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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Airborne observations and modeling of springtime stratosphere-to-troposphere transport over California" *by* E. L. Yates et al.

E. L. Yates et al.

emma.l.yates@nasa.gov

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Final author comments: The authors are most grateful to the reviewers for their thorough and thoughtful commentaries, which have improved the manuscript. We are pleased to respond as follows:

Reply to Anonymous Referee #1 This study presents in situ measurements and modeling analysis of a stratospheric cut-off and a tropopause fold over California, USA. A research aircraft sampled vertical profiles by descending in a spiral motion over the San Joaquin Valley (SJV) and an offshore location. The airborne instruments mea-

sured water vapour (H2O), ozone (O3) and carbon dioxide (CO2) and additionally, radiosonde data from Oakland is used to identify stable layers. Stratospheric air is identified by high O3, low CO2 and low H2O. The in situ ozone measurements are compared to ozone modeled by the Real-time Air Quality Modeling System (RAQMS) and the RAQMS combined with a Reverse Domain Filling (RDF) technique. The performance of RAQMS as well as RAQMS+RDF in terms of reproducing the observed ozone mixing ratios is compared. Finally, the impact on surface ozone levels and policy-making is discussed.

The most valuable aspect of this study is the data obtained from in situ measurements in complex meteorological situations. The measurements were performed at the right time in (nearly) the right place to clearly identify both the stratospheric cut-off and the tropopause fold. What remains unclear to me is the main goal of this study: is it the identification of the measured air as stratospheric, to show the relevance of CO2 for identifying stratospheric air, the analysis of the meteorological situation, the comparison between the performance of RAQMS vs. RAQMS+RDF or the quantification of the impact on surface ozone and air quality? All these points are discussed but I miss convincing and novel conclusions. Furthermore, a clear understanding of the difference between central concepts such as a stratospheric cut-off, a tropopause fold and Stratosphere-to-Troposphere transport (STT) is missing. The terms are used synonymously which at the very least is confusing if not erroneous. Nevertheless, I recommend the paper to be reconsidered subject to major revisions.

REPLY: The aim of this paper was to provide a detailed trace-gas analysis of two complex stratosphere-to-troposphere transport events. We aim to provide supporting evidence in terms of meteorological overviews, an assessment of the routinely-used air quality forecast model, RAQMS and a discussion of the likelihood that these events influenced surface O3 measurements. To clarify the context of the paper we have re-worded and added a few lines to the last paragraph of the introduction section. We have re-worded the introduction section discussion, adding additional detail related to the terms STT, tropopause folds, cut off lows etc. We have changed terms as suggested in the minor comments section to address the confusion over stratospheric cut-off, a tropopause fold and Stratosphere-to-Troposphere transport (STT).

Major comments: A) STT occurs when stratospheric air crosses the tropopause and enters the troposphere. In order to talk about STT events, one thus first has to define an appropriate boundary surface, i.e., the tropopause. This can be done in many ways (Thermal lapse-rate (WMO), Dynamical tropopause (using PV), Chemical tropopause (using O3 and gradients of O3)...) but without specifying what boundary surface is used, the concept of exchange is meaningless. In this study the tropopause, although never clearly defined, is regarded as some fixed height (365 hPa, p.10171, l.13, why this value?) and all stratospheric air which descends below this level is regarded as 'STT event'. A constant height (or pressure) is not a good tropopause definition because clearly, it neglects nearly all important temporal and spatial variations including the interplay between the tropopause and meteorological structures such as the jet stream and cyclones. Even though stratospheric air in a tropopause fold is often irreversibly mixed into the troposphere (STT) by turbulence and small-scale mixing, this is not always the case. The same applies for the air within a stratospheric cut-off - it might merge again with the stratosphere, resulting in no or very little STT occurring. It is important to investigate the transition from stratosphere to troposphere and not to take it for granted as soon as the air reaches some height level. REPLY: We apologize for the misunderstanding. The height/pressure referred to on p.10171, l.13 is in reference to the aircraft ceiling on these flight days, the tropospheric O3 amounts calculated in section 3.3 are from all measured data and not a result of using a predefined tropopause height. We have re-worded this section in hope to better explain this. RAQMS model uses the thermal tropopause to define the tropopause height and words have been added to section 2.2 to emphasize this point. We have also added a paragraph discussing the tropopause to section 2.2.

B) Concerning the Lagrangian analysis using back-trajectories calculated from RAQMS

output, the use of the term 'origin of the STT event' for the location of the backtrajectories 6 days before initialisation is very misleading. What makes this time step so special? Why is it not 5 or 7 days? A meaningful 'origin' would be the location where the trajectories fulfill some objective and relevant criterion, e.g. cross the tropopause. Although the back-trajectories (shown in Figs. 6 and 11) provide useful information about the history of the air sampled by the aircraft, the analysis of the 'Origin of STE Encounter' is thus arbitrary in the sense that its results ('origins' mostly located over the east coast of Asia etc.) highly depend on the choice of 6 days as 'time since origin' and the sensitivity to this choice is not discussed REPLY: The objective of the Lagrangian analysis is to determine the history of the STT sampled by the aircraft and the reviewer is correct in pointing out that referring to the location of the trajectories six days prior to the encounter as the "origin" of the STT event is misleading. We now use the term "location of the flight track STT encounter" instead of "origin"

C) In line with comment A), the problem of an ill-defined tropopause (as a constant pressure level) also applies to the conclusions reached in Section 3.3 (Stratosphere-to-troposphere implications). I find the choice of 'total column ozone below 365 hPa' an inappropriate measure of tropospheric ozone since any (reversible) displacement of the tropopause below this pressure level is interpreted as a transition from stratosphere to troposphere (STT) and thus as having an influence on tropospheric ozone. REPLY: See response to A).

D) The results from the 'Clean Continental PBL Exposure' analysis (Figs. 4 and 9, lower right) seem spurious to me. How can it be that air at 10 km height spent more than 75% of the last 6 days in a clean PBL (p.10169, l.24)? How did it get up there so quickly? What is the percentage of exposure to 'polluted' PBL? Even 40% (mentioned on p.10166, l.9) seem unrealistically high to me. And how do you explain the very sharp vertical gradients in PBL exposure between 6km and 10 km in Fig. 9 (bottom right)? What is the vertical resolution of your model in this region? REPLY: In responding to the reviewer's comments we carefully reviewed our post-processing of the RDF back-trajectories and identified an error in the calculation of clean and polluted continental PBL exposure which resulted in erroneously showing the percent of time spend in the marine PBL. We appreciate the reviewer pointing out this spurious result. The figures have been updated and now they show significantly lower percentages for clean continental PBL exposure overall, and no exposure to either clean or polluted continental PBL in the vicinity of the STT encounter for either events. We have also clarified that we include back-trajectories which are exposed to convective mixing over the continental PBL in our exposure calculations since convective mixing links the PBL to the back-trajectories and changes the mixing ratio within the air parcel. Rapid convective transport of PBL air aloft accounts for the relatively high exposure to clean continental PBL air above 8km in figure 9. Sharp vertical gradients in PBL exposure (and other RDF diagnostics) occur due to vertical shear along the back-trajectories, which led to sampling of very different air-mass histories at different altitudes. This has been noted in the discussion of the O3 gradients in figure 4.

Minor comments:

1) Page 10159, line 16: 'STT events are episodic in nature...winter-spring period': The correctness of this statement depends on what you mean by STT events and on the geographical location. The seasonal cycle of tropopause folds is shown in Sprenger et al. (2003) (doi:10.1029/2002JD002587) and the seasonal cycle of STT mass flux is presented in the STE climatology of Sprenger and Wernli (2003). A very recent STE climatology also shows the STT ozone flux (Skerlak et al. (2013) (ACPD doi:10.5194/acpd-13-11537-2013)). REPLY: We have re-worded this sentence/paragraph.

2) Page 10164, line 1: I would have appreciated a short explanation/definition of the 'Lagrangian average'. Same page, line 13: As already mentioned by the editor, I would replace 'isentropic PV' by 'PV' only. REPLY: Added brief definition to the sentence discussing Lagrangian average. Deleted "isentropic".

3) Page 10165, around line 18 and Figure 3: If you expect lower CO2 mixing ratios in

the stratosphere, how do you explain the strong increase above 350 hPa in Fig 3b)? And why does the measurement of CO2 in Fig 3c) stop at roughly 400 hPa? Furthermore, I don't see a clearly reduced CO2 'between 400 and 600 hPa' in the offshore profile but only between approximately 520 and 580 hPa. Also, the H2O seems to be generally very low above roughly 560 hPa - differences above this level are not visible and the region between 400 and 600 hPa is not drier than the air above it (or it is at least not visible from the plots). In the SJV profile (Fig 3c), it is very hard to see the minimum in CO2 (The curve looks quite constant, except for some very small variations and maybe a small local minimum around 630 hPa?). The (local) minimum of H2O and maximum of O3 is visible at 550 hPa, as described. REPLY: We carefully inspected our CO2 measurements and notice a fluctuation in the cavity pressure of the CO2 instrument at the start (top) of the profile, resulting in a peak in corresponding CO2. We have removed the contaminated data from the top of the profile in Figure 3b. The difference in top altitude of profiles between offshore (Fig 3b) and SJV (Fig 3c) is due to the longer warm-up period required by the CO2/H2O instrument before starting to sample. On this day it did not start recording until mid-way through the first (SJV) profile. In response the H2O description of Fig 3, the description has been updated in the text.

4) Page 10166, line 23: What is the conclusion from the 'large-scale mixing' analysis? REPLY: Paragraph has been reworded

5) Page 10167, line 25: What you call a 'significant amount of dispersion' looks like a quite coherent flow along the westerly jet to me. Nevertheless, the large zonal spread in 'origins' in a region of strong horizontal ozone gradients is likely the main reason for the very large standard deviation mentioned in the same paragraph. I am not absolutely sure I understand what you mean by 'high RDF O3 within the STT event': do you only consider the back-trajectories which have a Lagrangian average O3 greater than 120 ppbv and are sampled by the aircraft (nearest model level to flight altitude) or all the trajectories (from all vertical levels) along the flight curtain (with RDF O3 >

120 ppbv)? Also, it is unclear to me how the values of 97+-7.5ppbv in the 'analysed STT' (p.10168,l.12) can then be connected to the data presented in Fig. 5. If I understood this correctly, you are referring to the episodes where RDF O3 > 120 ppbv (one episode between approximately 18.3-18.4 h and one around 19.2 h) but the average measured O3 in these two episodes seems to be less than 97 ppbv? REPLY: High RDF O3 within the STT event" refers to all back-trajectories below 8km which have Lagrangian averaged O3>120ppbv, not just those back-trajectories that are sampled by the aircraft. The values of 97+/-7.5 ppbv are for all the back-trajectories below 8km with RDF O3>120ppbv, not just the periods sampled by the aircraft.

6) Page 10168, line 20: I would refer to this structure as a tropopause fold, rather than a stratospheric intrusion since the latter is a Lagrangian concept describing stratospheric air which entered the troposphere and the former is 'only' a deformation of the tropopause. Same page, line 27: The enhanced PV and O3 also extend below the jet core and even to the south of it. REPLY: Wording has been changed.

7) Page 10171, line 7: What do you mean by 'narrow STT event'? Line 8: Please elaborate how inertial gravity waves could have contributed to the very high ozone values. Line 9: I think you wanted to refer to Fig. 7 (not Fig. 11). REPLY: Changed description from 'narrow STT event" to "fine filament structure". Inertial gravity waves have been elaborated upon. Changed Fig reference to 7.

8) Page 10173, line 10: The observed peaks in hourly ozone from several measurement stations might indeed be influenced by the stratosphere (best candidate is the peak around 00:00 on 6th of June in South Pass, WY, Fig. 12 b) but closer analysis of the transport of the air from the stratosphere to this station would be needed. Since the South Pass station is not visible in Fig. 7 it is not possible to assess the meteorological situation over Wyoming. Also, it might be useful to investigate why the Zion National Park station (red in Fig. 12 b) shows two subsequent peaks whereas the Great Basin NP (black) and South Pass (cyan) only show one. REPLY: We have added a brief description of the meteorological overview for Wyoming during 5-6 June 2012 to the

discussion of the surface O3 enhancements. The second peak at Zion National Park occurs at 1500 local time on 6 June 2012. Given that this O3 enhancement occurs in the middle of the afternoon and that is separated from the earlier enhancement it may be difficult to determine the origins of the enhancement from the photochemically-induced O3 diurnal cycle.

9) Page 10174, line 25: I agree that 'total tropospheric ozone' (as defined in this paper) is not a very good indicator for tropopause folds, but I think the RAQMS analysis does a reasonable job in capturing both the cut-off and the tropopause fold (vertical cross-sections in Figs. 2 and 7). REPLY: We have re-worded this paragraph.

10) Page 10175, line 6: It seems quite obvious to me that a global model with comparatively coarse resolution (1°x1° x35 levels) cannot capture all the small-scale variations of ozone mixing ratio near tropopause folds and thus also underestimates the peak values. Why this is also the case for a nested meso-scale model with higher resolution, where for example the tropopause fold develops fully within the nested model domain, is not a priori clear to me and I am looking forward to seeing your results from the RAQMS/WTF-CHEM simulations. REPLY: The initial nested domain did not include the Gulf of Alaska where the filament was initially generated, we have re-run the nested domain over a larger area and resolve the fold better, but still underestimate the ozone within the fold.

Additional comments concerning the figures: Fig. 2: Why do you put the coordinate labels in the middle of the plots (top row) and not outside? The white wind vectors are barely visible. White contours not mentioned in the caption. REPLY: White wind vectors and contours have been changed to black. Contours in the bottom plots are wind speed in m/s which has been added to caption. Coordinate labels are in the middle of the plots because that's what IDL map_set does by default.

Fig. 3: Image quality is not very good. In contrast to the caption, there is no change in the ozone horizontal scale. REPLY: Plot and caption have been updated.

Fig. 6: Does 'STE Encounter' accurately describe the origin of back-trajectories with RDF $O_3 > 120$ ppbv? It would be very useful to indicate the 135° E meridian in the plot above. Although clear, mention that the wind is represented by white contours. I find it questionable to project all 'origins' onto the vertical cross-section at 135°E. The 'origins' are spread from South Korea to the middle of the North Pacific and thus I would only show the 'origins' located within a reasonable distance from the chosen cross-section. REPLY: We have added meridians to the plots. We acknowledge the reviewer's suggestion but prefer to show the location of all the back-trajectories from the STT encounter since that is what is shown on the map. We have re-worded the caption in relation to comment B).

Fig. 7: As Fig. 2. REPLY: Plot updated, see response to Fig. 2.

Fig. 8: Image quality is not very good. REPLY: Figure has been updated

Fig. 9: Very strange patterns (see comment D) Reply: Figure has been updated; see response to B).

Fig. 11. As Fig. 6 Reply: Figure has been updated, see response to Fig. 6.

Response to Anonymous Referee #2

General comments: This paper presents two very nice case studies of stratosphere-troposphere-transport above California using airborne in situ measurements and the Realtime Air Quality Monitoring System (RAQMS). The comparisons between the data and the RDF calculations are particularly interesting. The manuscript is well written and requires only minor revisions, primarily in the figures.

Specific Comments: 1. The study emphasizes the use of CO_2 as a "non-traditional" tracer for stratospheric air. Since CO, N_2O, and HNO_3 have much larger gradients between the troposphere and stratosphere, these compounds provide much better tracers of stratospheric air. What is the rationale for using CO_2? REPLY: The study took advantage of the Alpha Jet payload (O_3 and Picarro instruments). We noticed a con-

sistent and strong anti-correlation between CO_2 and O_3 when comparing tropospheric and stratospheric airmasses. Since CO_2 is not often reported within stratosphere-to-troposphere transport and because of the consistent trends in CO_2, we decided CO_2, in conjunction with other more well-associated stratospheric tracers (O_3, N_2O, CO etc) can be used to help detect stratosphere-to-troposphere transport as a first screening step, especially in assisting with identifying between and likelihood of stratospheric or lofted local- or long-range transport layers (where CO_2 increases often with O_3).

2. Much emphasis is placed on the utility of in situ measurements for probing STT events. Perhaps a word or two could be inserted to contrast the relative merits of in situ measurements versus lidar soundings, which at the very least are complementary and in some cases more advantageous. For example, although aircraft can sample other tracers besides ozone at higher resolution and under cloudy conditions, ground-based lidars can provide continuous 2-D measurements of ozone structures under clear sky conditions. REPLY: Reference to O_3 Lidar in capturing STT mentioned in the introduction and conclusion sections.

3. The precision for the 2B O_3 monitor is stated to be 2 ppbv for a 2-min average, but the data plotted in Figures 5 and 10 are clearly recorded at a much higher sample rate. What is this sample rate and what is the actual precision for these data? The flight speed of the Alpha Jet and the corresponding horizontal averaging time should also be explicitly stated. Do the 20 ppbv fluctuations in the ozone data near 19.0 UT in Figure 5 reflect ozone variability or measurement precision? Finally, since the 2B is sensitive to water, can the AlphaJet fly into clouds or are the measurements limited to clear sky (i.e. dry) conditions? REPLY: The precision described to in the manuscript is referring to data taken every 10 s over a period of 2 minutes (short term precision) and 1 hour (which we call instrument drift). We have re-worded the sentence referring to precision in the manuscript and added a concluding sentence of our understanding of the overall uncertainty (P7, L5, L10-11). Figures 5 and 10 have been re-plotted using 10s data (we record 2 s data but output 10 s data which has better precision and stability and

is in-line with data-handling procedures by other researchers/organizations using the 2B instrument). From our laboratory and chamber tests we did not see any effects that would influence instrument precision up to 20 ppbv. The data near 19.0 UTC is taken during the high altitude transit from the San Joaquin Valley to offshore. Given the instrument's response and precision observed in the profile(s) and boundary layer sectors (when we would expect precision to be most affected by changing conditions (pressure, turbulence etc)) and given results from the laboratory/chamber tests, we conclude that the data near 19.0 UTC of Figure 5 is a result of ozone variability and not a result of instrument precision. Agreed the instrument and Alpha Jet are biased towards clear sky or high altitude clouds conditions. We attempt to avoid flying into regions of condensed moisture, hence during the 14 June 2012 flight the Alpha Jet did not descend though the offshore marine stratus layer.

4. The discussion of surface impacts on pages 10172-3 is relatively weak and unconvincing compared to the rest of the paper. This section and Figure 12 should either be expanded and supplemented with additional meteorological information and trajectory calculations or deleted from the manuscript. REPLY: Some extra sentences detailing the meteorological conditions after the 5 June have been added to the description.

Technical corrections: p10160, L: "tropospheric" is misspelled. REPLY: Corrected.

p10160, L10: The reference for the 15-35 ppbv background concentrations should be Fiore et al. 2003 (already in the list). REPLY: Suggested changes made.

Figure 1: The legends on this figure and most of the others are too small and faint to be easily read. REPLY: Updated figure.

Figures 2 and 7: The wind vectors and aircraft flight tracks in the upper panels are very hard to see. It would be useful if the location of Oakland were also shown on these maps. REPLY: Figures and captions updated.

Figures 3 and 8: Since all of the other plots use altitude as the vertical coordinate,

it would be better to either change the axes in these figures or add altitude axes on the right hand side. Panel (a) would be much clearer if the potential temperature scale were expanded (270 to 340K) and water was plotted on the top axis as mixing ratios (in a different color) to make comparison with (b) and (c) easier. The green traces on (b) and (c) are hard to see. Maybe these should be changed to red? Despite the caption, the ozone scales appear to be the same. REPLY: Recommended changes have been made. Y-axis is altitude in line with other plots, wording in text has been changed to refer to altitude (and pressure) when referencing these figures. Figure a) has been re-plotted to expand the potential temperature scale, the color of CO_2 and H_2O have been changed in figures b and c.

Figures 4 and 9: The time axes should be plotted in hh:mm since these are the units referred to in the text. REPLY: We have added both HH:MM and decimal time to the text describing these Figures.

Figures 5 and 10: It would be helpful if the aircraft altitude were plotted on the right axis in a new color. Also, the time axes should be plotted in hh:mm since these are the units referred to in the text. REPLY: Figures have been updated. We have added both HH:MM and decimal time to the text describing these Figures.

Figure 6 and 11 (upper) A longitude scale on these plots would be helpful with a vertical line along 135 and 150 E, respectively, to orient the bottom panels. REPLY: Figures updated.

Figure 12: A third panel with a map of the US showing the locations of these surface sites should be added. REPLY: Map showing surface site locations has been added.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 10157, 2013.