

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

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The authors wish to thank anonymous reviewer #1 for their in-depth review and their many constructive comments. (I will identify reviewers comments in the reply below asC, my reply as R)

C: 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

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Discussion Paper



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.

R: Following the suggestions of the reviewers the authors will extensively rewrite and condense the paper. We will present the data first followed by the modeling details (at the suggestion of Jennifer Logan). We will also include a number of additional tables to summarize the results and thus make the text easier to read. Following the 1st reviewer's suggestions we will integrate section 5.1 and 3.3 to avoid a mid-paper summary. We also will simply the abstract to make it easier to read. In addition following the recommendation of reviewer 1 we will move the section discussing the equilibration of the model to the appendix and remove most of the paragraph in which we discuss components of the Mace Head ozone trend.

The authors will include a more detailed description of how Synoz was used in these model simulations. This description is summarized here: The stratospheric tracer synthetic tracer (Synoz) is described in McLinden et al. (2000). Synoz is a passive ozone-like tracer released into the equatorial stratospheric ozone production region (in our simulations defined between 10 and 70 hPa and 30 S – 30 N) at a rate equivalent to the cross-tropopause flux of ozone (specified in our simulations as 500 Tg/year). In our simulations below 500 hPa Synoz is relaxed to 25 ppbv with a timescale of 2 days. Ozone is set equal to Synoz above the tropopause ensuring the stratosphere to troposphere flux of ozone is equal to that of Synoz. At steady-state the cross-tropopause flux of Synoz is equal to its specified production rate (i.e. 500 Tg/year). Synoz has been used for many years in tropospheric chemical models with a high degree of success. For example Synoz was used in the GEOS-chem model until linearized ozone chemistry (LINOZ, see McLinden et al. 2000) was introduced in a beta version 24 February, 2010.

One of the advantages using Synoz as specified in McLinden et al (2000) is that the cross-tropopause flux of ozone is not sensitive to details of the stratospheric simula-

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tions. It is also not sensitive to interannual changes in stratospheric circulation. Because the production rate of Synoz production is fixed, as the stratospheric mean meridional circulation increases (decreases) an air mass spends less (more) time in the region where Synoz production is specified. This implies that as the circulation increases (decreases) the concentration of Synoz transported out of the equatorial source region (approximately equal to the amount of time that an air mass within the equatorial source region) decreases (increases). Thus, the flux of Synoz transported out of the equatorial source region (roughly proportional to the strength of the mean meridional circulation times the concentration of Synoz) is rather insensitive to changes in circulation strength.

In reality, as the stratospheric mean meridional circulation increases, ozone transport out of the ozone production region should also increase. Specifying the concentration of a Synoz like tracer (we will denote this tracer Synoz\*) within the equatorial production region (instead of its production) implies that its flux out of the equatorial source region will be sensitive to the strength of the circulation; an increase (decrease) in the stratospheric meridional mass flux will increase (decrease) the transport of Synoz\*. This is what we have done in this paper. The parameterization used in this paper was implemented as follows. (1) First we equilibrate the concentration of Synoz (McLinden et al., 2000) by running the model on the order of 10 years. (2) The equilibrated concentration of Synoz in the defined equatorial region (30 S – 30 N between and 10 and 70 hPa) is saved during a test-year producing an annual record of Synoz\* concentrations within the defined equatorial region within that year. (3) We re-run the test year, but instead of specifying the production rate of Synoz we specify concentrations of Synoz\* within the equatorial production region (obtained from step 2). We check that the two methods of specifying Synoz produce the same result during the given test year, and indeed they do. In other words the simulated concentrations of Synoz are very similar to those of Synoz\*. (4) We use the specified concentrations of Synoz\* within the equatorial region during all subsequent years. As stated above this allows us to parameterize the impact of the interannual variability of the stratospheric circulation on ozone while still retaining

the Synoz methodology. The stratospheric-tropospheric flux of ozone during the test year should be very close to 500 Tg/year. The stratospheric-tropospheric flux of ozone during subsequent years using Synoz\* should respond to changes in the strength of the simulated stratospheric Brewer-Dobson circulation in comparison to the test year.

As detailed in the expanded supplement to our revised paper (see figures accompanying our response to Jennifer Logan), and within the paper itself (Table 2) the methodology adopted here produces reasonable simulations of tropospheric ozone and its variability. In addition the STE (Stratospheric Tropospheric Exchange) of ozone using this parameterization is only a measure of changes in the stratospheric circulation and not of changes in stratospheric chemistry. This allows us to document the importance of circulation changes.

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

R: To be clarified in new version.

C: p. 22720 l 6 constant of all emissions? or only constant anthropogenic emissions?  
p. 22720 l.9 mention at which altitudes, this seems to general.

R: Both points to be clarified in new version. All emissions are interannually constant except for lightning.

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

R: To be clarified in new version. Ozone averaged at 150 hPa over the regions with a robust 150 hPa signal (Canada, Europe and Northern Europe) explains 69% of the interannual variability of ozone averaged over the tropospheric regions with a robust signal (Canada, Eastern U.S. and Northern Europe).

C: p. 22720 l.16 ...simulated variability at 500 hPa or entire troposphere?

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R: At 500 hPa. To be clarified in new version.

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

R: Agreed, we should present the trends corresponding to the correct sampling. However, it is also interesting to note the trends with ideal sampling (e.g., no missing data). We will also use a different method to sample the data in the revised version, one less likely to include spurious results due to incomplete sampling (see comment for p. 22725 I. 8)

C: p. 22721 I.4 150 hPa signal modelled?

R: The 150 hPa signal is measured. To be clarified in new version.

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

R: Thank you. To be clarified.

C: 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.

R: Thank you. Will do.

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

R: Thank you. We will include.

C: 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.

R: Thank you. We will clarify this point.

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

R: We agreed. Thank you.

C: p. 22722 l. 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 to air pollution mitigation).

R: Thank you. We will clarify the issue.

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

R: Thank you. We will do.

C: 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?

R: The reviewer is correct. We will clarify this sentence relating ozone variability and STE flux in our summary of results in Hsu and Prather (2009).

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

R: Yes. We will clarify.

C: 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.

R: We included ozone measurements from ozonesondes north of 30 N from the WOUDC database. Only those stations with long-term records that included at least most of the period from 1995-2005 using a single measurement technique (i.e., either ECC or the BM techniques) were used. We did not do an extensive analysis of the

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impact of incomplete datasets on derived trends. In our revised paper we will change our analysis method slightly: we will compute trends for each measurement site separately and then use a weighted average to compute the trends for each region. In the submitted version of the paper we averaged the measurements regionally, and then computed the trend for the region. This may cause spurious results for some reasons if there is incomplete data.

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

R: We used the selection criteria as given in Logan (1999): 0.8-1.2 for ECC sondes and 0.9-1.35 for BrewerMast sondes. These criteria will be clearly stated in the paper.

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

R: Thank you. The 6th region is Southern Europe, but since there is only one measurement site there we could not construct a robust regionally averaged ozone signal for Southern Europe. We will clarify this confusion.

C: 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 monthly value from the multi-annual average?

R: The running mean simply creates an annual average of the monthly deviations. We will change the term “annual average monthly deviation”. Thanks.

C: 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?

R: The average of six regions does not necessarily create a representative NH signal. However, our model results support the statement that the average of the well-sampled regions is representative of the hemispheric average. We intend to convey this point more strongly in the revised manuscript.

C: p. 22727 I 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.

R: This is a good point. We propose an alternative methodology for analyzing the regional measurement trends above (see our response to comment on p. 22725 I. 8)

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

R: Natural emissions (isoprene and terpenes) are prescribed with no interannual variability. Lightning emissions are not prescribed, but calculated from the convective frequency. The NO<sub>x</sub> and associated ozone produced from lightning emissions is budgeted as tropospheric ozone. This is clarified in the text.

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

R: Reanalysis. This is clarified.

C: 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.

R: There are two points to make here. In the first place, tagging stratospheric ozone and following its evolution in the troposphere is problematic because the results are sensitive to the tagging methodology in the vicinity of the tropopause, the location where the ozone gradient is largest. Calculating the stratospheric ozone as the difference between ozone and tagged tropospheric ozone does not have this problem. When we tag a stratospheric tracer of ozone (but with tropospheric ozone losses within the troposphere) we find large differences with the methodology used in this paper (see Hess and Lamarque, 2007). Secondly, the Synoz tracer is designed to

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get the stratospheric-tropospheric exchange correctly, but not to simulate the evolution of stratospheric ozone. The Synoz tracer has a simple tropospheric loss, whereas the loss of our tagged ozone is consistent with the simulated chemistry. With its simple source distribution and lack of chemistry Synoz looks rather different from stratospheric ozone.

C: 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.

R: The reviewer is correct that our statement "equivalence to switching off" is probably not terribly relevant for this paper. We will remove this statement, as it is extraneous to the main point. The tagged methodology is linear in the sense that if we tag individual NO<sub>x</sub> emissions the resulting sum of the tagged ozone for each of the individual emission sources is equal to tagged ozone obtained by tagging all of the sources. References to other tagging methodologies will be included in the revised paper.

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

R: Thank you. This is a good suggestion.

C: 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?

R: Please refer to the detailed description of using the Synoz flux method at the beginning of this review. Simply prescribing stratospheric ozone often results in too much STE. However, the original Synoz flux method does not allow for sufficient interannual

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variability.

C: 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?

R: The transport and chemistry operators in the model are conservative. However, we have no independent way to assess the net stratospheric input of ozone except as a residual term so we cannot check if the ozone budget is closed. Similarly we cannot independently test the budget of ozone production produced from colored NO<sub>x</sub>. The budgets will be different: ozone from produced from colored NO<sub>x</sub> is lost due to transport to the stratosphere, while the stratosphere represents a source of untagged ozone. Initializing the model with identical concentrations of NO<sub>x</sub> and colored NO<sub>x</sub>, ozone and colored ozone does provide a good test. We find that the evolution of the two distributions is almost identical initially, especially near the surface where the impact of the stratosphere is small.

C: 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.

R: In fact we believe it is more likely we underestimate the trend at least in the 1990s because we do not capture the ozone destruction from Pinatubo. For example in Figure 7 we do not capture the very low ozone values in the beginning of the 1990's at 500 hPa. The low measured values at 500 hPa are well correlated with the low measured ozone at 150 hPa and thus very likely due to Pinatubo. However, it is true we cannot capture the ozone trends due to trends in stratospheric chemistry; only due to changes

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in the circulation. Voulgarakis et al. (2011) shows the changes in ozone in 1999 are likely due to changes in circulation, not changes in stratospheric ozone amount.

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

R: Thank you. We will fix the language.

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

R: No – NOx emissions from lightning contribute to our tropospheric ozone component and are not attributed to the 'stratospheric component'.

C: p. 22730 l. 8 communicated=>propagated.

R: Thank you.

C: 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).

R: Thank you for this suggestion.

C: 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?

R: The correlations given here are between the simulated ozone averaged over the measurement sites and the simulated ozone averaged north of 30o N. Here we are examining the extent to which the measurement sites are representative of the large-scale average using the model simulation. We will make this clearer in the revised paper and likely include these type of correlations in a table.

C: 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-

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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?

R: It is a good suggestion to move this section to the appendix. Thank you. By using Synoz we are simply testing the sensitivity of the STE to transport processes. The timescales in Figure 2 are determined by the time for stratospheric ozone to adjust in the troposphere. Thus it should adjust on a tropospheric photochemical timescale.

C: 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.

R: See above for a better description of the Synoz parameterization. Indeed Synoz concentrations are prescribed in the stratosphere. However, it takes a number of years for the Synoz concentrations to reach an equilibrium concentration: the equilibrium is established between the concentrations specified in the stratospheric source region and the loss of Synoz in the troposphere. For example, if the concentrations were initially just prescribed in the stratospheric source region and set to zero elsewhere then stratospheric to tropospheric exchange would increase until equilibrium is established. We used Synoz initial conditions from a well spun-up simulation, so a spinup to the equilibrium was not necessary. However, to double check this we reported on a number of additional simulations (Figure 2) that clearly show there is no spinup problem.

C: 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.

R: Thank you. These timeseries correlated are the smoothed monthly residual time-series. We assumed if we had measurements for 10 out of 12 months then we could calculate an annual value for ozone.

C: p 22732 l.20 ....unlikely ... not correlated ... akward sentence, do you mean some-

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thing likely to be correlated? Japan and surrounding are known to have significant contributionsof stratospheric intrusions- which indeed maybe very different between 42 and 32 N.

R: Thank you. This has been clarified in the new text. We meant that the lack of correlation cannot be explained by the fact that at some sites (e.g., the Southern most ones) the 150 hPa level is sometimes in the troposphere. The lack of correlation at 500 hPa between these sites may very well be explained by their latitudinal differences. However, here we are discussing the lack of correlation at 150 hPa.

C: 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.

R: Thank you. We will.

C: p. 22734 l. 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.

R: Figure 3 shows the regionally averaged signal for different regions at 150 and 500 hPa (e.g., Canada, Northern Europe). Figure 4 shows the contributions of individual stations (e.g., Alert, Edmonton, Resolute etc) to the regionally averaged signal at 500 hPa. We will change the order of these figures in the revised text and clarify this explanation.

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

R: By most accounts the MOZAIC measurements are more accurate than those of the ozonesondes.

C: p. 22736 l. 18 Convective overshoots followed by adjustment?

R: Yes, due to penetrative convection into the lower stratosphere followed direct mixing

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with tropopause air and subsidence.

C: p. 22737 | 7 greater than.

R: Thank you.

C: p. 22737 | 10 the name is Sonnblick R: Thank you.

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

R: Thank you. Although as suggested we will likely eliminate this section in the revised text and move the important points to the paper inclusions.

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

R: As suggested we will move this section into the conclusions.

C: 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?

R: We have included the Pinatubo period in calculating the trends but not in calculating the correlations. This needs to be explained better. We will also check the importance of including (not including) the Pinatubo period in the measurements.

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

R: Thank you. We will clarify in the revised text.

C: 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

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consistency of model and measurements!

R: In Hess and Lamarque (2007) we explicitly compare this methodology with other studies. We show the methodology using tagged photochemical ozone results in a much lower stratospheric influence at the surface than tagging stratospheric ozone itself. It results in considerably less stratospheric influence at the surface than reported in Wang et al. [1998] and considerably less than Roelofs and Lelieveld [1997]. We will reference other studies examining this question in more detail.

C: p. 22743 l. 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.

R: Thank you. We will make this point clearer in the revised version.

C: p. 22743 l. 20 I am somewhat worried that the 'derived' stratospheric ozone component includes to some extent a signal of natural emission variability, please comment.

R: The only natural emission variability included is due to lightning. The lightning component is explicitly given as tropospheric as the resulting lightning NO<sub>x</sub> emissions are tagged as colored NO<sub>x</sub>.

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

R: Clarified. We mean the signal obtained by averaging all the robust stratospheric regional measurement signals together.

C: 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

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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.

R: We agree there is a lot of handwaving going on here. We will remove this paragraph, although retain the calculation about the Asian influence at Mace Head. This has not been previously published to our knowledge.

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

R: Thank you. We will do so.

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

R: Indeed. Examining Table S2 all three sets of MOZAIC measurements (Japan, the Eastern US and Europe) are correlated. This good correlation does not extend to the ozonesondes.

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

R: Agreed.

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

R: Thank you. Clarified.

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

R: Thank you.

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