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## ***Interactive comment on “Stratospheric impact on tropospheric ozone variability and trends: 1990–2009” by P. G. Hess and R. Zbinden***

**Anonymous Referee #1**

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This is a valuable and profound modelling study regarding the role of the stratosphere on determining tropospheric O<sub>3</sub> trends and variability for a 29 years time period. The authors make a convincing case from both a measurements and model perspective the stratosphere contributes to large-area interannual variability. However, there a number of places where the paper could better explain the assumptions made- especially regarding the use of Synoz in the model context.

The paper is very lengthy- and it is hard to stay focussed to the end. I suggest that the authors try to condense the manuscript somewhat (e.g. integrate 5.1 and 3.3; avoid a mid-summary. The abstract needs substantial improvement- to be able to read it stand-alone. I recommend this manuscript for publication after taking my suggestion into account.

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Detailed comments:

p. 22720 Abstract: it is not always clear about regions and altitudes the paper is talking. This makes the abstract quite confusing.

p. 22720 I.6 constant of all emissions? or only constant anthropogenic emissions?

p. 22720 I.9 mention at which altitudes, this seems to general

p. 22720 I.15 averaged over which regions (Canada, N and C Europe) or rather Canada-N.Eur-E.US?

p. 22720 I.16 ...simulated variability at 500 hPa or entire troposphere?

p. 22720 I.24 Why not directly present the numbers corresponding to the correct sampling?

p. 22721 I.4 150 hPa signal modelled?

p. 22721 I.5 the remote coastal site Mace Head, exposed to Atlantic ocean air. Note the spelling of Mace Head.

p. 22720-22721 You need to make clear why you want to show two time periods (1990-2009 and 2000-2009). Explain that one of the hypothesis of this paper is that the slowdown of trends in the 2000-2010 partly due to stratospheric ozone.

p. 22772 I. 18 a more recent evaluation of crop losses in Van Dingenen (Atm. Env. 2009)

p. 22721 I. 21 20 % of CURRENT ozone levels- as the authors know this is a non-linear system, and STE attributable surface ozone may have been larger in the past.

p. 22722 I. 1; I would recommend to delete 'deficient' in details; because most models are at least a factor of two higher than the observations.

p. 22722 I. 4 you introduce here the issue of trends, but some readers may not know what is the issue (reports on rising background; in some regions O3 going down due

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to air pollution mitigation.

p. 22722 l. 15 explain trends different in the 90s and 2000s.

p. 22724 l. 10 Check this sentence; doesn't sound correct that RMS variability of concentration is equal to a flux? Does this statement pertain to a specific part of the troposphere, or total tropospheric burden?

p. 22724 l. 20 Explain which region of the troposphere this paper is focussing on? Mainly 500 hPa?

p. 22725 l. 8 what is the exact criterium for a 'valid' dataset? Did you perform an analysis what this use of incomplete dataset means for derived trends? From the abstract I understood yes, but there seems not be a lot of emphasis.

p. 22725 l. 20 What was than the appropriate range for correction factors used in your study.

p. 22726 l. 24 here you speak of 6 regions (see confusion in abstract).

p. 22726 l. 19 step 2? Explain why creating a running mean is necessary? Does this change the trend results? annual average monthly deviation may be a confusing term: perhaps better the deviation of montly value from the multi-annual average?

p. 22727 l. 5 explain why simply creating an average of the six regions would give a realistic NH signal? Perhaps the sampled model results compared to the full model can demonstrate at least partly the validity?

p. 22727 l 15 I think that, although perhaps a good strategy, this procedure may create timeseries with from year-to-year- rather unequal amounts of measurements contributing to the region average ozone values. It is not clear to me whether this procedure may produce artificial trends? Can the authors comment on this? See also remark above.

p. 22727 Not clear what was done with 'natural' emissions, like isoprene/terpenes and

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lightning?

p. 22727 NCEP meteorology is that operational data, or re-analysis?

p. 22728 Tagging to calculate tropospheric ozone and calculating stratospheric ozone from the difference? In principle the Synoz tracer should also give you the stratospheric ozone? Explain.

p. 22729 I. equivalence to 'switching' off? Is this relevant for this paper? That is a linearity issue? It may further be worth to mention that several others have explored the 'tagging' approach; i.e. J.F.Mueller et al; Lelieveld and Dentener (2000), and probably a number as others as well.

p. 22729 Somewhere before it should be mentioned that (probably) there is not explicit stratospheric module, hence the need for Synoz.

p. 22729 I. 15 I am not very familiar with 'Synoz', and the authors modify the method here as well. I would like to ask the authors to explain better what is exactly done. E.g. at what levels Synoz concentrations are prescribed, and how often updated. Also what is the fundamental difference with just prescribing stratospheric ozone levels? Why did you have to abandon the 'original' synoz flux method?

p. 22729 Did the authors check the consistency of global ozone budget using this method? Does the tropospheric ozone production of the colored NO<sub>x</sub>, match that of the 'full' NO<sub>x</sub> ozone production, and is the budget of ozone production, destruction, deposition and stratospheric influx closed?

p. 22729 I. 17 The authors correctly mention that there is no variation in stratospheric chemistry (stratospheric composition?), while we know e.g. from total column measurements there is. In this respect I wonder whether there is not a risk that the stratospheric ozone input trend (as in Figure 1) into the troposphere is not overestimated (due to keeping stratospheric concentrations always the same). Please comment.

p. 22729 Figure 1 ... first 100 hPa is rather sloppy language , (surface pressure - 100

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hPa).

p. 22729 l. 27 again I wonder whether there could be for instance lightning NOx component in the signal attributed to the 'stratosphere'.

p. 22730 l. 8 communicated=>propagated

p. 22730 l. 10 Perhaps here it is time for a reminder to the reader that \*anthropogenic\* emissions were kept constant (especially those of biomass burning can cause ozone variability)

p. 22730 l. 20 Correlation of annual average ozone of model and measurements? I read in the table 2 correlations between 0. and 0.70; what values are discussed here?

p. 22731 l. 6-to l. 22732 again I am not too familiar with synoz and the modified version used here, but my main concern would be the use of a constant concentration- whereas in reality there is variability and feedback. Anyway I would suggest to move this section into an appendix, because it is somewhat distracting from the 'story'. The timescales in Figure 2 are those of stratospheric transport processes?

p. 22731 l. 8 I do not fully understand the concern about Synoz 'not in equilibrium', 'and adjustment towards' equilibrium. I understood that Synoz concentrations are prescribed, so I do not understand what can adjust? Please explain better.

p. 22732 l 11 explain in text and Caption of S1 what the correlation is looking at (annual values, the smoothed monthly residual), how missing values dealt with.

p 22732 l.20 ....unlikely ... not correlated ... akward sentence, do you mean something likely to be correlated? Japan and surrounding are known to have significant contributionso of stratospheric intrusions- which indeed maybe very different between 42 and 32 N.

p 22733 Figure 3; the 3 reddish colors are difficult to discern for my eyes. In final version try to make it as large as possible.

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p. 22734 I. 2 Can you explain the difference between Figure 4 and Figure 3b? I thought Figure 4 is an explanation of the average signal in Fig 3b, but it doesn't seem to fit.

p. 2273 what is meant : 'the MOZIAC record should give more accurate measurements? More accurate than what?

p. 22736 I. 18 Convective overshoots followed by adjustment?

p. 22737 I 7 greater than

p. 22737 I. 10 the name is Sonnblick

p. 22737 I. 21 A more appropriate title could be : Large scale correlation of stratospheric (150 hPa) and tropospheric (500 hPa) ozone from measurements.

p. 22739 I. 1-25 it is a bit strange mix with already model interpretation included; while the modelling section is still to come.

p. 22740 period between 1991 and 1995 excluded? Explain better because everywhere it is mentioned that the periods 1990-2000 and 1990-2009 are analysed. What about the earlier mentioned calculated trends? Do they include or exclude the Pinatubo period?

p. 22741 I 4 Pozzoli et al strictly talk about anthropogenic emission variability which in their case includes biomass burning. They did include variability of natural emissions.

p. 22741 there are a host of other studies that estimated stratospheric ozone at the surface, would be good to see if that is consistent with your study. Overall a very good consistency of model and measurements!

p. 22743 I. 1-10 this difference between large scale full model and 'measurement' sampled should probably be emphasized a bit more. Nevertheless qualitatively the conclusions remain the same.

p. 22743 I. 20 I am somewhat worried that the 'derived' stratospheric ozone component

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includes to some extent a signal of natural emission variability, please comment.

p. 22744 l. 4; the overall signal: you mean the large scale average ozone (surface, all troposphere?)??

p. 22745 0.18 ppbv/yr is a positive trend (the net effect of European and Asian emissions negative) correct? Also the 0.18 ppbv/yr (unfiltered) can be compared to 0.37 filtered (Table 2). Alternatively, with a lot of handwaving one could say that the European influence decrease the trend from  $0.37-0.18=0.21$  ppbv/yr. This is very different from the Fiore based estimate of minus  $-0.03$  to  $-0.05$ . However, I think the Fiore estimate can only be for all weather conditions, so perhaps the factor 0.3 doesn't make sense? Given too much assumptions I would recommend this paragraph to be deleted (or make it more robust). Please also check Pozzoli's simulation- which had the possibility to discriminate between meteo and emissions impacts on (unfiltered) Mace Head data.

p. 22746 l. 5 repeat why the focus on two periods (1990-2000) and 1990-2009.

p. 22748 l. 6 the Zbinden statement on Japan, seems to contradict with the 'ozone' sonde information?

p. 22749 l. 9 I suggest to swap conclusion 3 and 4.

p. 22750 l.1 But realize that some of the natural emission variability was hidden in meteorological variability.

Table 1: Names of stations De Bilt; Hohenpeissenberg, Mace Head.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 22719, 2011.

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