The manuscript “Ship-borne FTIR measurements of CO and O_3 in the Western Pacific from 43°N to 35°S: an evaluation of the sources” is revised according to the referees’ comments. A detailed point-to-point reply to the referees’ comments is given below. In addition, a revised manuscript is attached to this document.

Point-to-point reply

Anonymous Referee #1

<GENERAL COMMENTS>

Overall paper quality is good. It is worth accepting if revisions are made. The ship-borne FTS is providing excellent data set to confirm accuracy of the GEOS-Chem model. Firstly, what is newly found from measurements themselves is not clear. Secondly, relation between observation error and retrieval error of source or source region is not clearly described. Lastly, the authors have to discuss how to improve the measurements in the future.

We thank the referee for the positive general comments.

The manuscript shows a new and unique dataset of atmospheric CO and O_3 obtained during a ship campaign in the Western Pacific. The manuscript presents a very well reproducibility of the trace gas distributions by the GEOS-Chem model and it provides a detailed assessment of the source processes and source regions of Western Pacific CO and O_3 pollution. The analysis reveals that global scale transport of pollution from various different source processes and source regions cause several distinct plumes of elevated CO and O_3 contaminating large parts of the Western Pacific.

A statement to the observation error and the retrieval error of the sources is given in the “specific comments” part below. A possibility to improve measurements in the future is now discussed in the manuscript (see specific comments below).

<SPECIFIC COMMENTS>

1. (P22952) Abstract: (Lines 12, 21): Definition of “sources” and source “regions” is not clear. (P22952) Abstract: (Lines 14-20) How to distinguish between CO sources from biomass burning and from fossil fuel?

We agree with the referee in this aspect. The term “source” is meant to describe source processes, such as biomass burning and fossil fuel combustion, whereas the term “source regions” is meant to describe the location of the source, such as Asia and Africa. In order to clarify this aspect, we suggest to clearly distinguishing between source processes and source regions throughout the entire manuscript.

P22952, line 12: “…between different source processes and source regions.”
P22952, line 12: “The source regions are verified with HYSPLIT…”
P22952, line 21: “The source regions of the measured O_3 pollution…”
P22954, line 3: “… evaluation of the source processes and source regions”
P22958, line 9: “The tagged-CO simulation calculates source processes of CO…”
P22962, line 17: “… source regions of the observed CO and O_3 pollution…”
P22963, line 4: “To identify the contributions of different source processes and source repetitive regions…”
P22964, line 7: “In order to verify the source regions of the measured CO pollution…”
P22966, line 2: “In order to verify the source regions of the measured O_3 pollution…”
P22967, line 11: “This is verified with a subjective assessment of the source processes and source regions…”

2. (P22952) Abstract: (Lines 24-26): “In the Southern Hemisphere the impact of emissions from South America, South-East Africa and Oceania is important”: Expression of “important” is ambiguous. In addition, the areas are almost the same as land area of the Southern Hemisphere.

We replaced the expression “important” and changed the corresponding sentence. We agree that the mentioned areas are almost the same as land areas of the Southern Hemisphere. Those regions are tagged by the GEOS-Chem model and each show significant contributions to Southern Hemispheric O_3 contamination.

P22952, line 24: “In the Southern Hemisphere emissions from South America, South-East Africa, and Oceania significantly contribute to the measured O_3 pollution.”

3. (P22956) (Line 18): “DOF is an expression for the amount of individual layers” The word of “amount” is not clear. Do the authors mean “amount of information?

The DOF is rather an expression for the number of independent layers in the retrieval. We changed the sentence in the revised manuscript accordingly.

P22959, line18: “The degree of freedom (DOF) for signal is an expression for the number of independent layers in the retrieval (Rodgers, 1990).

4. (P22960 Line 9): How do the authors determine the criteria of wind direction of >30 deg and wind speed of 5m/s? Are there any technical documents to refer?

The criteria for the wind filter were empirically determined.

5. (P22963) (Lines 11 - 13): The contribution from regions such as ASFF 40% etc are presented. How accurate is the model estimation? Is the estimation error dependent on the distance of the source regions?

The estimation error of the source contributions in the model depends on many factors, such as emission rates, meteorology, and chemistry processes. Those factors are a major source of uncertainties in the source estimation. Emissions from NMHC, for example, are calculated to be 175-507 Tg/a (Duncan2007) offering only little constraint and potentially introducing large errors into the source estimation. Thus, a statement about the accuracy of the source estimation is very difficult. However, qualitative statements about the accuracy can be made by validating the model against observations as performed in this and other studies.

6. (P22967) (Line 8): Conclusions “Only surface CO concentrations in the mid-latitudes were largely over estimated by the model.” This conclusion is too simple. Brief explanation of the reason has to be described.

We agree with the referee and we now provide a brief explanation for the overestimation of CO surface concentrations by the model in the conclusions of the revised manuscript.

P22967, line 8: “In the beginning of the campaign, in mid latitudes, CO surface concentrations were largely overestimated by the model. Here, the model seems to mix free tropospheric air into the boundary layer too readily.”

7. (P22967) (Line 15): The expression of “The West Pacific” is too general. The northwest Pacific is not so remote and polluted from the Southeast Asia and China.

We agree with the referee and changed this aspect accordingly.

P22967, line 15: “Large parts of the West Pacific belong to Earth’s most remote areas. While much of the air observed in this region is considered the cleanest air world wide…”

8. The authors have to discuss how to improve the measurements in the future.

We agree with the referee and we inserted a new paragraph in the “summary and conclusions” section.

P22968, line 3: “The presented analysis provides a detailed evaluation of the source processes and source regions of Western Pacific carbon monoxide and ozone pollution measured during a ship campaign from Japan to New Zealand in fall 2009. Thus, the analysis is limited on the abovementioned period of time. In order to obtain a better knowledge of Western Pacific air contamination, future measurements in this region with high temporal and spatial resolution are needed as they are currently performed by, for example, the HIAPER Pole to Pole Observation (HIPPO) aircraft campaign \citep{Wosfy2011} and the SHIVA (Stratospheric ozone: Halogen Impacts in a Varying Atmosphere) campaign.”

<TECHNICAL CORRECTIONS>

1. (P22954) (Line 10) “Tropospheric sources of O_3 include the photochemical oxidization of CO, CH4…” The word “include” is not clear.

We agree with the referee and we changed the sentence in the revised manuscript.

P22954, line 10: “The main tropospheric source processes of O_3 are the photochemical oxidation of CO, CH_4, volatile organic compounds (VOC) in the presence of …”

2. (P22955) Section 2 is too short for section. The contents can be included in Section 1
We inserted a new section here, since we think that the description of the field campaign does not belong to the introduction part. We further believe that a separate section makes the manuscript easier to read.

3. (P22959) Lines 10-12: The sentence is not clear. “To account for the characteristics of the FTIR retrieval model profiles Xm…” Is “To account for the characteristics of the FTIR retrieval, model profiles Xm…” correct?
We agree with the referee. We inserted the comma in the revised manuscript.

P22959, line 10: “To account for the characteristics of the FTIR retrieval, model profiles Xm…”

4. (P22960 Line 15) and (p22961; Line 26) “Figure 4c-e show” Figure 5b-e shows” » Figures
We agree with the referee and we changed this aspect in the revised manuscript.

P22960, line 15: “Figures 4c-e show…”
P22961, line 26: “Figures 5b-e show…”

5. (P22985) (Fig A1): Only this figure has X-axis of the height. The Y axis of the vertical profile is clear to see as presented in Fig2.
We agree with the referee. Fig. A1 was changed in the revised manuscript according to the referee’s suggestion.”

Anonymous Referee #2

<GENERAL COMMENTS>

This paper presents a suite of measurements of carbon monoxide and ozone from a ship transit from 43 N to 35 S. Solar absorption Fourier Transform infrared spectrometry measurements of total and partial column amounts are complimented by in-situ FTIR measurements of concentrations of CO and other trace gases at the surface and occasional flask sampling and O_3 sondes. The chemical transport model GEOS-Chem is used in full chemistry mode and in tagged CO and Ox modes to help interpret the measurements and source regions of various pollution events sampled. Fire-maps and back trajectory analysis and HCN total column amounts measured from the ship are also used to help interpret the measured pollution events. The measurements presented here are important because they are from a region of the globe with a great scarcity of atmospheric measurements. The generally good agreement seen between the measurements and the GEOS-Chem model is heartening although a weakness in the model’s ability to get the vertical mixing correct is revealed. It is also evident from the discussion paper that a serious and careful effort has gone into the interpretation of these measurements. For this reason I think that this paper is worthy of publication and is certainly suitable for ACP although I recommend a number of minor revisions.
We thank the referee for the general positive comments. We considered the specific comments in the revised manuscript as far as possible (see “specific comments” part below).

<SPECIFIC COMMENTS>

1. My first specific comment is about the structure of the paper which I found somewhat cumbersome to read. I believe that the paper would be much easier to understand if the results section discussed each pollution event sampled in turn using all the available measurements and modeling to draw its conclusions. The current structure (where each technique is discussed in isolation so that the reader needs to be reminded about what the other evidence was) is not conducive to getting the main points across. However – I realize that to re-structure the paper might involve a great deal of work – so I recommend that the authors consider this point carefully and make up their own minds as to whether this change is worthwhile.

We considered this point carefully. The structure of the paper was chosen in analogy to many other studies, such as Velazco2005 (JGR, "Latitude and altitude variability of carbon monoxide in the Atlantic detected from ship-borne Fourier transform spectrometry, model, and satellite data"), Jaeglé2003 (JGR), and Staudt2001(JGR). We first show all obtained measurements in comparison to the GEOS-Chem model. In that way the reproducibility of the model can be evaluated and the variability of the measurements is apparent. Then, the source processes and source regions of the measured variability were evaluated using different methods and techniques one after the other. Thus, we find the structure in the manuscript more intuitive than discussing each pollution event individually.

2. In section 4.2 page 22962 lines 5-7 it is stated that the pollution event PE1 is captured by solar absorption FTIR measurements of O_3 and yet in the previous section 4.1 page 22960 lines 25-27 it is stated that solar absorption FTIR measurements were not possible because of bad measurement conditions as a result of typhoon Melor. These seem to be contradictory statements.

We agree with the referee that the statements might be confusing. The weather conditions during the ship campaign partly allowed to measure not more than a few spectra during the day. In particular, in the beginning of the ship campaign the weather allowed to measure not more than one to two good spectra during the day. For those spectra the spectral region from 700cm^{-1} to 1350cm^{-1} was preferred against other spectral regions. The preferred spectral region covers the ozone retrieval window from 1000cm^{-1} to 1005cm^{-1} but not the CO retrieval windows close to 2100cm^{-1}. Thus, the coverage is different for trace gases from different spectral regions. For clarification we suggest to insert a footnote stating this problem.

P22962, line 7: "PE1 within cruise section S1 is this time also captured by the solar absorption FTIR spectrometer \footnote{The weather conditions in the beginning of the ship campaign partly allowed to measure not more than one to two spectra during the day. For those spectra the spectral region from 700cm^{-1} to 1350cm^{-1} was preferred against other spectral regions. This spectral region includes the O_3 microwindow but not the CO microwindows} and the O_3-sondes.

3. Indeed – the coverage of O_3, CO, and HCN measurements shown in figures 4, 5 and 8 are different and the reasons for this are not explained.

See comment 2 (CO, O_3, and HCN were retrieved in different spectral regions. The coverage of the spectra including those spectral regions differs.)

4. The profiles and the column amounts do not seem to line up properly in Figure 4b and 4c and 5a and 5b. Is this just a formatting problem? The column amount of CO at around 40N has no corresponding profile – why is this?

We agree with the referee that the column amounts do not line up properly in Figure 4b, 4c, 5a, and 5b. This is a formatting problem, which we were not aware of so far. This is now corrected in the revised manuscript.

The column amount of CO at around 40°N has no corresponding profile since this spectrum was measured with a reduced resolution of 0.01cm^{-1}. The profile retrieval of such a spectrum is not reliable. The profile was therefore rejected.

5. Again – the solar absorption FTIR measurements of CO in figure 4b plot are shown with error bars but no explanation is given as to what these are – or why these are the only measurements for which error bars are given. Are they a standard deviation of a number of measurements or a theoretical uncertainty of the retrieval of a single measurement?

The error in the solar absorption FTIR total column amount of CO is the uncertainty of the retrieval of a single measurement. This aspect is now stated in the manuscript.

The error bars of the in-situ measurements are not shown since the errors of these measurements are small (Flasks: 0.3%, FTIR in-situ: 0.4%) and would not be visible in the corresponding plots.

The error of FTIR partial columns is still a matter of debate (and is not characterized by the retrieval software); Partial columns are calculated from the volume mixing ratios of the individual layers, which are each characterized by an error. However, the partial column error is not only the sum of the errors of these individual layers, because the layers are not independent. A discussion on that error would exceed the scope of this manuscript. For this reason the error of FTIR partial columns is not given, analogous to other studies (see Schneider 2008).

Additional information: the error in the total columns is about 2.5%. Schneider2008 found out that the error in FTIR tropospheric columns is about 5.4% by comparing to ozone sonde measurements).

P22979, Fig. 4, caption: "(blue markers, the error bars represent the theoretical uncertainty in the FTIR retrieval)."

6. Is the GEOS-Chem modeled tropopause height used to determine the partial column used for tropospheric O_3 from solar absorption FTIR measurements – or is there some other definition used?
The GEOS-Chem tropopause height is used to determine the partial tropospheric O_3 columns from solar absorption FTIR measurements. This is stated in the manuscript on Page 22959, line 27: "For the calculation of the FTIR tropospheric columns the model tropopause height is used. "

7. The agreement between sonde, FTIR and modeled tropospheric O_3 is remarkably good. I recommend that the authors stress this point and maybe include it in the conclusions.
We agree with the referee and we now describe the model-observation comparison in more detail in the "Summary and Conclusion" section and stress the point that the GEOS-Chem model outstandingly reproduces O_3 concentrations. Thereby, we also consider comment 6 from referee #1.

P22967, line 8: replace sentence "Only surface CO concentrations in the mid latitudes were largely overestimated by the model." by "The carbon monoxide variability is clearly expressed by the model, however, absolute values are underestimated due to a constant low bias in the simulation already known from previous studies. In the beginning of the ship campaign, in mid latitudes, CO surface concentrations were largely overestimated by the model. Here, the model seems to mix free tropospheric air into the boundary layer too readily. Ozone concentrations were outstandingly reproduced by the GEOS-Chem model during the entire ship campaign both in magnitude and variability."

8. In my opinion the last section of the conclusions needs to be rephrased as it seems to extrapolate from the four individual pollution events sampled to all pollution experienced in these areas. Maybe express as "in the pollution events sampled…"
We agree with the referee and we rephrased the last two sections of the conclusions.

P22967, line 21: "The analysis of several different carbon monoxide and ozone pollution events sampled during the RV Sonne ship campaign has demonstrated that in the Northern Hemisphere …"

P22967, line 24: "In the Southern Hemisphere biomass burning and fossil fuel combustion are determined to cause carbon monoxide and ozone pollution observed during the RV Sonne ship campaign. In particular, African and South American…"

<TECHNICAL CORRECTIONS>

1. Section 5 line 23 replace "measured" with "modeled"
We changed this aspect in the revised manuscript.

P22962, line 23: replaced "measured" by "modeled".

2. Suggest replace heading "conclusions" with "Summary and Conclusions"
We changed this aspect in the revised manuscript.

P22967, header: replaced "Conclusions" by "Summary and Conclusions"

3. Line 6 Abstract, suggest "results obtained" rather than "obtained results"
We changed this aspect in the revised manuscript.

P22952, line 6: "The results obtained are compared to the GEOS-Chem …"

4. Line 11 Abstract, suggest "make it possible to" rather than "allow to"
We changed this aspect in the revised manuscript.

P22952, line 11: "Tagged-CO simulations implemented in the GEOS-Chem model make it possible to differentiate between different sources and source regions."

5. Section 5.1, line 27, replace "origins" with "originates"

We changed this aspect in the revised manuscript.

P22963, line 27: "PE4 pollution mainly originates from …"

6. Conclusions, line 19-20 insert "with contributions" between "pollutants" and "from" at end line 19 and delete the word "partly" from line 20.
We have changed both aspects in the revised manuscript.

P22967, lines 19-20: "… the cause of this pollution is global scale transport of pollutants with contributions from source regions as far away as Europe."

Anonymous Referee #3

<GENERAL COMMENTS>

This paper presents a series of new measurements of O_3 and CO in the Western Pacific, one of the most remote areas on the planet thought to have the cleanest air. This area has impact on stratospheric composition because it lies below the preferred location for troposphere-to-stratosphere transport and hence has a global environmental impact. The measurements reveal the presence of pollution originated in sources far away from the location of the measurements. The authors use a series of modeling tools both Eulerian (GEOS-CHEM) and Lagrangian (HYSPLIT) to analyze the data and reveal the most likely origin of the pollution events. Agreement between models and measurements is fairly good and the interpretations are consistent. Although relevant for being among the first measurements in this region, the writing suggests perhaps too much generality. It may be adequate to underline the point-wise, episodic nature of the measurements and recommend actions for more robust statistical studies, maybe suggesting measures to improve the measurements (location, season, techniques, etc.) The paper would be more self- contained if some additional phrases containing key information (both for specialists and non-specialists) would be added. Specific comments point out some examples. The study is sound and interesting and therefore I recommend it for publication in ACP after the authors have addressed a few mostly minor remarks.
We thank the referee for the positive comments.
We now underline the point-wise nature of the measurements and we state a possibility to improve measurements in the future (see statement 8, referee #1). In addition, we now provide additional phrases containing more key information (see specific comments)

<SPECIFIC COMMENTS>

1. P 22952 L 24-25. Abstract: ARE significant, IS important?
We checked the grammar in both sentences.

2. P 22954 L 1. 'In the Southern… is analyzed.' This sentence is a bit awkward. It could be rewritten.
We agree with the referee and we rewrote the sentence.

P22954, line 1: "In the Southern Hemisphere (SH) biomass burning emissions and its global distribution are analyzed (Andreae et al., 2001; Thompson et al., 2001)."

3. P 22956 L 2-6. Not clear, radiosondes where launch and used together with NCEP winds as priors? The parenthesis is too long.
A priori pressure and temperature profiles were obtained from the NCEP database. Radiosonde observations were delivered to the WMO database to be assimilated into NCEP. This aspect is now clarified in the manuscript.

P22956, line 2-6: "A priori pressure and temperature profiles were obtained from the NCEP (National Center for Environmental Prediction, http://www.ncep.noaa.gov/) database. Meteorological observations from radiosondes, which were launched for times per day during cruise section S1, were delivered to the World Meteorological Organization (WMO) global database to be assimilated into the NCEP database."

4. P 22956 L 20. A brief sentence explaining the meaning of 3 or 5 DOF would be useful for a self contained explanation.

The explanation for the degree of freedom is given on P22956, lines 18-19: "The degree of freedom (DOF) for signal is an expression for the number of independent layers in the retrieval" (see also comment 3, referee #1).

5. P 22957 L 18. These where the radiosonde data where used as priors?

See comment 3.

6. P 22958 L 19. Does the word 'results' indicate model output?

Yes. The sentence was changed accordingly.

P22958, line 18: "Model output was generated for ozone production…"

7. P 22959 L 26. 'General' means 'standard' or a new general method is being proposed to handle the problem?

We assume that the referee refers to P22959 L 15. "Standard" was meant. Thus, we changed "general" to "standard" in the revised manuscript.

P22959, line 15: "The standard method for handling this problem is…"

8. P 22959 L 27. It would be clarifying to explicit the definition of the tropopause used both with model and measurements (i.e WMO?), also for the following references to tropopause.

We agree with the referee and we now give the definition of the tropopause (WMO) in a footnote.

P22959, line 27: "… above the tropopause\footnote{The tropopause is defined as the lowest level at which the temperature lapse rate decreases to 2 K km^{-1} or less, and the lapse rate averaged between this level and any level within the next 2 km does not exceed 2 K km^{-1} (WMO definition).}.

9. P 22960 L 19 and Figure: Right hand CO side appears remarkably accurate. A comment here on the cause of the disagreement on the left hand side would be useful for the reader.

The cause of the disagreement on the left hand side is discussed in the manuscript further below (P22963, line 18). However, we agree that a comment referring to that explanation would be useful for the reader.

P22960, line 12: "The cause of the overestimation in the beginning of cruise section S1 is discussed in Sect. 5.1."

10. P 22961 L 5. Is the Townsville underestimation problem related to the problem describing P1? What is the reason of overestimation of P1 by GEOS-CHEM?

In our opinion the overestimation of P1 is due to a wrong representation of the boundary layer in the model; the model mixes free tropospheric air into the boundary layer too readily in this meteorological situation (see comment 7, referee #2).

We believe that the Townsville underestimation is rather due to an underestimation of the emissions.

11. P 22961 L 14. It is not clear from the picture that PE3 reaches higher than 16 km.

We do not agree with the referee. We think that the enhancement (yellow color) reaches the tropopause height between 11/08 and 15/08. There, the tropopause is in 17 km altitude.

12. P 22961 L 18. PE4 seems to have a TTL component as well, not just near surface.

In our opinion, PE4 in Fig. 5c has a near surface and a mid tropospheric component (stated in the manuscript). The model (Fig. 5e) shows a near surface component only.

13. Figure 5, Panel (a). Is GEOSCHEM green line column average?

The green line in Fig. 5a represent the tropospheric columns of CO.

14. P 22962 L16. Wording: maybe "hypothesize" would be better than "assume".

We agree with the referee. This aspect has been corrected in the revised manuscript.

P22962, line 16: "…one can hypothesize that the sources of the observed CO and O3 pollution are related.

15. P 22963 L 16. Fix citation Jaegle/Bay
The citation is fixed in the revised manuscript.

16. P 22962 L 18. Do PBL O3 measurements suggest clear PBL air consistent with low CO?
PBL O_3 measurements (ozonesondes) show O_3 concentrations of about 40-50ppb and are, thus, quite consistent with low PBL CO in this region.

17. It the only problem that model excessively mixes FT and PBL air? Nothing related to emissions/ emission inventories?
In our opinion, the model shows the impact of a strong carbon monoxide source in the beginning of the ship campaign. Since the surface measurements do not show any hint to such a source, we concluded that the overestimation is due to a wrong expression of the wind fields transporting polluted air to this location. Based on the fact that the free troposphere is highly polluted we concluded that the model mixes free tropospheric air into the boundary layer too readily causing the enhancement in surface CO.

18. P 22963 L 17. wheras? -> whereas?
We agree with the referee. This is now corrected in the revised manuscript.

P22963, line 17: "…, whereas the FTIR…"

19. P 22963 L 18. suggests a problem?
We agree with the referee. We corrected the misspelling.

P22963, line 18: "This suggests a problem with the representation…"

20. P 22963 L 24 is the peak of VOC seen in Geos-CHEM relevant to PE2?
Yes. In fact, VOC emissions are relevant to PE2, PE3, and PE4. This is mentioned in the manuscript on P22964, line 5: "Oxidation of volatile organic compounds (VOC) is also responsible for part of the CO variability in the Southern Hemisphere."

21. P 22963 27 origins -> originates?
This has been corrected in the revised manuscript.

P22963, line 27: "PE4 pollution mainly originates from biomass burning..."

22. P 22964 L 10. Some more information about the parameters used for trajectory calculations would be helpful. Does "single" mean one trajectory per event, i.e. 4 trajectories in total? Accuracy of single trajectories over 315 hours is hardly robust. Ensembles are better. What is the size of the ensemble? What are the distribution of the initial locations in space (i.e. column releases?) and time (interval or single instant release?) Which winds and resolutions are the trajectories running on?
We agree with the referee and we now provide more information about the parameters used for the trajectory calculations. We agree that single trajectories are hardly robust. For this reason we used 120h ensembles and showed just one 315h single trajectory for each event, in addition.

P22964, line 7: "In order to verify the source regions of the measured CO pollution, backward trajectories were calculated with the NOAA HYSPLIT (National Oceanic and Atmospheric Administration Hybrid Single Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 1997, 1998) at the four characteristic points PE1, PE2, PE3, and PE4 (Fig. 7). For the calculations the NCEP-GDAS meteorological assimilation with a 1°x1° grid resolution was used. One single backward trajectory (max. 315 h) embedded by ensemble backward trajectories (120 h) are presented for each pollution event. The trajectory ensemble starts multiple trajectories (in total 27) around the starting location. Each member of the trajectory ensemble is calculated by offsetting the meteorological data by a fixed grid factor (one grid meteorological grid point in the horizontal and 0.01 sigma units in the vertical). The altitude of the backward trajectories is expressed by the adjacent color bar. In addition, MODIS (MOderate Resolution Imaging Spectroradiometer) firemaps are shown in Fig. 7 for the 5 day period prior to the corresponding trajectories in order to assess the influence of biomass burning.

23. Section 5.3: Is the main conclusion from this section that HCN supports the hypothesis that PE3 is mainly fossil fuel and PE 1, 2 and 3 are biomass burning originated?
The main conclusion from section 5.3 is that HCN support the hypothesis that PE1 and PE3 are mainly fossil fuel originated and PE2 and PE4 are biomass burning emissions affected.

24. P 22966 L 18. Only originates from Oceanian emissions or from Indonesia emissions?
In the tagged-tracer simulations the source region “Oceania” includes Indonesia.

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Ship-borne FTIR measurements of CO and O_3 in the Western Pacific from 43° N to 35° S: an evaluation of the sources

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Abstract. Carbon monoxide (CO) and ozone (O_3) have been measured in the Western Pacific (43° N to 35° S) during a ship campaign with Research Vessel Sonne in fall 2009. Observations have been performed using ship-based solar absorption Fourier Transform infrared spectrometry, flask sampling, balloon sounding, and in-situ Fourier Transform infrared analysis. The results obtained are compared to the GEOS-Chem global 3-D chemistry transport model for atmospheric composition. In general, a very good agreement is found between the GEOS-Chem model and all instruments. The CO and O_3 distributions show a comparable variability suggesting an impact from the same source regions.

Tagged-CO simulations implemented in the GEOS-Chem model make it possible to differentiate between different source processes and source regions. The source regions are verified with HYSPLIT backward trajectory calculations. In the Northern Hemisphere fossil fuel combustion in Asia is the dominant source. European and North American fossil fuel combustion also contribute to Northern Hemispheric CO pollution. In the Southern Hemisphere contributions from biomass burning and fossil fuel combustion are dominant; African biomass burning has a significant impact on Western Pacific CO pollution. Furthermore, in the tropical Western Pacific enhanced upper tropospheric CO within the tropical tropopause layer mainly originates from Indonesian fossil fuel combustion and can be transported into the stratosphere.

The source regions of the measured O_3 pollution are simulated with a tagged-O_3 simulation implemented in the GEOS-Chem model. Similar source regions compared to the tagged-CO simulations are identified by the model. In the Northern Hemisphere contributions from Asia, Europe, and North America are significant. In the Southern Hemisphere emissions from South America, South-East Africa, and Oceania significantly contribute to the measured O_3 pollution.

1 Introduction

Measurements of the global distribution of trace gases are indispensable to understand the dynamical and chemical processes in the atmosphere. Ground-based solar absorption Fourier Transform infrared (FTIR) spectrometry is a well established remote sensing technique for the observation of atmospheric trace gases (Rao, 1992; Notholt et al., 2003). Two global ground-based solar absorption FTIR networks (TCCON[1], NDACC[2]) are well established (Wunch et al., 2011; Kurylo, 1991). Besides observations from land-based FTIR stations, measurements have been performed in the Atlantic aboard Research Vessel (RV) Polarstern (Notholt et al., 2000; Velazco et al., 2005; Warneke et al., 2006). However, solar absorption FTIR measurements in the Pacific have not been previously performed. Here, we present the first solar absorption FTIR measurements in the Western Pacific complemented with in-situ observations.

The tropical Western Pacific is considered to be the main region of troposphere to stratosphere transport (Holton et al., 1995; Fueglistaler et al., 2004, 2009). In the tropical warm pool (Yan et al., 1992) upwelling processes are enforced by high sea surface temperature and high air humidity. Thus, in this region tropospheric air has a relatively high probability of reaching the stratosphere (Notholt et al., 2005). The entry of tropospheric air into the stratosphere has a strong impact on stratospheric composition. However, the significance of various transport processes for the transport of different species is not yet fully understood.

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[1] Total Carbon Column Observing Network (http://www.tccon.caltech.edu/)

[2] Network for the Detection of Atmospheric Composition Change (http://www.ndsc.ncep.noaa.gov/)

The Western Pacific is of great interest with respect to trans-Pacific air mass transport. In the Northern Hemisphere (NH) the influence of the increasing anthropogenic pollution due to the fast economic growth in Asia and its impact on Northern Hemispheric air quality is studied (Zhang et al., 2008; Jacob et al., 2003). In the Southern Hemisphere (SH) biomass burning emissions and its global distribution are analyzed (Andreae et al., 2001; Thompson et al., 2001).

In the present study we concentrate on the evaluation of the source processes and source regions of carbon monoxide (CO) and ozone (O_3) in the Western Pacific during a ship campaign with RV Sonne in fall 2009. CO is produced during fossil fuel combustion, biomass burning, and oxidation of methane (CH_4) and other non-methane hydrocarbons (NMHC). The major sink of CO is the oxidation by OH. The lifetime of CO ranges from weeks to several months. According to the 2007 IPCC (Intergovernmental Panel on Climate Change) report (Solomon et al., 2007) O_3 is the fourth most important contributor to global warming. The main tropospheric source processes of O_3 are the photochemical oxidation of CO, CH_4, and volatile organic compounds (VOC) in the presence of nitrogen oxides (NO_x) and hydrogen oxides (HO_x). The main sink of O_3 is photochemical degradation forming OH.

In this study measurements of CO and O_3 in the Western Pacific (43° N to 35° S) are presented. Since observations of CO and O_3 are rare in the Western Pacific, these measurements aim to contribute to a better understanding of the composition of the Western Pacific atmosphere. The observations are compared to model simulations which test the current understanding of Western Pacific composition. Furthermore, this study provides an evaluation of the source processes and source regions of Western Pacific CO and O_3 pollution to improve the knowledge of the contribution of these sources.

In Sect. 2 the underlying ship campaign is introduced. In Sect. 3 the instrumental setup and the model are described. This includes remote sensing and in-situ observations. In Sect. 4 results from the ship campaign and a model comparison are presented and discussed. Sources of CO and O_3 pollution are analyzed in Sect. 5.

2 Ship campaign

Measurements of CO and O_3 concentrations in the Western Pacific have been performed aboard RV Sonne in fall 2009. The campaign was performed as a North-to-South transit starting in Tomakomai, Japan, on 9 October 2009 and ending in Auckland, New Zealand, on 5 December 2009. An overview of the campaign is shown in Fig. 1. For a better visualization and interpretation of the dataset the transit was split into four sections according to Table 1. Section S1 was part of the TransBrom (very short lived bromine compounds in the ocean and their transport pathways into the stratosphere) campaign, while cruise sections S2–S4 were part of the SO-203 campaign. Both campaigns were organized by the Leibniz Institute of Marine Sciences at the University of Kiel (IFM-Geomar), Kiel, Germany. For a further description of the campaigns the reader is referred to http://www.rf-bremen.de/ and Quack and Krüger (2010).

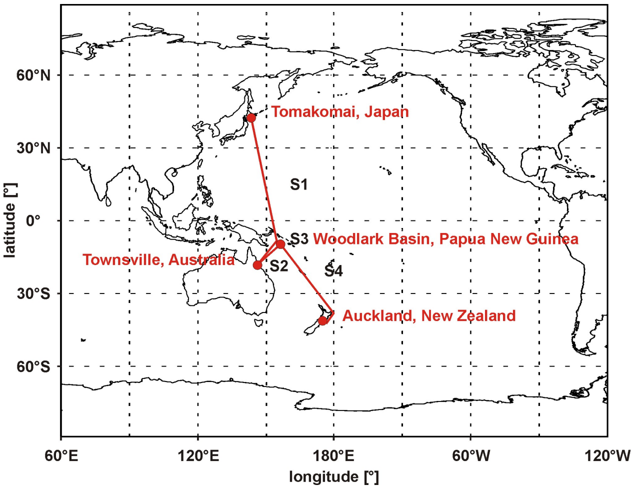

Fig. 1. Overview of the ship campaign with RV Sonne in the Western Pacific between 9 October 2009 and 5 December 2009 from Tomakomai, Japan, to Auckland, New Zealand, divided into four sections (S1–S4) according to Table 1. The red line indicates the approximate ship track.

3 Method

3.1 Solar absorption FTIR spectrometry

Solar absorption Fourier Transform infrared (FTIR) spectrometry was performed aboard RV Sonne using a Bruker IFS 120M spectrometer upgraded with new electronics equivalent to the newer Bruker IFS 125M instrument. The experimental setup for ship-borne measurements is described in Notholt et al. (2000). Solar absorption FTIR spectra were recorded in the spectral region between $700\,cm^{-1}$ and $15\,780\,cm^{-1}$ with a maximum resolution of $0.005\,cm^{-1}$. The meteorological conditions were permanently monitored to minimize the influence of clouds on solar absorption FTIR spectra. Small unavoidable intensity fluctuations caused by thin clouds were corrected using the method described by Ridder et al. (2011).

The retrieval of trace gas concentrations from solar absorption FTIR spectra was performed using the SFIT-2 (Spectral Least Squares Fitting) software developed at the NASA Langley Research Center and the National Institute for Water and Atmospheric Research in New Zealand (Rinsland et al., 1998). A priori pressure and temperature profiles were obtained from the NCEP (National Center for Environmental Prediction, http://www.ncep.noaa.gov/)

Table 1. Description of the four sections (S1–S4) of the ship campaign with RV Sonne according to Fig. 1.

Section	Location	Latitude	Date
S1	Tomakomai, Japan – Townsville, Australia	43° N–20° S	9 Oct 2009–24 Oct 2009
S2	Townsville, Australia – Woodlark Basin, Papua New Guinea	20° S–10° S	26 Oct 2009–30 Oct 2009
S3	Woodlark Basin, Papua New Guinea	10° S	30 Oct 2009–28 Nov 2009
S4	Woodlark Basin, Papua New Guinea – Auckland, New Zealand	10° S–35° S	28 Nov 2009–5 Dec 2009

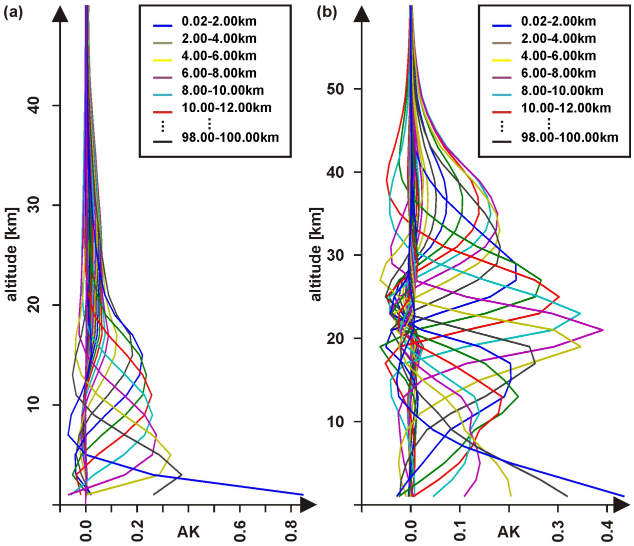

Fig. 2. Representative averaging kernels of the FTIR **(a)** CO retrieval and **(b)** O_3 retrieval. The colored lines indicate the sensitivity and resolution of each individual layer.

database. Meteorological observations from radiosondes, which were launched four times per day during cruise section S1, were delivered to the World Meteorological Organization (WMO) global database to be assimilated into the NCEP database. Spectral line parameters were taken from the HITRAN2004 (high-resolution transmission molecular absorption) database including the 2006 updates for H_2O. A priori profiles were kept constant for the whole campaign and were extracted from the WACCAM model (Garcia et al., 2007) at the location of the NDACC and TCCON station in Wollongong, Australia (34° S, 151° E). CO was retrieved simultaneously in three microwindows (2057.70 cm^{-1}–2057.91 cm^{-1}, 2069.55 cm^{-1}–2069.72 cm^{-1}, 2157.40 cm^{-1}–2159.20 cm^{-1}) including the interfering gases H_2O, N_2O, and O_3. O_3 was retrieved in a single microwindow (1000 cm^{-1}–1005 cm^{-1}) with a simultaneous fit of H_2O.

The sensitivity of the retrieval is expressed by the averaging kernels (AK) showed in Fig. 2. The height of each individual averaging kernel is an indicator for the sensitivity of the corresponding layer, while the width is an indicator for the vertical resolution (Rodgers, 2000). The degree of freedom (DOF) for signal is an expression for the number of independent layers in the retrieval (Rodgers, 1990). CO is mainly sensitive in the troposphere with a DOF between 3.0 and 4.0. O_3 is sensitive in the troposphere and stratosphere with a DOF of around 5.0.

3.2 In situ data

3.2.1 FTIR analyzer

Surface concentrations of CO were continuously measured aboard RV Sonne with a FTIR in-situ trace gas analyzer (Griffith et al., 2010; Esler et al., 2000) developed and constructed by the Centre for Atmospheric Chemistry at the University of Wollongong. The system uses a low-resolution FTIR spectrometer ($\mathrm{res} = 1\,\mathrm{cm}^{-1}$) to quantify the volume mixing ratios (vmr) of CO, CH_4, CO_2, N_2O, and $\delta^{13}C$ in CO_2. The FTIR spectrometer measures the transmission spectrum of an air sample within a multipass White cell (White, 1942). The transmission spectrum is then analyzed with the Multiple Atmospheric Layer Transmission (MALT) nonlinear least squares-fitting software (Griffith, 1996, 2002). A detailed description of the instrument can be found in Griffith et al. (2010).

3.2.2 Flasks

Approximately 30 flasks were taken aboard RV Sonne and were analyzed by the Max Planck Institute for Biogeochemistry in Jena, Germany. Air samples were dried during collection using magnesium perchlorate and compressed to 2 bar absolute yielding a sample size of 2 l. Concentrations of CO, CO_2, CH_4, N_2O, SF_6, $\delta^{13}C$ in CO_2, and $\delta^{18}O$ in CO_2 were determined by gas chromatography and mass spectrometry. For measurement accuracy the reader is referred to http://www.bgc.mpg.de/service/iso_gas_lab/gas_lab/techniques_results/index.shtml and to http://www.bgc.mpg.de/service/iso_gas_lab/techniques_results/irm_eams.shtml.

3.2.3 Balloon sondes

Daily O_3-sondes (Science Pump Corporation ECC-6A) were launched during cruise section S1. Meteorological condi-

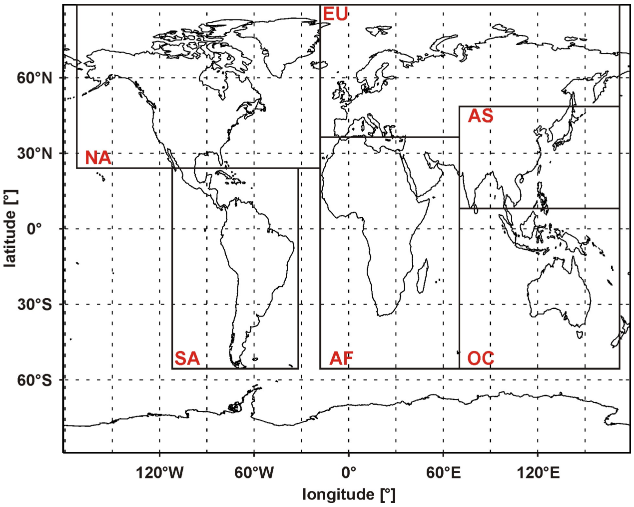

Fig. 3. Tropospheric source regions of the GEOS-Chem tagged-CO simulation. NA: North America, EU: Europe, AS: Asia, SA: South America, AF: Africa, and OC: Oceania.

tions obtained from radiosonde observations during S1 were delivered to the WMO global database.

3.3 GEOS-Chem

CO and O_3 concentrations in the Western Pacific were simulated using version v8-02-04 of the global 3-D chemistry transport model GEOS-Chem for atmospheric composition. The model is described in Bey et al. (2001a). GEOS-Chem is driven by GEOS-5 (Rienecker et al., 2008) assimilated meteorological observations from the NASA Global Modeling and Assimilation Office (GMAO). Model profiles were sampled along the ship track every three hours with a horizontal resolution of $2^\circ \times 2.5^\circ$ and a vertical resolution of 47 layers using the GEOS-Chem full chemistry simulation. Model results for carbon monoxide (CO) are scaled by a factor $\beta = 1.2$ to balance a consistent low bias in the GEOS-Chem simulation (Kopacz et al., 2010).

In order to identify CO sources from different source regions the tagged-CO simulation (Duncan et al., 2007) implemented in the GEOS-Chem model is applied using monthly mean OH concentrations from a previous full-chemistry run as the primary CO sink. The tagged-CO simulation calculates source processes of CO such as fossil fuel combustions, biomass burning, biofuel, oxidation of CH_4, and volatile organic compounds (VOC) from various source regions. An overview of the standard source regions within the tagged-CO simulation is shown in Fig. 3. Similar approaches have been previously used by Bey et al. (2001b); Staudt et al. (2001).

In order to identify the source regions of the measured O_3 pollution, the tagged-O_x simulation implemented in the GEOS-Chem model has been used. The tagged-O_x simulation calculates the contributions of the trace regions in Fig. 3 using daily production rates and loss frequencies of odd oxygen ($O_x = O_3 + NO_2 + 2 \times NO_3 + HNO_3 + PAN + HNO_4 + 3 \times N_2O_5$). In the following tagged-O_x will be referred to as tagged-O_3 since ozone generally represents more than 95 % of odd oxygen. Model output was generated for ozone production in the lowest five layers of the model; this simulation provides information about the direct production of O_3 over the continent and hints at the origin of the pollution. However, the simulation does not account for a secondary production of ozone from ozone precursors in the free troposphere and, thus, underestimates the simulated contributions. Similar approaches have previously been used by Sauvage et al. (2007); Li et al. (2002); Liu et al. (2002).

3.4 Intercomparison

CO and O_3 concentrations from different instruments and from model simulations are compared in this study. Thus, it is necessary to take the different characteristics of the different observing systems into account (Rodgers and Connor, 2003; Palm et al., 2005). In-situ observations and model results are generally compared directly to each other. However, when comparing solar absorption FTIR profiles to GEOS-Chem model profiles, the characteristics of the FTIR retrieval have to be considered; the FTIR measurements do not provide simple measurements of the true state $\boldsymbol{x}$, rather they provide the estimated state of the atmosphere $\hat{\boldsymbol{x}}$ which can be derived from the weighted contribution of the true state and the a priori state $\boldsymbol{x}_\mathrm{a}$ (Rodgers, 1990, 2000).

$$\hat{\boldsymbol{x}} = \boldsymbol{x}_\mathrm{a} + \mathbf{A}\,(\boldsymbol{x} - \boldsymbol{x}_\mathrm{a}) + \boldsymbol{\epsilon}_x \tag{1}$$

In Eq. (1) $\boldsymbol{\epsilon}_x$ represents the error in $\hat{\boldsymbol{x}}$ and $\mathbf{A}$ represents the averaging kernel matrix. To account for the characteristics of the FTIR retrieval, model profiles $\boldsymbol{x}_\mathrm{m}$ are expressed as smoothed model profiles $\boldsymbol{x}_\mathrm{s}$ retrieved with the resolution of the FTIR instrument in the absence of the error term.

$$\boldsymbol{x}_\mathrm{s} = \boldsymbol{x}_\mathrm{a} + \mathbf{A}\,(\boldsymbol{x}_\mathrm{m} - \boldsymbol{x}_\mathrm{a}) \tag{2}$$

The GEOS-Chem model simulates CO and O_3 concentrations only in the troposphere. For the smoothing in Eq. (2) a full model profile is needed. The standard method for handling this problem is to create a composite profile by combining the modeled tropospheric profile with the FTIR a priori profile above the tropopause[3].

CO total column concentrations calculated from the FTIR profiles and the smoothed composed model profiles are compared directly to each other in Sect. 4. Although the model does not reproduce stratospheric variations, the error in the column is negligible since more than 90 % of atmospheric

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[3] The tropopause is defined as the lowest level at which the temperature lapse rate decreases to $2\,\mathrm{K\,km^{-1}}$ or less, and the lapse rate averaged between this level and any level within the next 2 km does not exceed $2\,\mathrm{K\,km^{-1}}$ (WMO definition).

CO is located in the troposphere. Furthermore, the CO retrieval is not very sensitive in the stratosphere (Fig. 2a) and mainly reproduces the a priori profile.

O_3 tropospheric columns are calculated from the FTIR profiles and are directly compared to the raw model tropospheric columns and the tropospheric columns derived from the sondes following the approach in Schneider et al. (2008). For the calculation of the FTIR tropospheric columns the model tropopause height is used.

4 Results and discussion

4.1 CO

Figure 4 shows the CO distribution during the four sections of the ship campaign with RV Sonne. Each plot in Fig. 4 is divided into the four cruise sections described in Table 1; sections S1, S2, and S4 are plotted over latitude and section S3 is displayed over time. Figure 4a shows the CO surface volume mixing ratio measured with the FTIR in-situ analyzer compared to flask measurements and the GEOS-Chem model. To exclude contamination by the ship plume the FTIR in-situ data is filtered by wind direction ($\alpha \geq 30°$) and relative wind speed ($v_{\mathrm{rel}} \geq 5\,\mathrm{m\,s^{-1}}$). Individual peaks in the dataset may origin from nearby ship traffic. In general, CO concentrations from flask measurements and the FTIR in-situ analyzer agree well. Small-scale variations resolved by the FTIR in-situ analyzer are not reproduced by the model. The cause of the overestimation in the beginning of cruise section S1 is discussed in Sect. 5.1. Figure 4b displays the CO total column amounts derived from the solar absorption FTIR spectrometer and the GEOS-Chem model. The model reproduces the main features in the CO total column distribution. Figures 4c–e show the CO volume mixing ratio (vmr) profiles from panel c: the solar absorption FTIR spectrometer, panel d: the GEOS-Chem model smoothed with the FTIR averaging kernels, and panel e: the raw GEOS-Chem model. The agreement between the model profiles and the FTIR profiles is good. Additionally, the model tropopause height (TPH) is shown. A typical equatorial TPH of around 17 km is simulated as well as the typical tropopause height decrease at 30° N and 30° S, respectively. The simulated TPH coincides with the measured TPH from radiosonde observations during cruise section S1.

Figure 4 especially reveals the impact of several pollution events (PE). At the location corresponding to the beginning of cruise section S1 the GEOS-Chem model simulates a strong pollution event from the surface up to the tropopause (PE1). It was not possible to observe the pollution by the solar absorption FTIR spectrometer due to bad measurement conditions during that time period caused by the typhoon Melor. FTIR in-situ data, however, does not support polluted surface air simulated by the GEOS-Chem model and shows, on the contrary, clean surface air of about 60 ppb. Between sections S1 and S2 around 20° S all instruments show a further CO enhancement (PE2). Surface volume mixing ratios suggest local pollutants in the harbor of Townsville, Australia, at 19° S. Local pollutants are captured by the model but underestimated due to resolution effects. Total column concentrations suggest the influence of a further CO source in this region since the concentrations start to rise at 10° S. Profiles obtained by solar absorption FTIR spectrometry reveal enhanced CO concentrations in 4–8 km altitude to be the main contributor to the increased total column amounts. This mid tropospheric CO pollution is reproduced but underestimated by the GEOS-Chem model. Within section S3 at 10° S the solar absorption FTIR instrument and the GEOS-Chem model reveal a CO band in the upper troposphere (PE3). FTIR and smoothed model profiles, both reveal the pollution at similar heights between 6–12 km altitude. Thus, the actual height of the CO pollution can be determined from the raw model output, which pictures PE3 in 12–17 km altitude within the tropical tropopause layer (TTL). Based on Fueglistaler et al. (2009) it can be assumed that CO pollution within the TTL is transported by radiative heating into the stratosphere in this area. Between section S3 and S4 a fourth CO pollution event (PE4) occurs. Surface volume mixing ratios and model profiles, both picture PE4 as a near surface event. Solar absorption FTIR measurements also suggest the impact of mid tropospheric CO pollution.

An overview of the four pollution events discussed in this section is presented in Table 2.

Table 2. CO pollution events (PE1–PE4) during the ship campaign with RV Sonne in the Western Pacific (TPH: tropopause height, LT: lower troposphere).

Pollution	Section	Latitude	Altitude
PE1	S1	43° N–20° N	0 km-TPH
PE2	S1–S2	10° S–20° S	0 km, 4–8 km
PE3	S3	9° S	12–17 km
PE4	S3–S4	9° S–20° S	LT

4.2 O_3

Figure 5 shows the O_3 distribution during the four sections of the ship campaign with RV Sonne (Table 1). Figure 5a displays the tropospheric partial column concentration of O_3 derived from the solar absorption FTIR spectrometer in comparison to tropospheric columns derived from O_3-sondes and the GEOS-Chem model. Figures 5b–e show the O_3 volume mixing ratio profiles from panel b: the FTIR spectrometer, panel c: the GEOS-Chem model smoothed with FTIR averaging kernels, panel d: the raw GEOS-Chem model, and panel e: the O_3-sondes (only cruise section S1). The overall agreement between the model and the observations is very

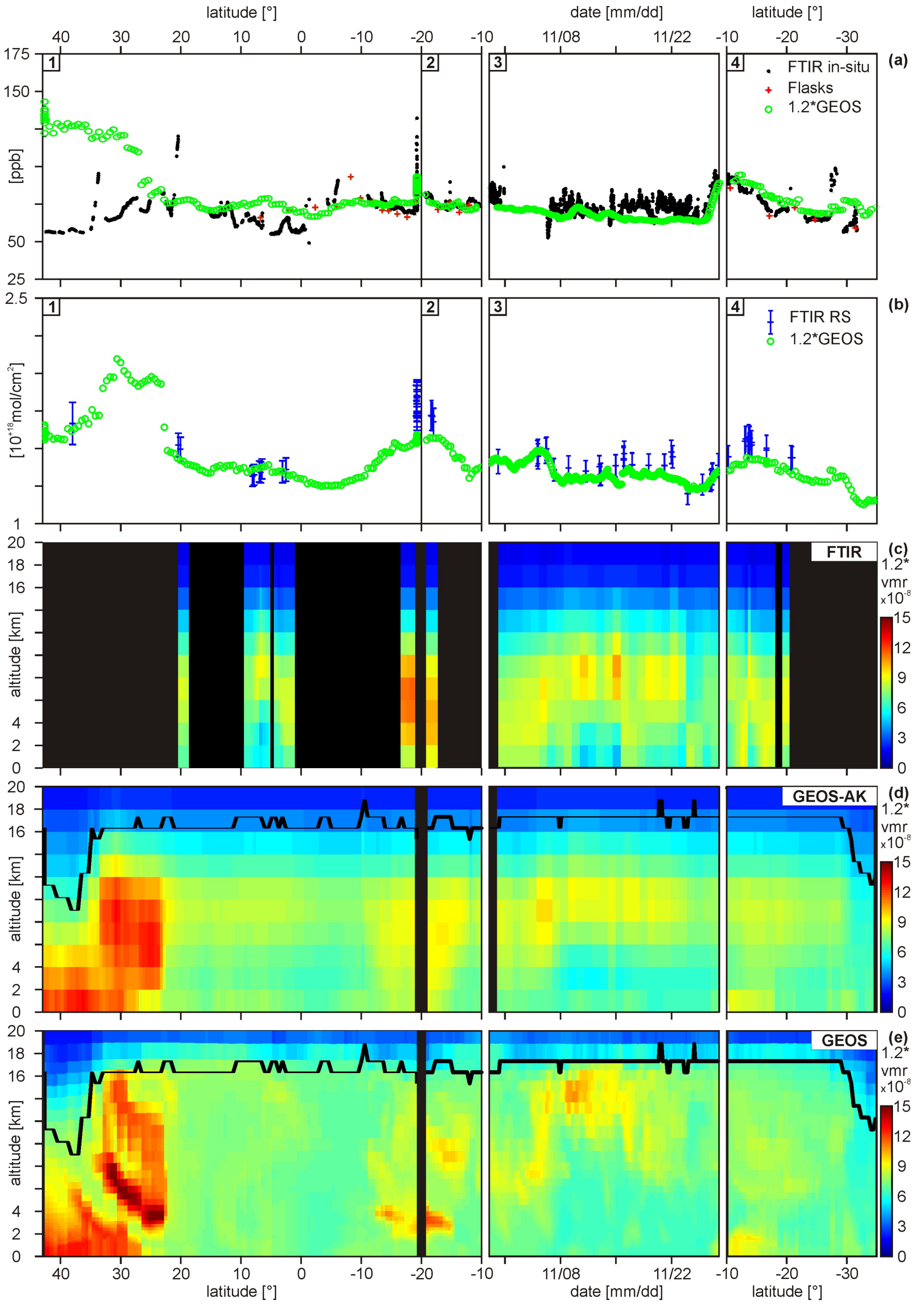

Fig. 4. CO distribution during the four sections of the ship campaign with RV Sonne in the Western Pacific; **(a)** surface volume mixing ratios obtained from FTIR in-situ measurements (black dots) and flask sampling (red crosses) compared to the GEOS-Chem model (green circles), **(b)** total column concentrations obtained from solar absorption FTIR measurements (blue markers, the error bars represent the theoretical uncertainty in the FTIR retrieval) compared to the GEOS-Chem model (green circles), **(c)** solar absorption FTIR profiles, **(d)** GEOS-Chem profiles smoothed with FTIR averaging kernels, **(e)** raw GEOS-Chem profiles. The black lines in **(d)** and **(e)** represent the model tropopause height.

good and the model reproduces the variability as well as the magnitude of the measurements.

In general, the O_3 distributions are quite similar to the CO distributions in Fig. 4 and all four pollution events (Table 2) can also be identified in the O_3 distributions. PE1 within cruise section S1 is this time also captured by the solar absorption FTIR spectrometer[4] and the O_3-sondes. Mid tropo-

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[4] The weather conditions in the beginning of the ship campaign partly allowed to measure not more than one to two spectra during the day. For those spectra the spectral region from 700 cm^{-1}

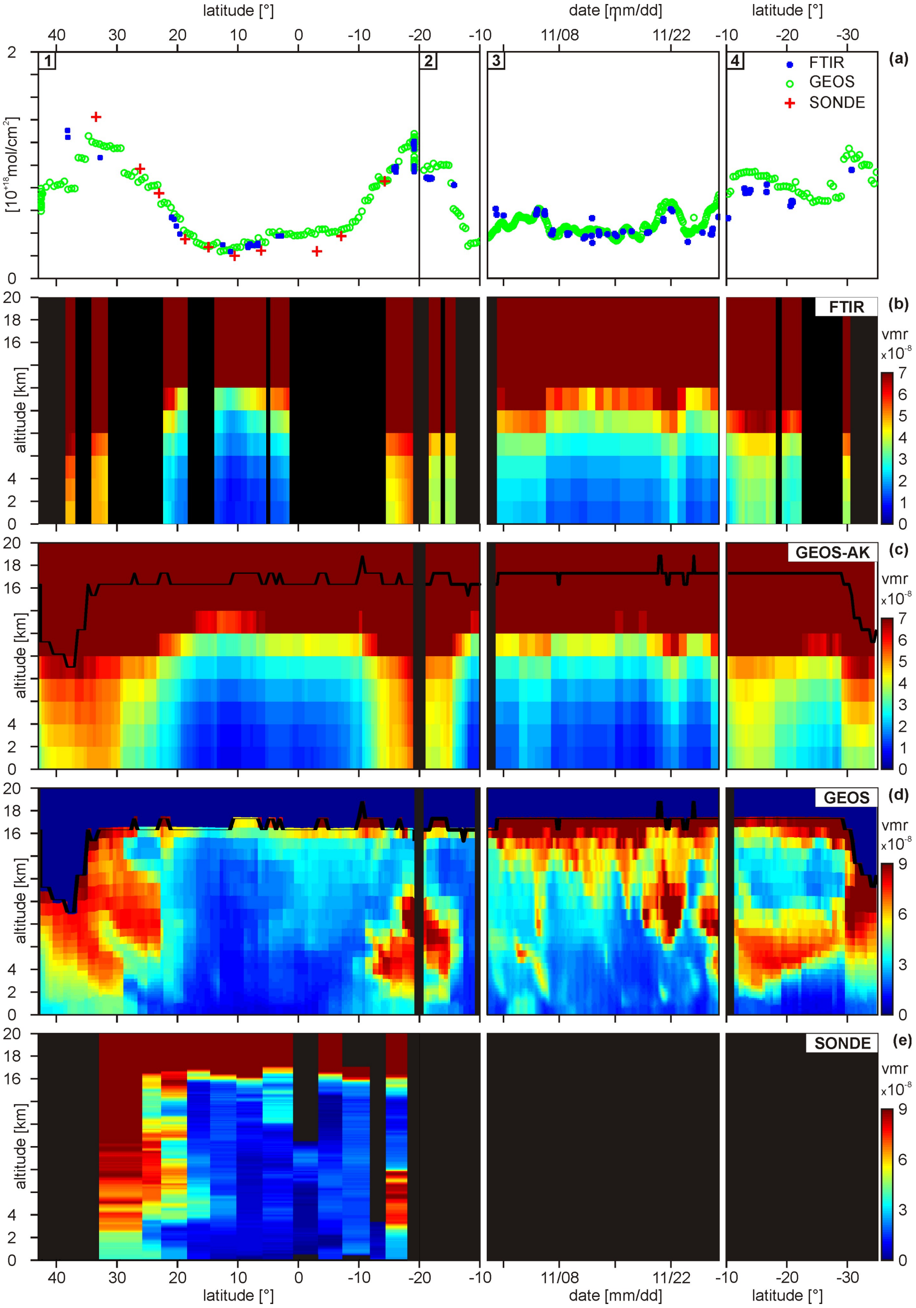

Fig. 5. O_3 distribution during the four sections of the ship campaign with RV Sonne; **(a)** tropospheric columns obtained from solar absorption FTIR measurements (blue dots) and O_3-sonde measurements (red crosses) compared to the GEOS-Chem model (green circles), **(b)** solar absorption FTIR profiles, **(c)** GEOS-Chem profiles smoothed with FTIR averaging kernels, **(d)** raw GEOS-Chem profiles, **(e)** O_3-sonde profiles. The black lines in **(c)** and **(d)** represent the model tropopause height.

spheric pollution (PE2) between cruise sections S1 and S2 is clearly observed by the FTIR spectrometer, the GEOS-Chem model, and the O_3-sondes. Upper tropospheric pollution in the Woodlark Basin (PE3) can not be resolved by the FTIR spectrometer, since high O_3 concentrations are convolved from the stratosphere into the troposphere by the averaging kernels and overlay the tropospheric O_3 signal. However, raw model volume mixing ratio profiles show enhanced upper tropospheric O_3. PE4 is seen in the mid troposphere rather than at the surface. Furthermore, enhanced O_3 at the end of cruise section S4 between 26° S and 32° S is observed.

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to 1350 cm^{-1} was preferred against other spectral regions. This spectral region includes the O_3 microwindow but not the CO microwindows.

Due to the fact that the O_3 and CO distributions are similar, one can hypothesize that the source regions of the observed CO and O_3 pollution are related. This aspect will be discussed in more detail in Sect. 5.

5 Sources of CO and O_3

To identify the source processes and source regions of the measured CO and O_3 pollution in the Western Pacific, the GEOS-Chem model tagged tracer simulations for CO (Sect. 5.1) and O_3 (Sect. 5.4) are applied. Model tagged tracer simulations deliver the contributions of various source processes and source regions to the modeled Western Pacific pollution. In addition, backward trajectory calculations combined with firemaps (Sect. 5.2) and solar absorption Fourier Transform infrared HCN measurements (Sect. 5.3) are presented. Trajectory studies reveal the transport pathways of Western Pacific air parcels. HCN measurements and firemaps show the impact of biomass burning. The discussion focuses on emissions from fossil fuel combustion and biomass burning.

5.1 Tagged-CO simulation

To identify the contributions of different source processes and source repetitive regions to the measured CO pollution (Fig. 4) in the Western Pacific with the GEOS-Chem model, CO is decomposed into tagged tracers as described in Sect. 3.3. The sum of all tagged tracers reproduces closely the results from the standard full-chemistry simulation shown at the top of Fig. 6. Below, the main CO contributions (with contributions $> 10\%$) calculated with the tagged-CO simulation are presented as the relative contributions to the total CO concentration.

During PE1 the main contribution is from Asian fossil fuel combustion (ASFF) ($\approx$40 %). European fossil fuel combustion (EUFF) ($\approx$15 %) and North American fossil fuel combustion (NAFF) ($\approx$10 %) contribute to lower tropospheric CO pollution between 43° N and 30° N. The impact of European and North American CO emissions on the Western Pacific and their transport pathways have been discussed in Jaeglé et al. (2003); Bey et al. (2001a). The GEOS-Chem model simulates contributions from ASFF, EUFF, and NAFF to the surface layer, whereas the FTIR in-situ analyzer suggest clean air concentrations of 60 ppb. This suggests a problem with the representation of the boundary layer in the model, which seems to mix free tropospheric air into the boundary layer too readily in this meteorological situation.

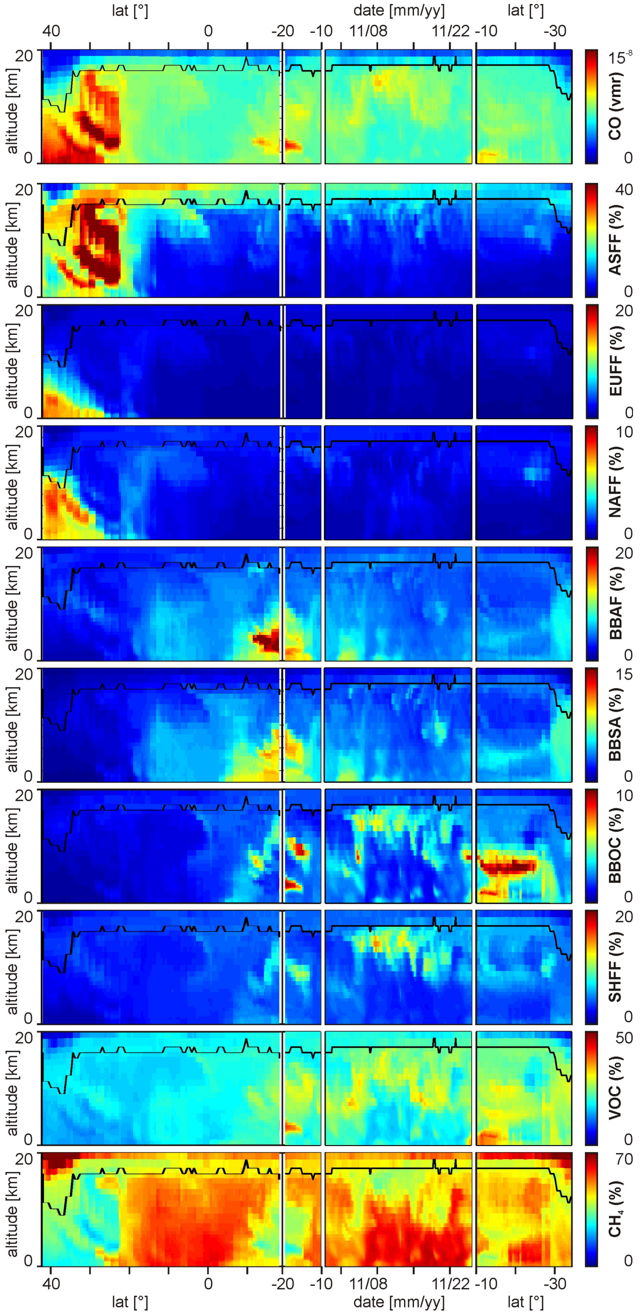

Fig. 6. Total CO vertical profiles (top) simulated with the GEOS-Chem tagged-CO simulation and relative contributions of various tracers from different trace regions (ASFF: Asian fossil fuel, EUFF: European fossil fuel, NAFF: North American fossil fuel, BBAF: African biomass burning, BBSA: South American biomass burning, BBOC: biomass burning from Oceania, SHFF: Southern Hemispheric fossil fuel, VOC: volatile organic compounds, CH_4: oxidation of methane). The black line in each graph indicates the model tropopause height.

The main contributions to the pollution close to Australia (PE2) are from African biomass burning (BBAF) (>20 %). Significant contributions ($\approx$15 %) originate from South American biomass burning (BBSA). Small contributions from biomass burning from Oceania (OCBB) ($\approx$10 %) are also apparent. The upper tropospheric pollution PE3 mainly originates from fossil fuel combustion in the Southern Hemisphere (SHFF) ($\approx$15 %). Biomass burning from the

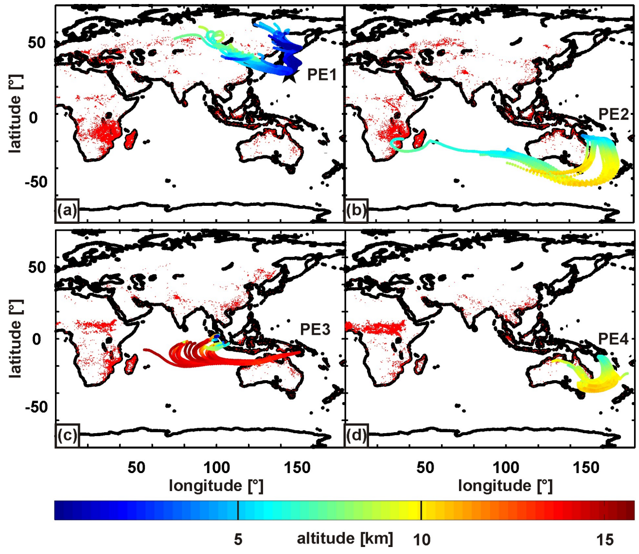

Fig. 7. HYSPLIT backward ensemble (120 h) and single (315 h) trajectories calculated at one representative altitude for the four characteristic locations; **(a)** PE1, **(b)** PE2, **(c)** PE3, and **(d)** PE4. The altitude of the trajectories is expressed by the color bar. MODIS fire counts (red dots) are displayed to picture the impact of biomass burning.

Oceania region contributes around 10 % to the total PE3 pollution. PE4 pollution mainly originates from biomass burning in Oceania (≈10 %). The most dominant background source is oxidation of methane (CH_4), which contributes up to 70 % in unpolluted regions. The strong variability in CH_4 relative contributions is mainly due to the variability in all other contributions. The absolute CH_4 contributions (not shown) ranges between 25 ppb and 35 ppb. Oxidation of volatile organic compounds (VOC) provides a background of around 20 % in unpolluted regions, but is also responsible for part of the CO variability in the Southern Hemisphere.

5.2 Backward trajectories

In order to verify the source regions of the measured CO pollution, backward trajectories were calculated with the NOAA HYSPLIT (National Oceanic and Atmospheric Administration Hybrid Single Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 1997, 1998) at the four characteristic points PE1, PE2, PE3, and PE4 (Fig. 7). For the calculations the NCEP-GDAS meteorological assimilation with a 1° × 1° grid resolution was used. One single backward trajectory (max. 315 h) embedded by ensemble backward trajectories (120 h) are presented for each pollution event. The trajectory ensemble starts multiple trajectories (in total 27) around the starting location. Each member of the trajectory ensemble is calculated by offsetting the meteorological data by a fixed grid factor (one grid meteorological grid point in the horizontal and 0.01 sigma units in the vertical). The altitude of the backward trajectories is expressed by the adjacent color bar. In addition, MODIS (MOderate Resolution Imaging Spectroradiometer) firemaps[5] are shown for the 5-day period prior to the corresponding trajectories in order to assess the influence of biomass burning.

Backward trajectories corresponding to PE1 show near surface air parcels (100 m) originating from northern directions indicating that European and American CO emissions (finding in Sect. 5.2) are transported over northern latitudes towards the Western Pacific. However, high altitude air parcels (2000 m) have their origin further to the west. Transport of pollution from Asia into the Western Pacific is verified. Backward trajectories corresponding to PE2 and starting at 4000 m show that enhanced CO concentrations close to Australia do not have their origin in the Australian continent. Air parcels are transported from the westerly Southern Hemisphere around the continent to the east of Australia. African biomass burning is confirmed to be the major source of the measured CO pollution since MODIS firemaps show strong fires in South-East of Africa during that time period. Backward trajectories corresponding to PE3 and starting at 14 000 m altitude show air parcels to be transported from Indonesia into the Western Pacific. The trajectories start at the Indonesian ground level, are elevated quickly into high altitude, and are then transported into the Western Pacific. Since MODIS firemaps only show a small influence of biomass burning, combustion of fossil fuel is the more likely source of the upper tropospheric CO pollution. PE4 corresponding trajectories starting at 150 m altitude show air parcels coming from the Australian continent, and MODIS firemaps show extensive fire counts in Eastern Australia supporting the impact of biomass burning to pollution PE4.

5.3 HCN

Hydrogen cyanide (HCN) is a common tracer for biomass burning emissions with a lifetime of a few months and is used to further investigate the influence of biomass burning (Holzinger et al., 1999; Li et al., 2000). HCN profiles derived from solar absorption FTIR measurements are used to investigate this influence on the measured pollution in the Western Pacific during the ship campaign with RV Sonne.

In Fig. 8 the HCN profiles derived from the solar absorption FTIR instrument are presented. Information about the retrieval are given in the appendix. Between section S1 and S2 enhanced HCN in the upper troposphere can be found supporting the assignment of air masses influenced by African biomass burning being transported to eastern Australia. A small enhancement in the beginning of cruise section S4 supports the influence of Australian biomass burning to pollution PE4. However, as no further HCN pollution can be determined from Fig. 8, other pollution events are obviously not significantly affected by biomass burning.

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[5] http://rapidfire.sci.gsfc.nasa.gov/firemaps/

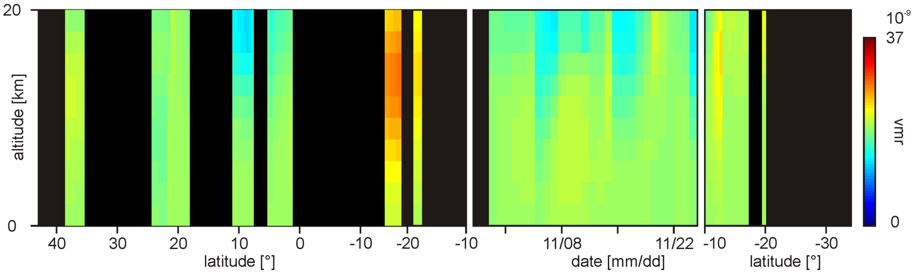

Fig. 8. HCN profiles retrieved from the solar absorption FTIR measurements during the ship campaign with RV Sonne in the Western Pacific showing the influence of biomass burning emissions on Western Pacific composition.

Particularly, air in the upper troposphere within cruise section S3 is somewhat depleted of HCN supporting the finding in Sect. 5.2 that fossil fuel combustion from Indonesia is a major source of upper tropospheric CO pollution in this area.

5.4 Tagged-O_3 simulation

In order to verify the source regions of the measured O_3 pollution in the Western Pacific, the GEOS-Chem tagged-O_3 simulation as described in Sect. 3.3 is used. The tagged-O_3 simulation closely reproduces the results from the full chemistry simulation shown at the top of Fig. 9. Below, Fig. 9 shows the relative contributions of various regions to the total simulated O_3 distribution. Since the simulation does not consider secondary production of ozone in the free troposphere from ozone precursors, the contributions of the tagged-tracers are underestimated. However, Fig. 9 provides an overview of the source regions of the measured O_3 distribution.

In the Northern Hemisphere, for PE1 the same source regions as identified by the tagged-CO simulation are revealed. Between 43° N and 35° N lower tropospheric pollution originates from European emissions, whereas middle and upper tropospheric pollution mainly originates from North American emissions. Pollution between 35° N and 20° N is caused by Asian emissions. The PE2 pollution in the middle troposphere close to Australia mainly originates from African emissions and South American emissions. Oceanian emissions show a gap in the middle troposphere verifying that Oceanian emissions are not a major source of the observed mid-tropospheric pollution. In cruise section S3 pollution PE3 only originates from Oceanian emissions supporting the conclusion that upper tropospheric pollution mainly originates from Indonesian emissions with small contributions from Oceanian fires as seen in Sect. 5.1. In cruise section S4 regarding pollution PE4 Oceanian emissions are the major source. However, between 26° S and 32° S enhanced O_3 amounts mainly originates from South American and African emissions.

In the Southern Hemisphere the contributions of African and South American emissions are small. Most of the O_3 pollution measured in the Western Pacific from these source regions is, thus, produced during the transport towards the Western Pacific through secondary production from ozone precursors such as CO.

6 Summary and Conclusions

In this study we have reported about the first measurements of CO and O_3 in the Western Pacific performed with solar absorption Fourier Transform infrared spectrometry during a ship campaign with RV Sonne in fall 2009. In-situ observations from three different instruments have also contributed to this study.

The observations have been compared to the 3-D chemistry transport model GEOS-Chem showing that the model generally reproduces CO and O_3 concentrations well. The carbon monoxide variability is clearly expressed by the model, however, absolute values are underestimated due to a constant low bias in the simulation already known from previous studies. In the beginning of the campaign, in mid latitudes, CO surface concentrations were largely overestimated by the model. Here, the model seems to mix free tropospheric air into the boundary layer too readily. Ozone concentrations were outstandingly reproduced by the GEOS-chem model during the entire ship campaign both in magnitude and variability.

The variability of the CO and O_3 distributions is largely similar suggesting impacts from similar source regions. This is verified with a subjective assessment of the source processes and source regions of the measured CO and O_3 pollution using tagged tracer simulations, MODIS firemaps, HYSPLIT trajectory calculations, and solar absorption FTIR measurements of HCN.

Large parts of the West Pacific belong to Earth's most remote areas. While much of the air observed in this region is considered the cleanest air world wide, the presented observations show the presence of distinct plumes of elevated carbon monoxide and ozone. Trajectory based transport studies and studies with a full chemical transport model show that the cause of this pollution is global scale transport of pollutants with contributions from source regions as far away as Europe.

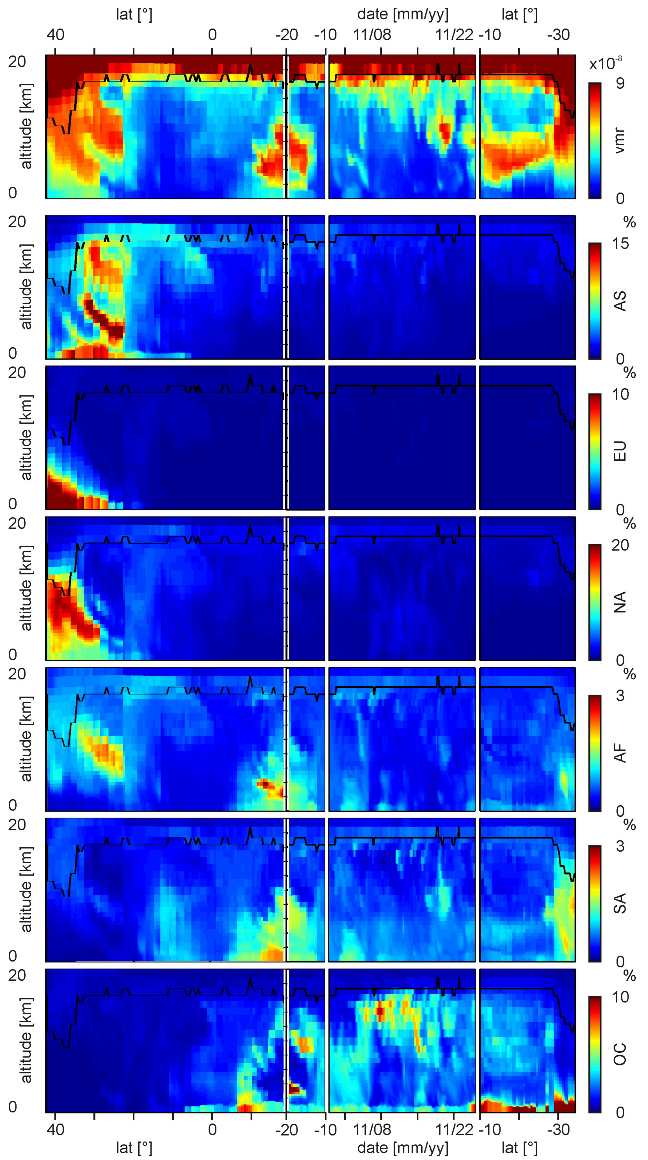

Fig. 9. Relative contributions of various GEOS-Chem tagged regions to the measured O_3 pollution calculated with the tagged-O_3 simulation. AS: Asia, EU: Europe, NA: North America, AF: Africa, SA: South America, OC: Oceania. The black line in each graph indicates the model tropopause height.

The analysis of several different carbon monoxide and ozone pollution events sampled during the RV Sonne ship campaign has demonstrated that in the Northern Hemisphere the main contributor to Western Pacific tropospheric air pollution is Asian fossil fuel combustion. Contributions from European emissions and North American emissions in the lower and middle troposphere are significant as well.

In the Southern Hemisphere biomass burning and fossil fuel combustion are determined to cause carbon monoxide and ozone pollution observed during the RV Sonne ship campaign. In particular, African and South American biomass burning significantly contribute to Western Pacific air pollution. Fossil fuel combustion in Indonesia is a major contribution to upper tropospheric pollution in the tropical Western Pacific. This pollution within the tropical tropopause layer is transported by radiative heating into the stratosphere and has an impact on stratospheric composition.

The presented analysis provides a detailed evaluation of the source processes and source regions of Western Pacific carbon monoxide and ozone pollution measured during a ship campaign from Japan to New Zealand in fall 2009. Thus, the analysis is limited on the abovementioned period ob time. In order to obtain a better knowledge of Western Pacific air contamination, future measurements in this region with high temporal and spatial resolution are needed as they are currently performed by, for example, the HIAPER Pole to Pole Observation (HIPPO) aircraft campaign (Wofsy, 2011) and the SHIVA (Stratospheric ozone: Halogen Impacts in a Varying Atmosphere) campaign.

Appendix A HCN retrieval

In Fig. 8 the solar absorption FTIR profiles of HCN during the ship campaign with RV Sonne were shown. HCN was simultaneously fitted in four microwindows ($3268.1800\,\mathrm{cm^{-1}}$–$3268.2700\,\mathrm{cm^{-1}}$, $3287.1800\,\mathrm{cm^{-1}}$–$3287.3200\,\mathrm{cm^{-1}}$, $3299.4600\,\mathrm{cm^{-1}}$–$3299.5800\,\mathrm{cm^{-1}}$, $3305.4000\,\mathrm{cm^{-1}}$–$3305.7000\,\mathrm{cm^{-1}}$) with H_2O, C_2H_2, and CO_2 also fitted as interfering gases. HCN reaches a DOF of around 2 with the main sensitivity in the upper troposphere. The corresponding averaging kernels are presented in Fig. A1.

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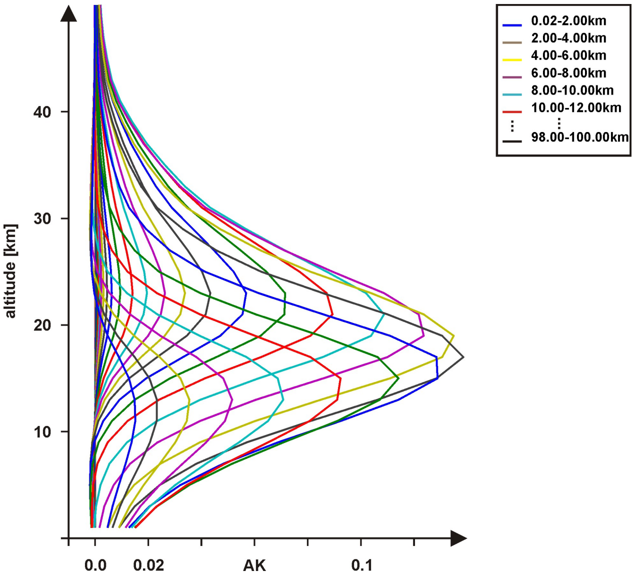

Fig. A1. Representative averaging kernels of the FTIR HCN retrieval. The colored lines indicate the sensitivity and resolution of each individual layer.

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