

We thank both referees for their very constructive comments and suggestions. We have revised our manuscript according to their comments and suggestions.

Anonymous referee #1

Review of Paper ACPD 14 (2014), 1823-1859

Influence of airmass downward transport on the variability of surface ozone at Xianggelila Regional Atmosphere Background Station, Southwest China,

by J. Ma, W. L. Lin, X. D. Zheng, X. B. Xu, Z. Li, and L. L. Yang

General Remarks

The manuscript describes results of atmospheric measurements obtained at a high-altitude station at the edge of the Tibetan Plateau. Observations in that region are highly relevant, but still sparse, which emphasizes the value of this manuscript. However, there are a few deficiencies the authors should consider addressing before publication in ACP. First of all, the investigations cited refer to work related to mid-latitudes. This is highly welcome, but downward transport in the subtropics is more controlled by the subtropical jet stream. Therefore, the relevant literature must be included. Also, the role of the monsoon in vertical exchange and the position of the jet stream require more details including literature.

Details

(1) Abstract: line 8: I think you mean "O₃ downward transport events". This is a deficiency throughout the paper: "transport event" is never clearly explained, but obviously preferentially assigned to STT events. Please, rephrase this expression clearer in the entire paper, also varying the wording (stratospheric intrusion, STT event, acronyms defined at the beginning, others).

Response: You are right. We use this word more carefully in the revised paper. The Y-indicator is used to evaluate the occurrence of O₃ downward transport. Also for being clearer, we have changed the title of the paper to "Influence of airmass downward transport on the variability of surface ozone at Xianggelila Regional Atmosphere Background Station, Southwest China".

(2) Abstract, lines 11-12: Is there a way to specify how significant this contribution is? Even a rough estimate ("of the order of") would be better than the phrase used.

Response: The sentence in lines 14-16 specifies the contribution of deep downward transport events in winter. We have moved this sentence ahead.

(3) Abstract: The comparison with the jet-stream seasonality is missing.

Response: As suggested, "The seasonal variation of O₃ downward transport, as suggested by the Y indicator at Xianggelila, is consistent with the seasonality of stratosphere-to-troposphere transport and the subtropical jet stream over the Tibetan plateau" is added at the end of the abstract.

(4) P. 1825, line 3: These stations are not necessarily remote, better remove "remote".

Response: accepted.

(5) P. 1825, following line 18: Here, an introduction to the importance of STT along the subtropical jet stream (STJ), its seasonality and the influence of the monsoon is expected. This

cannot be omitted since the situation in the mid-latitudes cannot be fully transferred to Tibet. The importance of the STJ had been underestimated until the late 1990s. This is a time when case studies started (e.g., Cammas et al.). Sprenger et al., *J. Geophys. Res.*, 108 (2003), 8518, doi: 10.1029/2002JD002587, found that the downward transfer along the STJ could be even more important than that STT in the mid-latitudes. This emphasizes the role of the high-lying stations in that area. There are indications of long-range transport of high-ozone air masses that emerged from shallow STT along the STJ (Langford et al., *GRL* 25 (1998), 2667-2670; Langford et al., *GRL* 26 (1999), 2449-2452; Trickl et al., *ACP* 11 (2011), 9343-9366). Koch et al., *Int. J. Climatol.*, 26 (2006), 283-301, published some model-based climatology. Unfortunately, I am not familiar with the literature on the influence of the monsoon on the STJ, but something on this topic should be added.

Response: To address the importance of the STJ, we have added the following text at the end of the first paragraph in the introduction: In the mid-latitudes, the subtropical jet (STJ) stream can have significant effect on the vertical ozone distribution and the STJ varies from a wintertime maximum to a summertime minimum (Bukin et al., 2011; Koch et al., 2006). Sprenger et al. (2003) found that the downward transfer along the STJ could be even more important than the stratosphere-to-troposphere transport (STT) in the mid-latitudes and there are indications of long-range transport of high-ozone air masses that emerged from shallow STT along the STJ (Langford et al., 1998; Langford, 1999; Koch et al., 2006; Trickl et al., 2011). Near the STJ, the occurrence frequency of double tropopauses shows a strong seasonal variation over North Hemisphere midlatitudes, with 50–70% occurrence in profiles during winter, and a small fraction (~10%) over most of the hemisphere during summer (Randel et al., 2007), and the multiple tropopause occurrence over the Tibetan Plateau can be as high as 80% during certain winters (Chen et al., 2011). In addition, the Asian and North American monsoons may have distinct effects on the upper troposphere and lower stratosphere (Gettelman et al., 2004).

Bukin, O.A., An, N.S., Pavlov, A.N., Stolyarchuk, S. Y., Shmirko, K.A.: Effect that Jet Streams Have on the Vertical Ozone Distribution and Characteristics of Tropopause Inversion Layer in the Far East Region. *Izvestiya, Atmospheric and Oceanic Physics*, 2011, Vol. 47, No. 5, pp. 610–618.

Chen, X. L., Ma, Y. M., Kelder, H., Su, Z., and K. Yang.: On the behaviour of the tropopause folding events over the Tibetan Plateau, *Atmos. Chem. Phys.*, 11, 5113–5122, doi:10.5194/acp-11-5113-2011, 2011.

Gettelman, A., Kinnison, D. E., Dunkerton, T. J., and Brasseur, G. P.: Impact of monsoon circulations on the upper troposphere and lower stratosphere, *J. Geophys. Res.*, 109, D22101, doi:10.1029/2004JD004878, 2004.

Koch, P., Wernli, H., and Davies, H. C.: An Event-based Jet-stream Climatology and Typology, *Int. J. Climatol.*, 26, 283–301, 2006.

Langford, A. O.: Stratosphere-troposphere exchange at the subtropical jet, contribution to the tropospheric ozone budget at midlatitudes, *Geophys. Res. Lett.*, 26, 2449–2452, 1999.

Langford, A. O., OLeary, T. J., Masters, C. D., Aikin, K. C., and Proffitt, M. H.: Modulation of middle and upper tropospheric ozone at Northern midlatitudes by the El Niño/Southern Oscillation, *Geophys. Res. Lett.*, 25, 2667–2670, 1998.

Randel, W. J., D. J. Seidel, and L. L. Pan (2007), Observational characteristics of double tropopauses, *J. Geophys. Res.*, 112, D07309, doi:10.1029/2006JD007904.

Trickl, T., Bärtsch-Ritter, N., Eisele, H., Furger, M., Mücke, R., Sprenger, M., and Stohl, A.: High-ozone layers in the middle and upper troposphere above Central Europe: potential import from the stratosphere along the subtropical jet stream, *Atmos. Chem. Phys.*, 11, 9343-9366, doi:10.5194/acp-11-9343-2011, 2011.

(6) P. 1825, line 19: Better "The Tibetan Plateau and the surrounding mountain ranges".

Response: accepted.

(7) P. 1826, line 4: "and, on the south rim, the Nepal Climate Observatory in the Himalaya range (Cristofanelli....)"

Response: accepted.

(8) Sec. 2.2: Give uncertainties of the instruments.

Response: We have added the uncertainties.

(9) P. 1828, line 9: Add citation on HYSPLIT (see web site) and web site.

Response: accepted.

(10) P. 1828, line 21: "the case study" was (if I did not miss it) not yet introduced. If this is the case, please, write "in a case study described in Sect. 3.3".

Response: accepted.

(11) P. 1829, lines 1 and 2: Lower stratospheric CO is not immediately low above the tropopause. Above the transition zone values down to 20 ppb have been reported; at mid-latitude summit stations mostly values of 80-100 ppb arrive in STT air. If this is different in Tibet this could be discussed somewhere in the paper (see (23) further below).

Response: Here, we refer a general inverse profile of CO when comparing with the surface level of CO. CO source is mainly in the lower troposphere.

(12) P. 1830, line 10: Better: "consistent with observations at background sites elsewhere in the Northern Hemisphere"

Response: accepted.

(13) P. 1830, line 11: Specify range of wintertime values for CO.

Response: We have added the range in the revised paper.

(14) P. 1830, line 18: ", where photochemically produced O₃ can accumulate starting in the late morning."

Response: accepted.

(15) P. 1830, lines 22-23: Better: "and then, until the beginning of the night, O₃ decreases with decreasing wind speed." Is there any idea about this behaviour? Does this mean that turbulent downward mixing from a reservoir diminishes and deposition becomes more important? Is the station located in flat terrain or elevated?

Response: The station is located on the top of a hill. We have added ".....and then, until the beginning of the night, O₃ decreases with decreasing wind speed when the turbulent downward mixing from a reservoir diminishes and deposition becomes more important."

(16) P. 1831, line 1: An introductory sentence is missing, e.g., "The amplitude of the diurnal variation of O₃ varies as a function of the season." Then, I would continue "The maximal amplitude was found in spring, the minimal one in winter." This spring maximum is a remarkable result that could be emphasized on. Does this indicate an importance of the photochemical origin of the spring peak (see Monks)? What about CO?

Response: We have included the introductory sentence and added "In spring, the average daytime level of CO is the highest among four seasons. A positive correlation between O₃ and CO (slope: 0.154, P<0.0001) during the daytime (10:00~18:00) in spring can be derived using the reduced-major-axis regression technique. Such positive O₃-CO correlation suggests photochemical production of O₃ from anthropogenic sources. This indicates the importance of photochemical origin of the spring peak."

(17) P. 1831, line 14: Describe how the clusters are defined (source region? Please, give some details!).

Response: Trajectories are grouped according to their spatial similarity. The number of clusters is judged in a plot of percent change of the total cluster spatial variance vs. the number of clusters. The iterative step just before the large increase in the change of TSV gives the final number of clusters. Details can be found in the user guide of Hysplit software.

(18) P. 1831, line 23: The relative importance of types 1-4 is missing (see line 17); this is an important issue because of the discussion on p. 1832. I suggest adding a column with the relative contributions to Table 2 in addition to mentioning them in the text (or refer to the insert in Fig 4; is this what "ratios" mean in line 15? "Fractions" or "relative contributions" would be easier to understand.).

Response: The "ratios" means "fractions". As the fractions are showed in the insert in Fig. 4 and mentioned in the text, no column is added to Table 2. The sentence in lines 14-15 is reworded as "The mean trajectory for each cluster, their fractions (the number of trajectories in each cluster to the total number of the trajectories), and their patterns are shown in Fig. 4."

(19) P. 1832, lines 11-12: The situation in Central Europe is not relevant here. If you want to point out the similarity you could do this in a separate sentence. Sprenger et al. show the seasonality of STT (which is important to mention) not of the STJ. However, it is reasonable to assume that the band of STT events they show is related to the STJ. The seasonality of the STJ is described by Koch et al. as far as I remember (this should also be mentioned). The STJ is present during a major part of the year. Sprenger et al. distinguish between shallow, medium and deep folds. The definitions are unclear. In Tibet medium and deep folds could be the species of interest. The images for the medium folds (Figs. 3c and d by Sprenger et al.), indeed, indicate a seasonal pattern for the eastern rim of the highlands, but there is not much as to deep folds. Thus, observational material is very important indeed.

Response: Yes, more text is added and additional references are cited: "This is consistent with the seasonality of stratosphere-to-troposphere transport (Sprenger and Wernli, 2003; Sprenger et al., 2003) and the subtropical jet events (Koch et al., 2006) in the Northern Hemisphere. The results from Sprenger et al. (2003) demonstrate that during winter, the frequency of shallow tropopause folds is highest above the Tibetan Plateau (see Fig. 3 in their paper). Škerlak et al. (2014) concluded that, as one of the clear hotspots of deep STT fluxes into the continental PBL, there are also intense deep STT fluxes over the Tibetan Plateau during the whole year, with a peak in winter. On the basis of the intensive radiosonde observations, Chen et al. (2011) concluded that the multiple tropopause, which is associated with tropopause folds near the

subtropical westerly jet, occurs in winter with a high frequency over the Tibetan Plateau, and as a result, the intrusion of air masses from the stratosphere may contribute to a higher upper tropospheric O₃ concentration in winter than in summer above the plateau.”

Chen, X. L., Ma, Y. M., Kelder, H., Su, Z., and K. Yang.: On the behaviour of the tropopause folding events over the Tibetan Plateau. *Atmos. Chem. Phys.*, 11, 5113–5122, doi:10.5194/acp-11-5113-2011, 2011.

Škerlak, B., Sprenger, M., and Wernli, H.: A global climatology of stratosphere–troposphere exchange using the ERA-Interim data set from 1979 to 2011, *Atmos. Chem. Phys.*, 14, 913–937, doi:10.5194/acp-14-913-2014, 2014.

(20) P. 1832, lines 15-17: do not mix heights and pressure levels. 200 to 400 hPa does not correspond to 1000 to 3000 m, otherwise you could put the altitude range in brackets. It is interesting to see a wintertime UTLS maximum in the pressure panel, but not in the height panel. Please, discuss this in the text!

Response: We have put the altitude range in brackets. We did notice the wintertime UTLS maximum in the pressure panel, but not in the height panel. Maybe the pressure levels are more suitable than the heights because the latter are terrain-following and described in m above ground level. To discuss this, we have inserted the following text in P. 1832, line 22: “It is interesting to see that there is a winter maximum around the UTLS region in the pressure panel of Fig. 5, but no maximum in the height panel. The actual reason for this is clear. The pressure levels are more comparable than the heights because the latter are terrain-following and given in m above ground level.”

(21) P. 1833, line 19: As said "transport event" is not sufficiently specific (see title of Sec. 3.4 and at many places)

Response: Changed to “A case of strong O₃ downward transport”.

(22) P. 1833, line 20: Replace "explain" by "demonstrate".

Response: accepted.

(23) P. 1834, line 3: What "higher layer"? Please, explain.

Response: We calculated the airflow in the different height of the endpoints. When the endpoint height is above 1200m, the phenomenon is evident. We clear it in the text.

(24) P. 1833: The exceptionally low value of CO (I see 20 ppb not 50 ppb) asks for a few more words; 20 ppb is a value corresponding to the full stratosphere outside the UTLS transition layer (there are numerous papers on this, I suggest, e.g., Hegglin et al., *J. Geophys. Res.* 114 (2009), D00B11, doi: 10.1029/2008JD009984; other names to search for are Zahn, Pan, Hoor, Vogel, ...). I am not aware of any other example of this kind (see (Trickl et al., 2010), for a few examples). Is there evidence from more examples in your station record?

Response: Our CO analyzer can give values as low as several ppb. However, the lower detection limit of the analyzer is 25 ppb so that the uncertainty of the lower measurements (say, < 50 ppb) is much larger. To be careful about this technical limitation, we have decided not to over-interpret the low CO values you mentioned.

(25) P. 1835, line 21: "average maximal height" looks contradictory. What do you

mean?

Response: We use the maximal height of a trajectory (the latitude, longitude position and height for trajectory are described every one hour) to obtain the mean value. It is changed to "averaged maximal height"

(26) P. 1838, lines 9-10: Why should be observations in China be consistent with findings for Europe? Better "and resembles that".

Response: accepted.

Figures:

(1) Fig. 1: Cite Google Maps?

Response: The sources are cited.

(2) Fig. 6, line 5: "actually negative, but"; axis title "Y indicator" should be slightly separated from "Correlation coefficient".

Response: Accepted.

(3) Figs. 8 and 9: The panels and the text is too small.

Response: We have modified the panels and re-sized the text.

(4) Fig. 10: I do not see any red feature.

Response: We have made the red pentacles brighter.

(5) Fig. 12: "above specific trajectory pressure" (levels?) is absolutely unclear.

Response: Changed to "above specific trajectory pressure levels".

Style

Response: We sincerely thank the reviewer's detailed correction. We have carefully checked the style and made corrections according to the suggestions.

(1) P. 1824, line 26: Change to "from the stratosphere". Articles are missing at many places in the paper. Please, re-examine carefully. This review is not necessarily complete.

(2) P. 1825, line three: Same!

(3) P. 1825, line 15: Remove semicolon behind "2012".

(4) P. 1825, line 15: Replace "often" by "are often".

(5) P. 1825: line 21: Better: "have great impact on air circulation, climate change, on scales."

(6) P. 1825, line 23: "from the upper troposphere and the lower stratosphere"

(7) P. 1825, line 24: "natural input of"

(8) P. 1826, line 15: "which is located"

(9) P. 1826, line 20: "impact"

(10) P. 1827, line 7: "of the Yunan"

(11) P. 1827, line 11: Better: "for monitoring".

- (12) P. 1828, line 16: "The height of the endpoint"
- (13) P. 1829, lines 2-3: Replace "rich" and "poor" by "high" and "low".
- (14) P. 1830, lines 20-21: "the O₃ mixing ratios increase sharply with increasing wind speed."
- (15) P. 1831, line 2: "In the monsoon season"
- (16) P. 1831, line 14: "grouped into 7 clusters"
- (17) P. 1831, line 21: "abundant rain". I think "abundant precipitation" is more adequate in winter because of there is the possibility of snowing.
- (18) P. 1831, line 26: "airmasses from the free troposphere to the surface"
- (19) P. 1832, line 1: "level"
- (20) P. 1832, line 5: "in fall, but the Y indicator"
- (21) P. 1832, line 18: "large Y indicators" or "the occurrence of a large Y indicator"
- (22) P. 1832, lines 16 and 23: Do you mean "possibility" or "probability"?
- (23) P. 1832, line 22: "a tiny peak in the kernel probability density at pressures around 430 hPa"
- (24) P. 1833, line 10: "from the upper"
- (25) P. 1833, line 16: "rain"
- (26) P. 1833, line 26: "detection limit of the CO analyzer"
- (27) P. 1833, line 27: "7 day" or "7-day"; "trajectories initiated at 00:00 UTC"
- (28) P. 1834, line 4: "in the higher layer"
- (29) P. 1834, line 7: "origination" does not exist. You could use "origin" or "source region".
- (30) P. 1834, line 11: Replace ", which" by "that".
- (31) P. 1834, line 13: Remove "there was".
- (32) P. 1835, line 4: "3.3, the Y indicator"
- (33) P. 1835; line 7: "there may be a high probability of an intrusion event."
- (34) P. 1835, line 13: "To analyze further" (no split infinitive allowed in English grammar!)
- (35) P. 1835, line 24: "The pressures covered by the trajectories in winter"
- (36) P. 1836, line 12: "Because there is still monsoon influence in fall"
- (37) P. 1837, lines 1 and 3: "the Y indicator"
- (38) P. 1837, line 3: "there are still open questions such as"
- (39) P. 1838, line 7: "pressure level"

Response to Anonymous referee #2

General comments

The paper discusses an analysis of stratospheric intrusion of ozone-rich air masses detected in a two-year period at the Xianggelila background station located at high altitude in south-western China. A normalized indicator was developed to evacuate the occurrence of cases of downward transport of ozone. This is an interesting topic suitable for ACP. I have some suggestions that could be taken into account in the revision detailed in my specific comments.

Specific comments

In abstract (Line 8-10). “it is show that most frequent transport events……”. It is not very clear if authors intend that in winter the events are more frequent or that these events are more intense. Further, it would be possible to include a quantitative (even if approximate) estimation of the mentioned contribution.

Response: According to the results of Table 3, the occurrence of cases of the downward transport of O₃-rich air is the most frequent in winter, but the most intense event is found in June as seen in section 3.3. It seems not easy to select a threshold value of Y to get the quantitative contribution from all O₃ downward transport events. On the basis of Y larger than 4, 39% of cases of downward transport of O₃-rich air occurred in winter. An increase of O₃ caused by deep transport event is estimated as 21.0% (+9.6 ppb) in winter, by subtracting the winter average ozone level (45.8 ppb) from the average O₃ mixing ratio (55.4 ppb) in the period with both trajectories pressure level lower than 400 hPa and Y over 8. The sentence in lines 14-16 “A 9.6 ppb increase (21.0%) of surface ozone is estimated based on the impacts of deep downward transport events in winter” conveys this information. Therefore, we have moved this sentence ahead.

The Y indicator developed here is quite similar to the SI indicator developed in Cristofanelli et al. (2009) even if this last is based on Be the general idea and structure of the two indicators are similar. It would be interesting if author try including a discussion to compare the performances of the two indicators.

Response: Yes. The general idea and structure of the two indicators are similar. The seasonality of the occurrence of cases of downward transport of O₃-rich air is similar to the results of Cristofanelli et al. (2009). Both show a seasonal cycle with a winter maximum and a summer minimum. We have included in section 3.4 in the revised paper “The seasonal cycle of our Y indicator (see Table 3 and Fig. 6) resembles that of the SI frequency at Mt. Cimone obtained by Cristofanelli et al. (2009) using a stratospheric intrusion index. Both indicators reveal that the downward transport of upper air is strongest in winter and weakest in summer.”

In several parts of the paper it is spoken generically of “ozone transport”. I believe that it is better to specify when it is needed that it is downward transport otherwise it could be confused with advection.

Response: We agree with you. Downward transport is specified.

In section 2.2 there is a detailed description of the instruments and of the calibration procedures used. It is missing a description of the measurement uncertainties. Please add it.

Response: Yes, we have added the uncertainties of the instruments.

Section 3.3. Please change “ detect limit” with “detection limit”.

Response: Accepted.

In caption of Fig. 11 Pleae change “bigger” with “larger”.

Response: Accepted.