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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric ozone variations at the Nepal climate observatory – pyramid (Himalayas, 5079 m a.s.l.) and influence of stratospheric intrusion events" *by* P. Cristofanelli et al.

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Review of "Tropospheric ozone variations at the Nepal climate observatory. . ." by Cristofanelli et al . This paper presents an analysis of ozone and related data from the NCO-P observatory at 5 km in the Himalayas. It is significantly improved over an earlier version that I also reviewed. The analysis focuses on the influence of stratospheric intrusions and the fraction of tropospheric ozone due to S-T exchange. While the analysis appears to be reasonable and the data are extremely valuable, there are some key points that are not clear and several important caveats that the authors fail to

discuss. I believe that once these points are clarified, the manuscript could be ready for publication.

We thanks the referee for the very useful and detailed comments which help us in strongly improving the paper. All points were accurately evaluated and point-to-point discussed in the following. We reported modifications now introduced along the paper by italic characters .

1)Important caveats: The method to derive the total stratospheric influence relies on the identification of specific time periods with strat influence. This method fails to account for stratospheric contribution which is part of the background. Certainly the high value of average O3 during the pre-monsoon period is partly due to mixing of stratospheric air into the background. This would result in an under-estimate of the strat contribution to trop air.

We agree with the referee that this analysis can underestimate the amount of stratospheric contributions already part of the background. In fact, the paper is mainly focused on the analysis of "stratospheric intrusion events", as clearly indicated in the title. To make more clear this point, we modify the paper title in *"Tropospheric ozone variations at the Nepal Climate Observatory – Pyramid (Himalayas, 5079 m a.s.l.) and influence of deep stratospheric intrusion events"*. Moreover, to further clarify this point, we introduced the following sentence in the introduction: *"It should well clarify that part of this work is aimed in evaluating the contribution of relatively "fresh" stratospheric inputs, i.e. still having a clear fingerprint of their stratospheric origin".*

Finally, in the discussion paragraph we cited that: *"It should be noted that in this analysis we assessed the contribution from relatively "fresh" SI, for which still clear stratospheric "fingerprints" were observable."*

Concerning the high O3 values observed in the pre-monsoon, certainly a contributions from stratospheric air is expected, however as shown by Bonasoni et

al. (2010) also contributions related to pollution transport (from biomass burning and industrial/traffic/energy sources) are very likely. As already reported in the paper (Section 3): ***"during the pre-monsoon period, both STE and long-range/regional transport from Central Asia, North Africa and Middle East (Wang et al., 2006; Sudo and Akimoto, 2007; Bonasoni et al., 2010) can contribute to the observed O3 concentrations".***

2)The other important caveat is the specific method to identify time periods with stratospheric influence. The method likely over-estimates the amount of time with strat influence (although it is a bit difficult to be sure given some confusing points in the description.) These two caveats MIGHT balance out, but not likely. While I don't think they completely invalidate the analyses, the authors need to discuss these uncertainties.

In the revised paper we re-organized this section. Following also comments by other referees, we better motivated the adopted thresholds. Moreover, a discussion on criteria sensitivity has been introduced in the paper (see also the answers to referee 3).

We don't agree that the methodology is likely for strongly over-estimating the time with stratospheric influence. As described in the section 5, a careful analysis of in-situ O3, BC and RH were conducted to identify the time-period for which stratospheric air-mass influenced the measurement site during the SI identified by the methodology described in the section 4.1. However, with the purpose of better evaluate the uncertainties connected with the selection of SI, as reported above, we present a sensitivity study on the criteria applied. As now reported in section 4.1: ***"The simultaneous application of the four criteria identifies only air masses characterized by extremely clear stratospheric properties, while ignoring other weaker SI events, thus leading to the identification of a very low number of days (4) influenced by SI. It should be noted that the last criterion was the most active in selecting possible SI. As being based on the analysis of not completely unambiguous stratospheric tracers (i.e. RH and O3), we introduced***

additional thresholds for other tracers (i.e. AP, PV, TCO) to minimize this ambiguity in the SI identifications. Moreover, we performed a sensitivity analysis by adopting different RH values. For instance, assuming the widely used 40% threshold for RH (e.g. Stohl et al., 2000; Trickl et al., 2010) the total number of selected SI reduced only by 2%. On the other side, the (iv) criterion should lead to the selection of further 15 SI days if the additional thresholds on AP, PV and TCO were not considered."

3)As for the confusing part, I found the values in Table 2 impossible to follow. First, is N the number of 30 minute data points?

Yes

If so, then why are there only 1600 hrs (N=3183) in all seasons?

Yes, this number represents the hrs during which stratospheric air-masses was present at the measurement site according to the selection methodology. This is in agreement with the value (expressed in hrs) reported in the paper (Section 5): 1,591 hours (10% of the analysed time period). Probably, the table caption wasn't clear enough, thus we decided to modify it: ***"Table 2: For periods affected by SI at the NCO-P: seasonal averaged O3 concentration (O3, where N represent the number of 30-min data), O3 excess (△O3) and maximum O3 integral (O3S, see section 5 for definition)."***

Second, I can't reproduce the values for ppbv*hr. For example under pre-monsoon, for N=1396, assuming 25% time is strat influence, gives approx N=400 or 200 hrs. 200hrs x 9 ppbv = 1800 ppbv*hr, not 4.5e4. The authors need to do a better job of explaining these values and how the calculations were actually done.

As reported in the formula (1) and in the manuscript (Section 5), the "integral" O3 was calculated by summing the 30-min O3 concentrations and NOT the excess of △O3. Obviously, in this case, one should assume that all the O3 detected during

a SI can be traced back to stratosphere. This measure was provided to give information about the upper limit of SI contribution to O3 at NCO-P. This point was now better explained in the abstract, Section 5, Table 2 caption as well as in the "Conclusion" section (see the following relating answer), also discussing the uncertainty related to this estimate. To this aim, we provided the lowest limit of SI contribution to surface O3 at NCO-P by using $\triangle$O3.

Even if the "integral" SI contribution to surface O3 at NCO-P can be characterised by a large uncertainty, the authors think that the obtained results provide a clear indication of the potential impact of SI on the surface O3. As now reported in the conclusions: *"Even if the present estimates are associated to a rather large uncertainties, SI can play a not-negligible role in determining the levels and variability of tropospheric O3 over the southern Himalayas, having significant potential impacts on the regional radiative budget, particularly during the strongest events."*

Other points: 1484, Line 15: Whether S-T is the largest natural input to the trop is arguable.

OK, the sentence was modified.

1489, line 12: Is uncertainty for 1 min, 1 hr, ??

1 min, as now reported in the manuscript.

1490, line 4: 273 or 298K?

273 K

1492, line 29: Are there significant emission sources within one day, if not, this explanation is not believable.

Yes, this is possible. As reported by Bonasoni et al. (2010) "NCO-P is located away from important anthropogenic sources of pollutants, and only small vil-

lages are present along the valley: Lobuche, Pheriche, Tyangboche, Namche Bazar (the biggest village with about 800 inhabitants), Phakding and Lukla. The closest major urban area is Kathmandu (1 081 845 inhabitants; 2001 census), situated in the valley of the same name (estimated population of the valley in 2009 was about 3 million). The city, located about 200 km South-West of the measurement site and more than 3.5km lower ...". Observational evidences collected during events of extended pollution and biomass burning in the Indogangetic Plain and Himalayas foothills, suggest that pollutants and spread haze are pilling up against the Himalayas ridges and impinging valleys and so transported to the measurement site in 1 single day.

However, we rephrased as following: *"This behaviour suggests the transport of air-masses richer in O3 from along the valley or possibly influenced by a weak local photochemical production."*

1494, line 5: The filtering method is not well described. Please explain what is the goal of this filtering.

We think that this section was now improved (see also answer to referee2, 3 and the second comments in this letter)

1494, line 15: My sense is that this method is going to over estimate the amt of time of S-T exchange. For example, you calculate an array of trajectories and consider the time S-T if even ONE trajectory gets to a PV of 1.6.

Basing on our experience in analysing specific SI episodes at other mountain sites and at NCO-P as well (Cristofanelli et al., JGR, 2003;2006; Bonasoni et al., Atmos Env, 2000; Bonasoni et al., Sci Tot Env,2008), we think that this approach is suitable to detect SI influence. Moreover, it should be noted that in this analysis the PV is not a "stand alone" criterion, but for leading SI identification it had to be corroborated by at least one other tracer (AP, TCO or RH+O3). However, this choice was better motivated in the text (Section 4.1): *"In particular, we de-*

cided to select as influenced by possible SI, the days for which at least one of the 14 back-trajectories ending in a vertical range extending up to 50 hPa from the NCO-P altitude showed PV > 1.6 pvu, basing on the experience gained in the analysis of case studies of stratospheric intrusions at other high mountain stations (Cristofanelli et al., 2006; Cui et al, 2009) and at the NCO-P itself (Bonasoni et al., 2009)"

1495, line 7: Again filtering needs better explanation. As described, one 30 min point can result in selection as an S-T time. This is likely to over-estimate influence.

We remove the words "(basing on 30-min)" as confounding. The analysis was based on daily basis but considering 30-min O3 and RH data. In fact, the greatest part of identified events had a duration grater than 8 hours (only 3 events had durations lower than 8 hours).

1497, line 24: The equation does not seem correct. It seems from the description that only one summation is actually done. I assume n is each individual S-T event. Why is O3 used, shouldn't it be DELTA O3?

Please, see answer to major point 3. Moreover, with the purpose of presenting a "confidence interval" about the possible contribution of SI to ozone, we also provide a conservative estimation of the SI contribution by using Delta O3. In the "Conclusion" (section 6): ***"In order to calculate the maximum fraction of tropospheric O3 due to SI at NCO-P, the integral stratospheric O3 contribution (O3S) was calculated basing on the assumption that all the O3 observed at the NCO-P during an SI can be traced back to the stratosphere. Through this analysis it was estimated that up to 13.7% of surface O3 recorded at NCO-P can be attributed to SI. On a seasonal basis, the lowest SI contributions were found during the summer monsoon (less than 0.1%), while the highest were found during winter (up to 24.2%). Such values, which represent an upper limit of the O3 contributions by SI at NCO-P, are in good agreement with the results of Sudo and Akimoto***

(2007) who, by running a tagged tracer simulation with a global chemical transport model, estimated that STE contributed from 10% to 20% of the annual TCO over the southern Himalayas. However, it should be noted that the present estimates can vary if a different SI selection methodology is applied to screen SI at NCO-P or if different measures of the stratospheric O3 input are considered. In particular, with the purpose of defining the lowest limit of SI contribution to surface O3 at NCO-P, we adopted the very conservative assumption that during an SI only the observed O3 excess (△O3) can be traced back to the stratosphere. In this case, on a yearly basis, the 2% of surface O3 could be directly attributed to SI."

Now we cited also in the abstract the large uncertainties related with these estimation: ***"Even considering the rather large uncertainty associated with these estimates, the provided results indicated that, during non-monsoon periods, high O3 levels could affect NCO-P during SI, thus influencing the variability of tropospheric O3 over the southern Himalayas."***

Table 2: See comments above.

OK

Figure 2: Caption should mention that PV is maximum along xx trajectories, where xx is the total number of trajectories.

OK

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 1483, 2010.