, line 13) We add the model calculation results into both the D17O-N2O and d18O-d17O plots for comparison (Figures 2a and 3). The comparisons between the observation and the model show an under-

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estimation of D17O maximum in the model that may be brought from overestimation of the O2 photolysis contribution to CO2 and O(1D) reaction in the mesosphere in the model.

(5) Referee comment (p. S8116, line 14 to p. S8116, line 26) According to referee comments, we provide some new information for clarification of details of observation, analysis, and discussion. First, N2O mixing ratios are shown in Table 1. Second, several constants to calculate d17O by Kawagucci et al. (2005) method are presented in Section 2 as follows: lambda = 0.516 (Santrock et al., 1985); K = 0.0099235 (Santrock et al., 1985); 13RVPDB = 0.0112372 (Craig, 1957); 18RVSMOW = 0.0020052 (Baertschi, 1976). Third, D17O and d18O isotope fluxes from stratosphere to troposphere are calculated and presented respectively as +48 permil GtC/yr and +38 permil GtC/yr with ˜30% uncertainties by similar way presented in Boering et al. (2004).

(6) Referee comment (p. S8117, line 3 to 12) Most specific question at a start of this study was the slope difference on d18O-d17O plot between previous papers (Thiemens et al. (1995); Lammerzahl et al. (2002)). In addition, it is also the remained question that other previous studies tried to interpret the slope difference as the gradual slope change (Kawagucci et al. (2005); Liang et al. (2007)) while the gradual change has not been observed. We discussed and answered the questions as (2) in this reply. In addition to the discussion for the slope, we also estimated CO2 oxygen isotope fluxes as (5) in this reply.

(7) Referee comment (p. S8117, line 13 to 18) The expression of [correlation fade away…] is deleted.

(8) Referee comment (p. S8117, line 20 to 21) [recently] is removed.

(9) Referee comment (p. S8117, line 22 to 26) The expression for oxygen isotope compositions of tropospheric CO2 is rewritten to [Tropospheric CO2 has an almost constant 18O value (˜ +41permil) with little 17O anomaly due to rapid oxygen isotope exchange between troposphere CO2 and surface water, such as leaf water and

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seawater (Thiemens et al., 1991; Ciais et al., 1997; Hoag et al., 2005).].

(10) Referee comment (p. S8118, line 1 to 13) Further details of sampling system, sample storage over a decade, and analytical procedure are additionally described.

(11) Referee comment (p. S8118, line 14 to 18) The expression for sampling location compared to tropopause height is rewritten to [We obtained 53 whole stratospheric air samples with 5 tropospheric air samples during seven launches over Sanriku and Kiruna (Table 1).]

(12) Referee comment (p. S8118, line 19 to 23) We used the value of [1.4 ppmv/yr] proposed from stratospheric CO2 mixing ratio observation by Aoki et al. (2003) that used same samples in this study.

(13) Referee comment (p. S8118, line 24 to 25) [correlation to] and [high-latitudinal] are rewritten to [correlation with] and [high-latitude], respectively.

(14) Referee comment (p. S8118, line 26 to p. S8119, line 3) [lower stratosphere above 25 km] in the discussion paper is inept. We define lower-middle-upper stratosphere as [tropopause˜25km], [25km˜36km (our observation range)], and [36km˜stratopause (out of our observation range)], while the definition is unspecified in the literature.

(15) Referee comment (p. S8119, line 4 to p. S8119, line 7) Expressions of [almost positive] and [almost negative] are deleted.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 15723, 2007.

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Interactive Comment