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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Mixing between a stratospheric intrusion and a biomass burning plume" *by* J. Brioude et al.

J. Brioude et al.

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Reply to review 1 and 2:
We respond to the reviewers 1 and 2 and a revised version of the manuscript will be submitted to the editor. We would like to thank the reviewers and the editor for their time and effort and appreciate their positive reviews of our paper.

Introduction

Referee 1: " the introduction and the discussion are too short (...). In particular, more information can be given concerning the Stratosphere-Troposphere exchange, and the role of biomass burning plume in the atmospheric chemistry.

=> The introduction has been expanded as follows:

Since the pioneering work of Danielsen (1968), tropopause folds have been a focus of atmospheric research because they are one of the main sources of stratospheric ozone flux into the troposphere. They occur in cutoff lows (Price and Vaughan, 1993) or in frontal systems, and are more frequent near the subtropics (between 20^{o}N and 40^o) than further poleward (Sprenger et al., 2003). The net ozone flux across the tropopause is downward from the stratosphere to the troposphere at midlatitudes, with a maximum in winter/spring and minimum in summer/fall (Sprenger and Wernli, 2003). Using a 15-year climatology of cross tropopause exchange, James et al. (2003) have shown that stratospheric airmasses over United States reach the lower troposphere above southeastern United States and the northern part of the Gulf of Mexico in winter. In summer, the impact of stratospheric airmasses is much weaker and is most prominent above the northern United States. Much attention has been paid to the decay of tropopause folds in the troposphere because the ozone that they contain contributes to the OH radical concentration which affects the chemical budget of different trace gases in the troposphere (Esler et al., 2001). Kentarchos and Roelofs (2003) have shown that the stratospheric ozone flux contributes up to 15% to the tropospheric OH radical concentration in the Northern Hemisphere. To have a large impact on the tropospheric ozone concentration, the stratospheric air in the fold must mix irreversibly with the surrounding tropospheric air. Previous studies have shown that tropopause folds can disperse into decaying warm conveyor belts (Cooper et al., 2004), and they can also be interleaved with anthropogenic pollution (Cho et al., 2001; Parrish et al., 2000). Irreversible Stratosphere-Troposphere Exchange (STE) occurs between these layers at small scales resulting in a layer with elevated ozone and reduced CO relative to adjacent unimpacted tropospheric air. Another common, but completely unrelated, influence on the composition of the troposphere arises from biomass burning (BB) emissions. BB is known to be one of the main sources of aerosols, carbon monoxide (CO), volatile organic compounds and nitrogen oxides (NOx) in the troposphere (Andreae and Merlet, 2001). During the summer 2004 NEAQS-ITCT 2k4 study period, plumes from Alaska and western Canada were sampled on multiple flights above the

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eastern United States (de Gouw et al., 2006). In the region of New York and Boston, biomass burning was more important than anthropogenic CO sources at altitudes between 3 and 5 km (Warneke et al., 2006). BB can also affect ozone concentrations at the continental scale. During the summer of 1995, large forest fires in northern Canada caused high CO mixing ratios at ground level in the southern and eastern United States, and were responsible for 10 to 30 ppbv ozone enhancements over a period of 2 weeks (Wotawa and Trainer, 2000; Mckeen et al., 2002).

While BB plumes and stratospheric intrusions occur frequently in the troposphere, to date, mixing between the two has not been reported. In this case study, we document the first clear case of mixing between a tropopause fold and a biomass burning (BB) plume in the free troposphere. The event occurred during the Texas Air Quality Study/Gulf of Mexico Atmospheric Composition and Climate Study (TexAQS/GoMACCS), conducted during August-October 2006 across eastern Texas and the northwestern Gulf of Mexico to investigate the influence of anthropogenic trace gases and aerosol emissions on the air quality and radiative balance of the region. In this paper, we use in-situ measurements of ozone, carbon monoxide, aerosol extinction coefficient, acetonitrile, nitric acid and relative humidity measured from the NOAA P3 aircraft to study the chemical signature of mixing between a biomass burning plume and a stratospheric intrusion. We use a Lagrangian particle dispersion model and satellite imagery to investigate the transport mechanisms that bring together the tropopause fold and the biomass burning plume. Section 2 presents the in-situ measurements and the modeling methods. Section 3 presents the analysis of observations, model calculations and satellite imagery. Finally, conclusions are drawn in Section 4.

Conclusion:

Referee 1: the introduction and the discussion are too short, and a real conclusion is missing. The meaning of the percentages given in the discussion is not clear, so the authors must discuss this in details.

Referee 2: P 8025 The discussion of the statistical analysis is very sparse and

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could be improved in clarity. I have trouble understanding the motivation and what the relevance is. Is this a surprising or expected result?The conclusion that this kind of mixing can influence tropospheric chemistry is not really substantiated and the claim about possible impact on surface air quality seems pretty far fetched. Do the measurements demonstrate anything like that? If anything, O3 seems to decrease in the mixed part of the stratospheric intrusion. With all the concurrent measurement from the P3, I would think the authors have some evidence for this statement.

=>The conclusion has been expanded. We have changed also the statistical analysis to make it clearer.

Ozone, carbon monoxide, aerosol extinction coefficient, acetonitrile, nitric acid and relative humidity measured from the NOAA P3 aircraft during the TexAQS/GoMACCS 2006 experiment, show evidence of mixing between a tropopause fold and a biomass burning plume above east Texas at 16:00UTC on 25 September 2006. We used the FLEXPART Lagrangian particle dispersion model to simulate biomass burning transport and stratospheric intrusions. We found that a biomass burning plume originating north of Los Angeles was mixed with air from a tropopause fold that originated above the central United States. This Lagrangian-based study confirms that mixing does occur between biomass burning plumes and stratospheric intrusions in the free troposphere. But is this a common occurence? To answer this question we performed a statistical analysis using FLEXPART forward simulations from 15 August to 1 October 2006 to assess the frequency of such mixing in the Texas region (110^{o}W 90^{o}W, 35^{o}N 28^{o}N) during September. We used the same FLEXPART parameterization for stratospheric tracers and biomass burning as used for this case study, but assuming an area burned of 180 ha per fire detection (Stohl et al., 2007). A stratospheric intrusion was deemed to be present within a grid cell if the ozone tracer mixing ratio exceeded 30 ppbv, and a biomass burning plume was deemed present if the biomass burning tracer

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mixing ratio exceeded 10 ppbv. Two stratospheric intrusions were found at the surface of the Texas region in September. These intrusions were mixed with biomass burning plumes in 26% of the grid cells related to the stratospheric intrusions. Between 100 m and 4 km of altitude, six stratospheric intrusions were found over Texas, and 40% was mixed with biomass burning plumes. Between 4 and 8km, nine stratospheric intrusions were found over Texas, and 8% was mixed with biomass burning plumes. This percentage decreases with altitude because the biomass burning plumes are less frequent in the free and upper troposphere. These results show that mixing between stratospheric intrusions and biomass burning plumes were relatively common in the lower/free troposphere above Texas during September. This study indicates that mixing between stratospheric intrusions and biomass burning plumes can influence tropospheric chemistry, but further studies are required to determine if such events have any implications for surface air quality.

Specific comments

Referee 1 The y-axis labels on Figure 23 ("ST" and "BB") must be explained in the legend.
The labels have been added.

referee 2 p 8019 Instrument and model description is concise and to the point.
We agree

p 8021, l22. The authors state that the HNO3: O3 relationship is 0.0022*O3- 0.055 and then say that typical UT/LS HNO3 is 300 ppt. It is not quite clear how this equation relates to the concentration in the UTLS, it would be more useful to quote the relationship found in the UT/LS. Please add units to be used. E.g." HNO3 [ppb] = 0.0022*O3[ppb]..."
The sentence has been corrected as follows: The HNO3 to ozone slope is HNO3[ppbv]=0.0022*O3[ppbv]-0.055, and is consistent with previous measurements obtained in the lower stratosphere (NOy[ppbv]=0.0029*O3[ppbv]+0.16, Neuman et al.,

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2001).

P 8022, L 4-5: I can't really see horizontal or vertical branches, though I think I know what the authors are talking about. I suggest also in the context of the discussion of the P3 data, to overlay the different branches with labeled lines that can then easily be referenced in the text.
=> The branches have been labeled.

L25 the modeled trace is also not correlated after 16:20, where measured CO stays flat at an elevated approximately 100 ppb, whereas modeled CO goes to 0, which seems to say that the model does not capture the source for the spikes after 16:20.
=>After 16:20, the aircraft was changing altitude from 4 to 5km. This explains why the anthropogenic CO tracer is decreasing from 20 to 10ppbv. The fact that the measured CO stays at 100ppbv with small peaks while the anthropogenic CO tracer decreases may be due to the presence of a narrow biomass burning plume near the aircraft at this time. FLEXPART is probably not accurate enough to calculate such small and narrow CO peaks. Besides, FLEXPART is not able to resolve the actual size of the two CO peaks at 16h10.

Figure 2 As mentioned above, add labeled lines for clarity and easy reference in text. Adding a colorbar for the grayscale may be useful and make these measurements more quantitative.
=> Rather than adding a grayscale colorbar, we prefer to add the number of MOZAIC flights and quote the number of MOZAIC data related to the white grid cell. The gray shading is based on 27 MOZAIC flights. The most frequent ozone/CO grid cell measured by the MOZAIC flights (white color) is related to 194 data points in the whole troposphere, and 62 data points in the free troposphere.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 8017, 2007.