

## ***Interactive comment on “Tropospheric observations of CFC-114 and CFC-114a with a focus on long-term trends and emissions” by Johannes C. Laube et al.***

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As an initial remark we would also like to acknowledge the work of this anonymous reviewer which has further improved this manuscript. Below please find responses to all comments.

Referee comment

Pg. 5, Line 25: It seems that Laube et al 2010 offers the best description of the dilution method. I think Laube et al 2012 should be Laube et al 2010.

Author response

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The sentence was changed to “Calibration scales were established for CFC-114 and CFC-114a by a two-step dilution process described in Laube et al. (2010) which was improved later (Laube et al., 2012).”

Referee comment

Pg. 7, Line 17: The sentence “The recommended values . . .” is missing something.

Author response

Both the sentence “The recommended values mentioned above are based on work by and.” and the text above it are intended to convey a similar message. Hence, sentence and text have been revised as follows:

“The rate coefficients of  $1.43 \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  and  $1.62 \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  are applied for the reaction of O(1D) with CFC-114 and CFC-114a. The recommended values for CFC-114 and CFC-114a mentioned above are based on work by Baasandorj et al., (2013) and Baasandorj et al., (2011), respectively.”

Referee comment

Pg. 7, Line 21: I believe that Carpenter and Reimann adopted lifetimes based on SPARC (2013), so it would suffice to use SPARC (2013) as the reference for the 100 and 189 lifetimes.

Author response

CFC-114a was not included in the SPARC report which is why we feel it is necessary to keep both references.

Referee comment

Pg. 7, Line 5: Not really a complaint, just an observation: You used a relatively sophisticated model, and yet the model is driven largely by data from one site (Cape Grim) or firn air (a smoothed record), and UV absorption data needed to be highly tuned

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using the parameter  $F$  to achieve the recommended total lifetimes. It seems a simple model might suffice given the limited data. I suppose you used a model that was readily available.

Author response

We are using a model that has been proven to work well for a variety of atmospheric trace gases but in particular CFCs.

Referee comment

Pg. 8, Line 11: Not quite sure what is meant by “early day”

Author response

This was one of the first models to infer atmospheric trace gas trends from firm air observations. Many improvements have been made since and we have provided some details in section 3.4

Referee comment

Pg. 9, Line 10: In, “These two facts imply that increasingly higher emissions of CFC-114a would be needed to sustain increases in mixing ratios above those of CFC-114. “ Do you mean that higher emissions of CFC-114a would be required to sustain the observed growth rate of CFC-114a? The mixing ratio of CFC-114a was never higher than that of CFC-114, so I’m not sure what you mean here.

Author response

We agree with the referee and have changed the sentence to: “These two facts imply that increasingly higher emissions of CFC-114a would be needed to sustain relative increases (as a percent of the abundance) above those of CFC-114.”

Referee comment

Pg. 9, Line 16: Carpenter and Reimann (2014) state that the assumption of 10%

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relates to the abundance of CFC-114a relative to CFC-114, based on measurements from 1990. Please provide a reference for the "current" assumption that the emissions fraction is 10%.

#### Author response

We agree with the referee in that Carpenter and Reimann (2014) state that their assumption is based on data from the 1990s. However, Carpenter and Reimann (2014) apply this assumption to reported current CFC-114 trends and mixing ratios (Table 1-1) and in fact there is no recent evidence in the peer-reviewed literature challenging this assumption.

#### Referee comment

Pg 12, Line 21: I can't find GWP(100)=8490 in Carpenter and Reimann (2014). In the 2014 Ozone Assessment, GWPs were listed in Chapter 5 (Harris and Wuebbles), where I see CFC-114 listed as 8530.

#### Author response

We have corrected the statement two-fold: 1) The GWP was reported by Hodnebrog et al., 2013, which has been added to the reference and 2) The number in the text was wrong and has been changed to 8,590 (the calculation was correct).

#### Referee comment

Figure S1: I don't know what "scen" means on the titles of the two left-most figures.

#### Author response

The sentence in the figure caption was modified to: "Left: Atmospheric time series corresponding to the modelled firn profiles (black lines, scen: scenario) in comparison to Cape Grim air archive data (blue dots)."

Minor suggestions/corrections:

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Pg. 2, Line 18: Consider using “phase out” in place of “ban”. (minor)

Pg. 2, Line 18: Even though you define consumption = production + imports – exports, and the MP does list control in terms of consumption in many cases, it is more general to say that the MP regulates “production and consumption”. This would then be consistent with the statement on pg 2, line 26.

Pg. 2, Line 20: Suggest “has started to decline” in place of “started to reduce”

Pg. 5, Line 5: Is this column commercially available? If so, please tell us where you purchased it.

Pg. 6, Line 6: Consider using “mole (mass)” in line 6, since it is used in line 7.

Pg. 8, Line 20: Suggest “. . . ratios below 4%”. Given the uncertainties, anything below 4% is clearly not well known.

Pg. 10 Line 11: Suggest “AFEAS data, which suggests rapidly increasing emissions to more than 5 Gg/year in the late 1940s, are inconsistent with our emissions estimates.”

Pg 10, Line 12: Suggest “Emission rates above 5 Gg/yr, suggested by AFEAS, are unlikely to have occurred before the mid-1950’s. . . .”

Figure 3 caption: The caption should read: “Global emissions of CFC-114 and CFC-114a derived from Cape Grim observations (solid lines), with uncertainties represented by dashed lines. The dotted lines represent emissions derived purely from firn air data.”

Author response

All changes were made as requested.

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