

1 *Author's Reply to comments from the referee #1*

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3 We would like to thank the referee #1 for his/her interest in our work and helpful comments to
4 improve our manuscript. Listed below are our replies to the referee's comments and
5 suggestions, which we will revise our manuscript according to.

6

7 <Referee> I still would like to see more scientific discussion of the data, e.g. seasonal
8 variation, longitudinal and latitudinal gradients. By now a significant amount of data
9 has been collected, and I recommend Section 5 (preliminary results) to be somewhat
10 extended. The Western Pacific region has been studied comparatively well in the past
11 using various platform types, especially in the northern hemisphere, and the new data
12 could for example be compared to data from coastal stations in the region. To my
13 knowledge there exist or have existed similar measurement programs deploying
14 commercial ship in the Pacific that should be referenced.

15 <Reply> Thanks very much for your suggestions, in particular, keen eyes to past observations
16 in the western Pacific region. In this paper submitted to Atmos. Meas. Technol., we
17 paid great attention to the instrumentation of continuous measurements of
18 atmospheric CO onboard cargo-ships, and tried to focus on technical aspects of this
19 shipboard system in detail. We are currently in the process of finalizing the observed
20 data from both continuous measurements and bottle samples. Detailed analysis of
21 seasonal, latitudinal, and longitudinal characteristics of CO and co-measured species
22 such as CO₂ and O₃ on the basis of several ships will be presented in a separate paper,
23 which we plan to submit to Atmos. Chem. Phys. in a due course. We will refer to
24 previous measurement programs deploying commercial ship in the revised paper for
25 sure.

26

1 <Referee> The authors mention that besides CO also O₃ and CO₂ are measured. Why is none
2 of this data shown? It is mentioned that commercially available instruments are used
3 for this. What is the data quality of these measurements? Section 3.4 explains that
4 CO₂ measurements are used for quality control of the data. How about CO/CO₂
5 correlations? Also, the flask samples seem to be analyzed for a variety of other
6 compounds that could help investigate CO source types.

7 <Reply> As described above, this paper focuses on the methodology of shipboard CO
8 observations and its field deployment on commercial ships. Hence we briefly
9 mentioned CO₂ and O₃ by referencing to our previous papers. Detailed analysis of
10 CO including relationships with CO₂ and O₃, and other gaseous species from flask
11 samples is currently in preparation and will be submitted to Atmos. Chem. Phys.
12 soon.

13

14 **Specific comments**

15 <Referee > The expression “over the Pacific Ocean” on first reading implies measurements
16 are performed onboard aircraft. Although it is clearly said, that these are ship based
17 measurements, it should be “in the Pacific” or “in the Pacific region” or “in the
18 Pacific boundary layer”.

19 <Reply> We will replace “over the Pacific Ocean” with “in the Pacific”.

20

21 <Referee > Is the same air intake used for all measurements (flasks, continuous CO, O₃,
22 CO₂)? The position of the intake is mentioned a couple of times. Please make this
23 clear and omit afterwards.

24 <Reply> We have individual intakes for flasks samplings, and continuous CO, O₃ and CO₂
25 measurements. We will clarify this description.

26

1 <Referee > How much time is in between flask sample collection and GC analysis? What are
2 typical O₃ values encountered? Has CO growth in the cylinders been investigated?

3 <Reply> This is really an important point for CO analysis in flask samples. The time between
4 sampling and analysis depends on ship's routes (i.e., TF5, FTW, SKB) and sampling
5 locations (i.e., first or second half of the cruise). Flask samples are analyzed
6 immediately after the flasks have returned to laboratories at NIES. This means, all
7 the flask samples are typically analyzed within 1 to 2 month after collection of
8 ambient air. Typical O₃ mixing ratios are different depending on the ship's route. The
9 O₃ mixing ratios are less than 20 ppbv in the open ocean but are more than 40 ppbv
10 in coastal area. We examined the CO growth in the cylinders of gas standards as
11 shown in Fig. 5. Since the growth is significant for ppbv-level standards used for
12 flask analysis, we have applied correction factors before comparison to continuous
13 data.

14

15 <Referee > It is not clear to me what the different measurement time scales are.

16 <Reply> This sentence means that 10-sec data from CO, CO₂, and O₃ instruments are merged
17 into a same electric file on the GPS time, and then 1-min mean data are produced for
18 further data processing.

19

20 <Referee > P4507, L23-P4508, L4: To my knowledge CO measurements are also performed
21 as part of the Japanese CONTRAIL program. If so, this should also be mentioned
22 here. Yashiro et al in JGR 2009 also discussed a similar project of ship-based
23 sampling that might be a suitable reference here because similar measurements are
24 carried out in the same region.

25 <Reply> We will revise the introduction section by adding CONTRAIL program and
26 ship-based flask sampling observation by Yashiro et al. [2009] to the Reference list.

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<Referee > P4510, L20/21: “including gases”: Later calibration gases and the use of nitrogen are discussed, presumably only potentially poisonous gases are meant?

<Reply > Yes, it means any hazardous or flammable gases. We will correct the text to make it more understandable.

<Referee> P4512, L22: At which time resolution are the different types of raw data recorded?

<Reply> All the continuous instruments: CO, CO₂, and O₃ analyzer provide 10-sec data.

<Referee> Section 2.2: The title is confusing because the term continuous system has been used before to distinguish between the continuous CO measurement and the flask sampling. The importance and length of this paragraph does not justify the separate treatment. It can be merged into section 3. More details about the analyzers used should be given, e.g. what are the measurement techniques, detection limit, precision, time resolution...

<Reply> We will integrate sections 2.2 and 2.3 to make a new section. Technical details of analyzers are given in references.

<Referee> P4511, L6: the term “air sample” should be used careful to avoid confusion with the flask samples.

<Reply> To avoid confusion, we will use a term “continuous air sample” and “flask air sample” in a revised manuscript.

<Referee> P4511, L16: Why is the number of samples limited to seven? This is a very small number for the large distances traveled.

<Reply> As shown in Fig. 1, 21, 14, and 7 samples are collected in total for CO analysis

1 along the route of TF5, SKB, and FTW, respectively. Each air sampling equipment
2 contains seven glass (or stainless) flasks. Several air sampling equipments can be set
3 on the automatic air sampler unit depending on the space of the observation room.
4

5 <Referee> P4512, L4/5: What is the actual sampling time for one flask?

6 <Reply> After the flask is rinsed, air sample is collected for 4-5 min with ambient air
7 pressurized up to 1.6 kgf for glass flasks and 2.5 kgf for stainless steel flasks,
8 respectively. We will add this phrase to a revised manuscript.
9

10 <Referee> P4513, L5ff: Please mention that continuous measurements are performed in
11 addition to flask sampling.

12 <Reply> We will update section 3 according to the referee's comments.
13

14 <Referee> P4514, L25: In section 2.1, it was mentioned that no gases were used onboard,
15 Please clarify.

16 <Reply> As mentioned above, only hazardous or flammable gases are unusable.
17

18 <Referee> P4516, L22: 20 min zero-air injection time seems rather long. The daily calibration
19 time is only 10 min. How long does the instrument need to stabilize after changing
20 from sample to zero air? Is it ensured that flask sample collection takes place while
21 ambient air is measured?

22 <Reply> We apply relatively long zeroing period to obtain zero-levels more precisely,
23 especially for low CO mixing ratios typically seen in the southern hemisphere. We
24 believe that this is acceptable for measurements on ships that are moving "relatively"
25 slowly. For comparison of continuous measurements and flask analyses, we only
26 compared concomitant data within 1 hour.

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<Referee> P4516, L25: Does that mean that a 40 min averages is taken or is the mean only calculated for the drift correction? At which time resolution are the raw data recorded? This needs to be clarified.

<Reply> For CO, 10-sec instant and 1-min mean data are available. To obtain highly precise CO mixing ratios from GFC measurement, data for a few tens of minutes were averaged. We calculate CO mixing ratios from 40-min data.

<Referee> P4519, L24: How about the fourth route (PX)? How many samples were collected?

<Reply> On board PYXIS, 14 flask samples were collected but continuous CO measurements were not made (Please see Figure 1). Hence we do not describe the onboard observation of PYXIS.

<Referee> Section 4.1: I suggest to skip this section. It does simply state that there is reasonable agreement between flask sampling and continuous measurements, the quantitative details of this are presented in section 4.2. It is no big surprise and does not need to be discussed in such detail that high resolution measurements capture more details than coarse flask sampling, and unsurprisingly this is more relevant in regions with high spatial variability.

<Reply> We believe that this section is one of the most important sections in this paper, because it demonstrates reasonably good agreement between two independent techniques to determine atmospheric CO, in very different environments observed in TF5, FTW, and SKB routes. Technically, it would also be difficult to illustrate the CO variations from Figure 9 only.

1 <Referee> P4520, L6: It is mentioned in section 2.2 that 10s and 1min data are reported, so
2 why are 40 min mean values used? See also my question on the 40 min averaging
3 above. I was unable to find the exact time resolution of the continuous instrument
4 and I am not sure what the sampling time is, but it should be possible to calculate a
5 well matching integral value from continuous measurements for each flask sample.

6 <Reply> Please see our reply above.

7

8 <Referee > P4522, L2ff: “four distinct regimes” may be a better expression to use here. For
9 better understanding refer to the colors used in Fig. 6. The statement about the Asian
10 monsoon circulation needs be clarified. In the current version the discussion of the
11 FTW route winter cruise (P4521, L25 – P4522, L22) is very difficult to follow.

12 <Reply > Thanks for this comment. We will revise this sentence accordingly, and add the
13 explanation of Asian monsoon circulation.

14

15 <Referee > I suggest to merge Figure 1 and 9 into one figure and to include a table
16 quantifying the number of cruises on each route.

17 <Reply > We think that flask sampling locations should be provided along with the ship’s
18 route. It is difficult to clearly show the locations in Figure 9 only.

19