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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global and regional emissions estimates of 1,1-difluoroethane (HFC-152a, CH_3CHF_2) from in situ and air archive observations" *by* P. G. Simmonds et al.

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Referee 1. We thank this referee for his constructive comments and important corrections which are addressed below:- General comments. Document has been edited for punctuation, etc. Information in the results section (P21352, L10-24) has been moved to the methods section 3.4 as requested. The discussion section has been improved where necessary to clarify which method is being discussed. (See response to specific comments). The relative contribution of HFC-152a radiative forcing has been added to the end of the Results and Discussion as follows:- "As reported by Rigby et al, (2014) the major long lived synthetic greenhouse gases (SGHG) which include

CFCs, HCFCs, HFCs and perfluorocarbons (SF6 and NF3) ; as well as CH3CCl3 and CCl4 were responsible for 350 $\pm$ 10mW/m2 of direct radiative forcing in 2012. The radiative forcing of HFC-152a, determined from the AGAGE 12-box model in this study, was 0.61 $\pm$ 0.2 mW/m2 in 2014, which represents only a tiny fraction ($\sim$0.2%) of the global radiative forcing of the SGHG". Specific comments. P21337, L1-6: This sentence has been changed to the following two sentences "High frequency, in situ observations from eleven globally-distributed sites for the period 1994âĂŠ2014 and archived air measurements dating from 1978 onward have been used to determine the global growth rate of 1,1-difluoroethane (HFC-152a, CH3CHF2). These observations have been combined with a range of atmospheric transport models to derive global emission estimates in a top-down approach". P21337, L12: HFC-152a lifetime ($\sim$1.5 years) has been added to abstract. P21337, L25-26: "significant underestimate (> 20 Gg)" has been added. P21339, L4: Changed to "concentrated in Germany" P21341, L24: "dating back to 1978" has been added. P21342, L19 and L21-22: Sentence has been clarified as follows: "Baseline in situ monthly mean HFC-152a mixing ratios were calculated by removing enhancements, due to local and regional pollution influences, using the AGAGE pollution identification algorithm" A brief description of the algorithm has been added as requested. P21343, L27: Missing height has been added. P21344, L2: "Emissions were estimated between 1989 and 2014. . ." P21344, L5-6: The overall uncertainty calculation includes contributions from the observations, the prior and the atmospheric lifetime. The methods have been discussed in detail elsewhere; therefore we have added a reference to the method used: "Global emissions were derived that included estimates of the uncertainties due to the observations, the prior and the lifetime of HFC-152a, as detailed in the supplementary material in Rigby et al. (2014)" P21344, section 3.2: We have modified the final sentence of this section. As above, we have decided that a full description of the uncertainty quantification is not appropriate here, as it has been well documented elsewhere (Ganesan et al., 2014 and Lunt et al., 2015 in this case): "Emissions were estimated using a hierarchical Bayesian inverse method (Ganesan et al., 2014; Lunt et al., 2015) and all high-frequency observations

from 10 of the 11 sites listed in Table 1, excluding Shangdianzi due to its short time series. The hierarchical Bayesian method includes uncertainty parameters (e.g. model "mismatch" errors and a priori uncertainties) in the estimation scheme, reducing the influence of subjective choices on the outcome of the inversion." P21345, section 3.3: Following text has been added to Section 3.3 "The uncertainty estimation used within InTEM is described in detail elsewhere (Manning et al 2011). The uncertainty space was explored by a) solving the inversion multiple times with a range of baseline mole fractions within the baseline uncertainty estimated during the baseline fitting process and b) by altering the 3-year inversion time window by one month throughout the data period thereby solving over a particular one year period many times using different observations. In total for each annual estimate up to 111 inversions were performed, the median and 5th and 95th percentiles were used as the final total and spread. For the Australian estimates data between 2002 and 2011 inclusive was used, for the NW European estimates data between Nov. 1994 and Dec. 2013 were used". P21345, section 3.4: The required information was moved from section 5.2.2 to 3.4 and further details concerning uncertainty were added to the text. P21346, section 3.5: The referee brings up an important point and the following discussion has been added to the text – "ISC works best for co-located sources – however extensive modelling has shown that by the time the Melbourne/Port Phillip plume reaches Cape Grim (300 km from the source) it is well mixed and the likely inhomogeneity of the source regions (for CO and HFC-152a in this case) does not have a significant influence on the derived emissions. It should be noted that in order to obtain a significant sampling of Port Phillip pollution episodes at Cape Grim, data from 3 years (for example 2011-2013) are used to derive annual emissions (for 2012). (InTEM also uses data from 3 years to derive annual emissions. The ISC uncertainties given in the paper include (1) the uncertainties in the estimates of CO emissions from Melbourne/Port Phillip (2) the uncertainties in the overall correlation between CO and HCFC-152a as seen in pollution episodes at Cape Grim (3) the uncertainties in the geographic extent of the HFC-152a and CO source regions impacting on Cape Grim and their entrained population." P21348, L19-20: Figure

6 shows the "baseline filtered" semi-hemispheric average observations that were used in the inversion. The solid line shows the a posteriori model run. We have clarified this line to reflect this: "Figure 6 shows the mole fractions output from the AGAGE global 12-box model, along with the monthly-mean semi-hemispheric average observations used in the inversion. The figure also shows the running mean growth rate, smoothed using a Kolmogorov–Zurbenko filter with a window of approximately 12 months (Rigby et al., 2014)." Figure 6: Caption changed to: "Top panel: AGAGE 12-box model mole fractions for the two NH and two SH latitudinal bands (solid line). The points show the semi-hemispheric monthly mean observations from the 5 AGAGE stations used in the inversion (MHD, THD, RPB, SMO, CGO). Lower panel: HFC-152a annualised growth rate (see Rigby et al., 2014 for smoothing method) for each semi-hemisphere, with the heavy blue line and shading showing the global average and its uncertainty." P21349, L6: "using the 12-box model" has been added. Figure 9: We think that you mean Figure 8, where the information has been expanded to improve the description of the figure. P21352, L10-24: Information has been moved to the methods section 3.4 as requested. P21354, L21: "HCFC-22 as the reference tracer" has been added. P21355, L4-5: Replaced with this text: "SE Australian emissions of HFC-152a are estimated using the positive enhancements above baseline or background concentrations observed at Cape Grim using interspecies correlation with CO as the reference species (ISC: Dunse et al., 2005; Greally et al., 2007) and inverse modelling (InTEM: Manning et al., 2003, 2011)."

P21355, L20-21: Yes, the emission estimates are part of this study as an update to a previous study (Greally et al., 2007) – the Australian emissions have been reported separately to the Australian Government Department of the Environment (DoE) (references give) in accord with DoE requirements for funding of the Australian component of this research.

P21356, L8-9: Replaced with this text: "It is unusual for Australian emissions of an industrial chemical to be as low as 0.1% of global emissions. For other HFCs, CFCs

and HCFCs (for example HFC-134a, CFC-12, HCFC-22), Australian emissions as fraction of global emissions are typically 1-2%, similar to Australia's fraction of global gross domestic product (GDP, 1.9%, 2014) but significantly larger than Australia's fraction of global population (0.33%, 2014) (Fraser et al., 2014b)." (Note: Discussion of CO as a proxy has been included in Section 3.5) Following text has been added to the InTEM discussion: "The method for estimating the InTEM uncertainties are discussed above. No additional uncertainty was applied to the estimates through the process of up-scaling from Victoria/Tasmania to Australian totals. The assumption was made that the use of HFC-152a per head of population was identical across Australia; we have no further information to use." Technical comments P21337, L10: Sentence has been changed. "The concentration of HFC-152a has grown substantially since the first measurements in 1994,. . .. . P21337, L11: Changed as requested. P21338, L17: Changed as requested. P21339, L4-5: Changed as requested. P21339, L8: Changed as requested. P21339, L19: Changed as requested. P21339, L22: Changed as requested. P21346, L25: Changed as requested. P21348, L7: Changed as requested.

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