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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temporal changes in the emissions of CH_4 and CO from China estimated from CH_4 / CO_2 and CO / CO_2 correlations observed at Hateruma Island" *by* Y. Tohjima et al.

Anonymous Referee #1

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Summary: Tohjima et al present the long-term data set of the atmospheric observations of CO2, CH4 and CO observations from the GAW station Hateruma Island (HAT). After data selection and fitting/detrending they interpret their hourly data set in regards to the observed synoptic scale variability (SSV). Trace gas concentrations are attributed to fluxes of CO2, CH4 and CO from East Asia using an approached based on interpreting the interspecies concentration ratios. Tohjima et al. also report the long-term trends of these ratios (DCO/DCO2, DCH4/DCO2 and DCH4/DCO). The source region for the concentration signal recorded at HAT is identified using forward simulations using the FLEXPART model. Using available bottom-up data from EDGAR V4.2 and CDIAC

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Discussion Paper

Tohijma et al. derive flux estimates for CH4 and CO for their area of influence (AOI) assuming the CO2 emissions are well-known. The calculated CH4 and CO fluxes and their inter-annual trends are finally compared to different bottom-up emission data sets and other top-down derived emission estimates.

General comments: The paper is well-written and the presented observational data sets are of very high quality. The description of the measurement techniques, calibration and quality assurance methodology is concise - yet exhaustive. The data processing technique applied (Thoning-filtering) and interpretation of SSV data is a standard technique (cf. Tohjima 2011 et al., ACP), but is novel for hourly data from this location. The methodology of using observed atmospheric trace gas ratios e.g. DCH4/DCO2 in combination with a known inventory is a well-established and suitable technique. The estimated uncertainty of 15% for the CH4 and CO emission estimates derived using this technique seems, however, very low. Besides the large uncertainty in the emissions in this region (e.g. Guan et al. 2012, Nature Climate Change) other sources of uncertainty should be considered. The cited uncertainty of emissions of 15% (Gregg et al. 2008, JGR) is valid for national totals - the EDGAR inventory does, however, have known limitations concerning the spatialisation of GHG emissions within large countries (e.g. Canada: Vogel et al. 2012, JIES; Nassar et al. 2013, JGR). As the sensitivity of the footprint differs across China (cf. Fig. 5) an erroneous spatial pattern of CO2 emissions could possibly alter the emission estimate beyond the assumed 15% uncertainty. “What is the typical uncertainty for the effective footprint area?” Is an open question that could be answered by comparing the EDGAR and e.g. the PKU emission data set for the EFA. Beyond this the observation-model comparison also displays great differences. The observed concentration ranges differ by a factor of two for CO2, factor of two for CH4 and a factor of six for CO (cf. Figure 3). This needs to be addressed in more detail here as the FLEXPART modelling result is crucial for identifying the source region (EFA) in this study. Another important improvement could be to compare the results of this study with the latest version of the REAS inventory (V2.1). It extends well beyond 2003 and also differs significantly from the previous

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version (V1.1), used in this study (cf. Kurokawa et al. 2013, ACPD). This data has not been available at the time of submission of this study, but seems valuable to add now. Overall, the observational data from HAT, the observation-based emission estimates and the interpretation of the results presented here are of great importance. HAT is one of very few long-established, high-precision observatories in East Asia - a region of growing importance for the global anthropogenic Greenhouse Gas budget. After addressing the general and specific comments this study will undoubtedly be a valuable addition to ACP and of interest to its readership, which is why I recommend its publication thereafter.

Specific comments:

Page 22894 Line 10ff. The author state that they observe a gradual trend over the whole period, but no trend 1999-2004 and no trend 2005-2010. This seems to indicate an abrupt change in 2004-2005, which would be the opposite of a gradual trend. Looking of the data I would argue that the trend seems to be gradual, but the noise/variability in the data does require a long time-series (>5a) to detect this trend.

Page 22894 Line 12-14 Please expand how FLEXPART proved that the emission changes are caused by emission variations and not by variations in atmospheric transport? E.g. are you sure the spatial distribution of EDGAR is realistic and up-to-date? (See general comments)

Page 22894 Line 25 Please mind that CH4 is only the third most important GHG after water vapor and CO2, but the second most important anthropogenic GHG.

Page 22898 Line 1 Please clarify the term “cleanup air”

Page 22899 Line 24 Did you test the sensitivity of the result to the choice of cut-off frequency?

Page 22900 Line 8 Please elaborate why you chose a threshold of |R|>0.8 . Another concern here is the lacking selection according to the temporal evolution of the con-

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centration (increase vs. decrease). A positive correlation can be found for an increase in CO2 and CH4, which is caused by emissions within the EFA. A positive correlation can, however, also be found for a concurrent decrease of both CO2 and CH4 due to the dilution with clean-air. This signal is not reflective of the emission ratio within the EFA, but a mere mixing signal.

Page 22900 Line 21 Could you give a quantitative estimate of the effect of the seasonally changing CO emissions?

Page 22902 Line 20 Please change “atmopsheric” to “atmospheric”

Page 22902 Line 22 Why did you not use the EDGAR V4.2 inventory for CH4 (as for CO2 and CO)?

Page 22903 Line 14 The comparison of model and data shows some similarity, but the scales of the signals are significantly different (see general comment).

Page 22903 Line 24 The findings of Tohjima et al. 2010 are based on the assumption of similar spatial distribution of CO2, CH4 and CO fluxes in East Asia – did you test this assumption by comparing the spatial distribution of the emissions in EDGARV4.2 and the REAS data sets for the study presented here?

Page 22904 Line 14 As most of the changes occurred 2004 to 2005 it would seem more logical to show this year rather then 2001/2002 or 2003/2004.

Page 22905 Line 16 What is your explanation for the inability to reproduce the leveling off?

Page 22908 Line 4 The discussion of the CH4 emission estimates is extremely short and should be expanded. The latest version of the REAS should be used (see general comments). Carbontracker methane or CarboScope results could also be interesting as basis of comparison.

Page 22908 Line 10 Please change “EDGARR” to “EDGAR”

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Page 22909 Line 17 Please clarify if you meant to say “underestimate” here. The difference between EDGARV4.2 and your results in Figure 10 seems to suggest that EDGAR underestimates the real emissions.

Figure 1. Please include the CH4 and CO emissions for the USA to be consistent

Figure 3. Please consider using same scales to allow a direct comparison of model and observations

Figure 7c. Please consider using “initial and corrected emission ratio”, to clarify the difference of “emission ratio” and “Sim (estimated emissions)”

Figure 9. Please correct “READ” to “REAS”

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 22893, 2013.