

Interactive comment on “Atmospheric radiocarbon measurements to quantify CO<sub>2</sub> emissions in the UK from 2014 to 2015” by Angelina Wenger et al.

Anonymous Referee #1 Received and published: 3 December 2018

*Reviewers comment: This paper aims to use <sup>14</sup>C measurements and modelling to examine fossil fuel CO<sub>2</sub> emissions in the UK. The concept is good, and I think that the results could be quite useful. Unfortunately though, this work is not yet ready for publication. The writing is so disorganized that it is not possible to assess the quality of the science. As I read through the paper, I started to make notes on specific points that were unclear. After five pages of noting my confusion with every section beyond the introduction, I gave up in frustration. I am very sorry to have to be so harsh, but this paper needs to be entirely rewritten so that the (likely very good) research can be reviewed and the meaning can become apparent.*

Authors response: The comments given on the paper are not entirely useful in providing guidance on how the paper can be improved. In response to the comments from the reviewer we have answered the examples they have given.

Changes to manuscript: We have rewritten sections of the paper to make more understandable to the reader. This included adding more sub headings, introductions to each section and more information added to improve the clarity. For example, added to section 3.3.1.:

*“For the calculation of ffCO<sub>2</sub> with Equation 4, 80‰ was used as the <sup>14</sup>CO<sub>2</sub> signature of heterotrophic respiration ( $\Delta_{HR}$ ). The mole fraction enhancement due to CO<sub>2</sub> emitted from heterotrophic respiration (CO<sub>2 HR</sub>) was derived from the NASA CASA biosphere model and atmospheric back trajectories (more details about the modelling can be found in section 3.1).*  
“

*Reviewers comment: Just to choose two examples: Section 3.2 is titled “Isotope modelling”. The content of this section is a series of equations, many of which are inappropriate or even wrong. For example,  $\delta^{14}\text{C}$  is explicitly detailed in an equation, but the equation is incorrect. Further,  $\delta^{14}\text{C}$  is never actually used in the paper, rather  $\Delta^{14}\text{C}$  is used but never explained in any equation. It is also not clear what these equations are used for. Are they used to determine simulated <sup>13</sup>C and <sup>14</sup>C values based on the model simulation? Or are they meant to show how the <sup>13</sup>C and <sup>14</sup>C content of the measurements was calculated? Whichever of these is the intent, it needs to be clear what it is that is being determined from the equations, and the equations presented should be pertinent to the results that are being shown later in the paper.*

Authors response: The reviewer has provided no input regarding what aspect of the equations they consider to be “incorrect” or inappropriate. We concede that Equation 2 might not be written in accordance with best practices. While this makes it a part of the wide problem of non-standardised isotope notations it certainly does not make it wrong. Our Equation 2 is equivalent to the early definition of the  $\delta$  value by Stuiver & Polach (1977), the second equation on page 361.

$$\delta^{14}\text{C} = \left[ \frac{{}^{14}\text{C sample} / \text{C sample}}{{}^{14}\text{C std} / \text{C std}} - 1 \right] \times 1000$$

Equation 2 from the discussion paper.

We have changed this to conform to more modern notation standards of Coplen 2011 page 2554 and added it to the supplementary material.

$$\delta^{14}\text{C} = \frac{{}^{14}\text{C sample} / {}^{12}\text{C sample}}{{}^{14}\text{C std} / {}^{12}\text{C std}} - 1$$

The definition of the  $\delta^{14}\text{C}$  was included as it is utilised in the definition of the  $\Delta^{14}\text{C}$  and also serves more generally as the definition of the small delta value. We have learnt from the reviewers comments that the inclusion of the basic definitions and equations caused confusion and hinders the flow of the reader. For this reason, we have put the definitions of the small and large delta value as well as the intermediate derivations used for the modelling in to the supplement. Only the equations directly used for simulations and calculations (Equations 1, 5, 7 and clearer version of Equation 10) remain in the main text.

*Reviewers comment:* Section 3.3 and 4.1. The discussion of biogenic and nuclear corrections looks interesting, but it is never explained where the values in Figure 2 come from. Are these model simulations? Or based on observations? Further, the values plotted in Figure 2 are not defined in the text or in any of the equations that are presented in the paper. A very knowledgeable reader would guess that these correction terms are meant to represent the bias term in equation 10, but with that bias term split out into “biogenic” and “nuclear” rather than lumped together into “other”.

*Authors response:* The Authors acknowledge the use of the correction term in Equation 10 (other) is confusing and prevents the reader from easily understanding how exactly the correction is applied. The former equation 10 (now Equation 4) has been changed to include the specific corrections that were applied in this work. The whole result section has been reorganised so the

*Changes to manuscript:* Equation 10 (now Equation 4) has been changed to be more descriptive.

$$\text{CO}_2 \text{ ff} = \frac{\text{CO}_2 \text{ bg} (\Delta_{\text{obs}} - \Delta_{\text{bg}})}{(\Delta_{\text{ff}} - \Delta_{\text{obs}})} - \frac{\text{CO}_2 \text{ hr} (\Delta_{\text{hr}} - \Delta_{\text{obs}})}{(\Delta_{\text{ff}} - \Delta_{\text{obs}})} - \frac{\text{CO}_2 \text{ nuc} (\Delta_{\text{nuc}} - \Delta_{\text{obs}})}{(\Delta_{\text{ff}} - \Delta_{\text{obs}})}$$

A short description of the nuclear and biospheric correction was added near Equation 4 and Equation 4 is referenced in both the detailed description of the corrections 3.3.1 and 3.3.2 as well as the results section and the legend of Figure 3 (old Figure 2).

*“This means that each correction can be evaluated for its impact on the final ffCO<sub>2</sub> value individually. The equation given in Turnbull et al., 2009 was adapted to have a correction term for heterotrophic*

*respiration (hr, section 3.3.1) and emissions from the nuclear industry (nuc, section 3.3.2) and is given in Equation 4.”*

*Reviewers comment:* I cannot evaluate the science in this paper until it is more clearly presented, and therefore have no choice but to recommend rejection and resubmission of a new, completely rewritten paper based on the same underlying research. I recommend that the more experienced coauthors expend some effort in assisting with the writing.

*Authors response:* The authors acknowledge that the manuscript was hard to follow. We have extensively revised and restructured the paper to facilitate the reading process. We have added more subsections, a clear introduction to each section and added more details and references in to the sentences.

*Changes to manuscript:* Examples of changes that have been made include expansion of the sections 3.3.1 and 3.3.2 (ffCO<sub>2</sub> correction terms) and simplification of the equations that are included in the main text. In addition to that, the result section is now better structured and clearly separates the result of the modelling framework from the method:

#### *4 Results*

##### *4.1 Comparison of modelled and observed data*

##### *4.2 Fossil Fuel CO<sub>2</sub> derived from $\Delta^{14}\text{CO}_2$ observations*

###### *4.2.1 Influence of the corrections applied to the ffCO<sub>2</sub> calculation*

###### *4.2.2 Results of ffCO<sub>2</sub> derived from $\Delta^{14}\text{CO}_2$ observations at TAC*

###### *4.2.3 Increase the temporal resolution of ffCO<sub>2</sub> using CO ratios?*

**Anonymous Referee #2 Received and published: 28 February 2019**

*Reviewers comment:* This paper makes interesting use of measurements of atmospheric <sup>14</sup>C in CO<sub>2</sub> in order to attempt to estimate fossil fuel emissions from the United Kingdom. This is an interesting and potentially useful approach and the publication of the data would certainly be beneficial. As far as I can gather, the methodology appears to be relatively thorough and robust. Whilst it is disappointing that the measurement uncertainty appears to prohibit a thorough understanding of the emissions, I still feel that it merits documentation. However, the manuscript in its current state feels disorganised and lacking in detail. It includes a number of vague or confusing sentences that do little to clarify the reasoning of the authors, and the reader is forced to work quite hard in order to follow the science. Unfortunately, it therefore needs a substantial rewrite before can be accepted for publication.

*Authors response:* The authors thank the reviewer for their time and their constructive remarks. The authors have taken the time to substantially reorganise the manuscript. The manuscript has been edited with a special focus on eliminating ambiguous sentences and to make it easier to read. Point per point answers to the reviewers comments are given below.

As major changes have been made to the manuscript, not all sections that were edited are included in the “Changes to the manuscript” part of the answers.

*Reviewers comment:* Whilst I have described some of the more significant problems below, I should reiterate that much of the paper is hard to follow and more detail is needed in most sections. This is a particular problem where equations have been included. Many of these are difficult put into context, and some do not even use the same variables for what I assume to be the same parameters. I found myself having to refer to the supplied references to understand the context of these equations. The jump from equation 9 to equation 10 is particularly jarring. The reader is forced to work hard to follow the logic here and so much more care is needed.

*Authors response:* We agree that the section with the equations was hard to read. We added clearer descriptions to the equations in the manuscript and took care to have consistency in the variable names. While we agree for the need to add more context to the equations in general, we removed many of the equations from the main text to ease the reading flow. Only the end version of the equations that were utilised in this work are now in the main text, while derivations are in the supplement. The equations used to calculate a parameter is now clearly referenced throughout the paper. We have changed the text to clarify why Equation 10 (now 4) was chosen to calculate ffCO<sub>2</sub>, however this merely aims to justify why the well established method developed by Turnbull et al., 2009 was used.

*Changes to manuscript:* An introduction to the “isotopic modelling” section is added to aid the reader:

*“This section describes the method and the equations used to model <sup>12</sup>CO<sub>2</sub>, <sup>13</sup>CO<sub>2</sub> and <sup>14</sup>CO<sub>2</sub> at TAC. The modelling of the two stable CO<sub>2</sub> isotopes was necessary in order to be able to simulate the <sup>14</sup>CO<sub>2</sub>. A framework to simulate <sup>14</sup>CO<sub>2</sub> was developed to have a tool investigate the observations and possible constraints of the radiocarbon method. A basic mass balance (Equation 1) was used as the basis of the modelling. Where the observed atmospheric mole fraction of CO<sub>2 obs</sub> can be described as the sum of CO<sub>2</sub> from individual sectors (CO<sub>2 obs</sub>) and a background contribution. This simple concept was adapted to the different CO<sub>2</sub> isotopes, by using the definition of the small delta (δ) value for <sup>13</sup>CO<sub>2</sub> and the definition of the large delta (Δ) <sup>14</sup>CO<sub>2</sub> as defined in Stuiver & Polach (1977). The simulated <sup>13</sup>CO<sub>2</sub> was calculated with Equation 2 and the Δ<sup>14</sup>CO<sub>2</sub> with Equation 3.”*

*“The Δ<sup>14</sup>CO<sub>2</sub> observations at TAC and MHD are used to calculate the recently added CO<sub>2</sub> from fossil fuel burning (ffCO<sub>2</sub>). This method takes advantage of the fact that fossil fuels have been isolated from other carbon pools for so long that they are completely devoid of <sup>14</sup>C, recent additions of CO<sub>2</sub> from fossil fuel burning therefore lead to a depletion in the atmospheric Δ<sup>14</sup>CO<sub>2</sub>. We followed the approach of Turnbull et al., 2009, this approach was chosen as the calculation of the uncorrected ffCO<sub>2</sub> is separated from the corrections. This means that each correction can be evaluated for its impact on the final ffCO<sub>2</sub> value individually. The equation given in Turnbull et al., 2009 was adapted to have a correction term for heterotrophic respiration (hr, section 3.3.1) and emissions from the nuclear industry (nuc, section 3.3.2) and is given in Equation 4. The reasoning behind the need for the corrections for heterotrophic respiration and emissions from the nuclear industry are explained in detail in the next two sections.”*

*Reviewers comment:* Some parts of the paper feel a little rushed and the number of mistakes included lead me to wonder if the text has been properly proof-read. For example, Figure 4 is never mentioned in the main text although it appears to be a fairly important model observation comparison.

Authors response: We agree with the reviewer that there were avoidable mistakes that were not caught in the proof read stage. More care has been taken in the editing of the revised paper. All Figures are now referred to and their meaning is more clearly described in the text.

Examples of changes to the manuscript include:

“In Figure 4 we present the results  $\text{ffCO}_2$  calculated with Equation 4 from  $\Delta^{14}\text{CO}_2$  observations at TAC station ( $\text{ffCO}_2_{\text{observed}}$ ) and compare it with modelled emissions obtained from the simulations performed in Section 3.1 ( $\text{ffCO}_2_{\text{simulated}}$ ). 1 ppm of  $\text{ffCO}_2$  causes a depletion of approximately 2.5 ‰ in  $\Delta^{14}\text{CO}_2$ . Figure 4 shows that most observed values are not significantly different from the modelled values. This implies that the  $\text{ffCO}_2$  derived from  $\Delta^{14}\text{CO}_2$  observations at TAC agrees well with the values simulated using emissions inventories and an atmospheric model (Section 3.2). However, the uncertainties associated with the observed  $\text{ffCO}_2$  is relatively large, while the  $\text{ffCO}_2$  emissions from the UK are comparatively low. This means that in the UK only very large deviations from the reported emissions in bottom up inventories would be captured by  $\text{ffCO}_2$  derived from  $\Delta^{14}\text{CO}_2$ .”

*Reviewers comment:* Some sentences, such as the first sentence of Section 3.3.1, do not make any sense.

Authors response: The authors agree with this comment, section 3.3.1 has been edited.

Changes to manuscript: Changes to beginning of section 3.3.1

“In the 1950s and 1960s extensive nuclear weapon tests caused a sudden sharp increase in the atmospheric  $^{14}\text{CO}_2$  content, this is commonly referred to as the bomb spike (Levin et al., 1980; Manning et al., 1990). This bomb  $^{14}\text{CO}_2$ , has gradually been assimilated into other carbon pools. Carbon that is exchanged from the biosphere to the atmosphere can have a different  $\Delta^{14}\text{CO}_2$  signature depending on when the carbon was originally assimilate in to the biosphere.”

*Reviewers comment:* The values discussed in the first paragraph of Section 4.1 do not seem to correlate with those shown in Figure 2.

Authors response: We would like to apologise for this oversight, the values reported in the text had not been updated to the newest version while Figure 2 had been recalculated already. We are very grateful that this mistake was spotted by the reviewer.

Changes to manuscript: The averages that are given in section 4.1 (now 4.2.1) reflect Figure 2 (now 3).

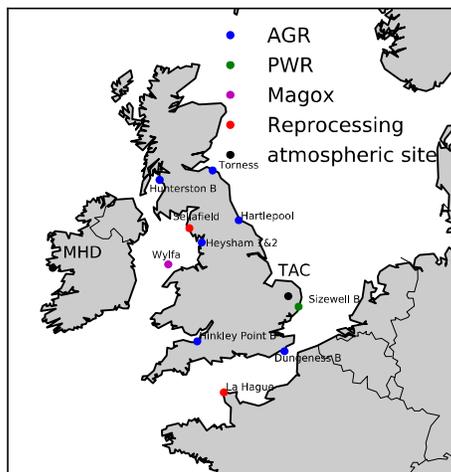
“The mean of the correction applied (over the whole study period 2014-2015) was 0.34 ppm  $\text{ffCO}_2$  equivalent for the heterotrophic respiration and 0.25 ppm for the nuclear emissions.

*Reviewers comment:* The locations of TAC and MHD are not displayed anywhere in the main paper!

Authors response: The authors agree with the reviewer that having a map with the location of the observation sites would be a positive addition. The location of MHD and TAC have now been included in the map showing the location of the nuclear power plants (Figure 1).

Changes to manuscript:

Addition of Figure 1:



“The TAC tall tower measurement site was set up in 2012 as part of the UK DECC (Deriving Emissions linked to Climate Change) network (Figure 1).”

“MHD, located on the west coast of Ireland, was used as the background site for this study and weekly sampling was performed when air masses were representative of clean air coming from the Atlantic (Figure 1).”

*Reviewers comment:* Finally, I’d suggest that just a little more justification for some of the authors’ decisions are necessary. For example, why is the 15<sup>th</sup> percentile used in order to estimate the background <sup>12</sup>CO<sub>2</sub> concentration?

Authors response: We agree that the justification for using the 15<sup>th</sup> percentile was very minimalistic, we have now slightly more extensive justification. The aim in the background choice was to get a smooth curve that represents the seasonal variability of CO<sub>2</sub> in the atmosphere and for that curve to have values that correspond to low background observations at TAC. The 15<sup>th</sup> percentile is up to a point a somewhat arbitrary choice, only the extremely low (<5) and high (>60) percentile values did not create a smooth curve. And any percentile between 10-20 fitted the low CO<sub>2</sub> concentrations in TAC very well in scope during winter and autumn.

Changes to manuscript:

“The 15<sup>th</sup> percentile of the MHD data was chosen for the background curve over other percentiles because it successfully removed short term concentration changes and pollution events. In addition

to creating a smooth curve, the 15<sup>th</sup> percentile of the MHD data was also fitted low concentrations observed in TAC well outside of the growing seasons (not much CO<sub>2</sub> uptake due to photosynthesis).”

*Reviewers comment:* Why is the CO and a concentration ratio used as a proxy for ffCO<sub>2</sub> in the forward NAME runs in the final section instead of using the EDGAR inventory to directly simulate CO<sub>2</sub>?

Authors response: This is a very good question especially since we established earlier in the paper that the use of the CO ration is not necessarily a very good proxy at TAC. We concede that using EDGAR emissions would have been more consistent with the rest of the work. Unfortunately adapting emission maps to be used in a NAME forward model is not trivial. When running the NAME model in the forward mode, theoretical particles are released in the same way as described in section 3.1 for the back trajectories. Release rates and locations corresponding to the EDGAR emissions would have had to be added for the whole modelling domain as individual point or area sources. It was decided to use a CO emission file that was already available in the correct format instead. We adapted the manuscript to explain why the CO emissions were used.

Changes to manuscript:

“A 1 year forward run was performed in NAME for both CO and <sup>14</sup>CO<sub>2</sub> (June 2012-June 2013). CO was used as a proxy for fossil fuel CO<sub>2</sub> instead of the EDGAR 2010 emissions as there was a CO emission file correctly formatted for the use in NAME available to the authors.”

*Reviewers comment:* Does a consistent 40m cut-off for the boundary layer (BL) in NAME model affect the results, or would a BL that varies with the time/season produce different footprints?

Authors response: We use 0-40 m as the range where NAME particles are influenced by the surface. This 40 m is not equivalent to the BL height. The BL height does vary with time, depending on meteorological conditions. Within the model, we have set a minimum BL of 40m, which comes into effect only under the relatively rare situation where the model-predicted BL is lower than 40m. This is done so the BL is never lower than the chosen surface influence region of 0-40m. A smaller value (than 40m) could be used as the surface influence but this would reduce the number of NAME particles interacting with the surface and cause more noise in the results. Further details are provided in earlier publications, which we have added to this line in the manuscript (Manning et al., 2011 and Arnold et al., 2018).

Changes to manuscript: References containing more detailed information and explanation have been added to the relevant passage.

“It is assumed that when a particle resides in the lowest 0 - 40 m of the model atmosphere, pollution from ground-based emission sources is added to the air parcel (Manning et al., 2011 and Arnold et al., 2018). “

*Reviewers comment:* I’d stress that the work in this manuscript appears to be good, but more care and time is necessary before it is ready for publication. I’d recommend that the authors make major revisions to the text of the manuscript, but that the paper could be accepted if these are carried out.

Authors response: We would like to thank the reviewer for their time and the fair and constructive suggestions they have given. We have made major changes to the whole manuscript and have aimed to implement all the suggestions given by the reviewer.

*Reviewers comment:* Brief suggestions: Slightly more detail about fractionation in Section 3.2. What exactly is it and why is it a problem?

Authors response: More details on fractionation have been added to section 3.2.

Changes to manuscript:

“The  $\Delta^{14}\text{C}$  is normalized to a  $\delta^{13}\text{C}$  value of  $-25\text{‰}$ , this is done to account for fractionation of the sample. Fractionation is the discrimination against one isotope in favour of the other in physical processes and chemical reactions. This discrimination takes place as the additional neutron in  $^{13}\text{C}$  alters both the weight of the carbon and their chemical bonding energies. Biological processes such as for example photosynthesis and evaporation selectively favour the lighter isotope. Fractionation effects discriminate against  $^{14}\text{C}$  twice as much as for  $^{13}\text{C}$  (Stuiver and Polach, 1977). Normalising  $\delta^{14}\text{C}$  measurements to a common  $\delta^{13}\text{C}$  should, remove reservoir specific differences caused by fractionation.”

*Reviewers comment:* Include a Figure showing an example NAME footprint for the site, and also examples of the emission distributions used with these footprints to create the simulated mole fractions.

Authors response: We agree that an example of a NAME back trajectory and an emission maps would be informative. We have added these to the supplementary material.

*Reviewers comment:* Section 3.3 should be expanded as it is currently too brief and confusing.

Authors response: We have taken this valuable suggestion on board and have changed the whole section 3.3 extensively. We have added an extent introduction to the chapter and clarified the approach chosen to calculate the  $\text{ffCO}_2$ . We feel that it is important to clarify that we did not develop a new approach to calculate  $\text{ffCO}_2$  and merely implemented the technique established in Turnbull et al., 2009. An extract of the revised section 3.3 has already been added above.

*Reviewers comment:* Also, a more detailed description of why the biospheric and nuclear corrections are necessary and how they are applied.

Authors response: We acknowledge that investigating the corrections needed for the  $\text{ffCO}_2$  calculation are an important part of this work and a more detailed description would be helpful to the reader. We have expanded section 3.3.1 and 3.3.2 and clarified how exactly they were calculated. Adding the explicitly used correction terms to Equation 4 should clarify how exactly the correction was implemented.

These sections 3.3.1 and 3.3.2 now read:

“3.3.1 Biospheric correction

In the 1950s and 1960s extensive nuclear weapon tests caused a sudden sharp increase in the atmospheric  $^{14}\text{CO}_2$  content, this is commonly referred to as the bomb spike (Levin et al., 1980; Manning et al., 1990). This bomb  $^{14}\text{CO}_2$ , has gradually been assimilated into other carbon pools, a Figure showing this gradual mixing can be found in the supplement. Carbon that is exchanged from the biosphere to the atmosphere can have a different  $\Delta^{14}\text{CO}_2$  signature depending on when the carbon was originally assimilated into the biosphere. To account for this, biospheric emissions were split into two sources, autotrophic and heterotrophic. Autotrophic respiration of plants generally contains recently assimilated carbon (<1 year). Therefore,  $^{14}\text{CO}_2$  from autotrophic respiration is generally assumed to be in equilibrium with the atmosphere. While recent work has indicated that autotrophic respiration may also contain older carbon (Phillips et al., 2015), it is assumed to be negligible for this work. Heterotrophically respired  $\text{CO}_2$  contains carbon from older pools (for example decaying biomass) and can be significantly enriched in  $^{14}\text{C}$  compared to current atmospheric  $\text{CO}_2$  (Naegler and Levin, 2009). To simulate the  $\Delta^{14}\text{CO}_2$  from heterotrophic respiration, the 1-box model developed by (Graven et al., 2012) was used, it is assumed that two-thirds of heterotrophic respiration originates from older carbon pools. This resulted in a  $\Delta^{14}\text{CO}_{2\text{hr}}$  of 67-91‰ for 2014-2015. For the calculation of  $\text{ffCO}_2$  with Equation 4, 80‰ was used as the  $^{14}\text{CO}_2$  signature of heterotrophic respiration ( $\Delta_{\text{hr}}$ ). The mole fraction enhancement due to  $\text{CO}_2$  emitted from heterotrophic respiration ( $\text{CO}_{2\text{hr}}$ ) was derived from the NASA CASA biosphere model and atmospheric back trajectories (more details about the modelling can be found in section 3.2). A similar disequilibrium exists with between the atmosphere and the ocean, but it was considered negligible for this work.

### 3.3.2 Nuclear correction

Radiocarbon emissions from nuclear reactors have a large temporal variability, making them difficult to correct for. Although the emissions are small, they have a  $\Delta^{14}\text{C}$  value of  $\sim 10^{15}\text{‰}$ , and can, therefore, influence radiocarbon observations significantly. During the study period, 3 types of nuclear power plants were in operation in the UK (Figure 1). Of these, both the AGR and the Magnox Reactor are cooled with  $\text{CO}_2$  gas. This creates an oxidising condition in the reactor, resulting in the majority of the released  $^{14}\text{C}$  being released in the form of  $^{14}\text{CO}_2$ . It is produced in the reactor from reactions of neutrons with  $^{14}\text{N}$ ,  $^{13}\text{C}$ ,  $^{17}\text{O}$ . Most of the  $^{14}\text{CO}_2$  emitted from the AGRs and Magnox plants originates from  $\text{N}_2$  impurities in the cooling gas (Yim and Caron, 2006). The UK also has one running pressurised water reactor (PWR), Sizewell B (52.21 °N, 1.62 °E), in the east of England. They contain a reducing reactor environment, leading to  $^{14}\text{C}$  being released predominantly in the form of  $^{14}\text{CH}_4$ . As  $^{14}\text{C}$  is constantly produced in nuclear reactors, parameterized emissions (an average emission factor per plant type that is multiplied with the power production of a plant) are a good approximation. However, the production of  $^{14}\text{C}$  is highly dependent on the number of impurities present in the reactor and only a small part of the produced  $^{14}\text{C}$  is ever emitted. Emissions can be caused by leakage as well as operational procedures, known as blowdown events. Reported emissions are therefore more informative. To apply a correction for these nuclear industry emissions in the calculation of  $\text{ffCO}_2$  in Equation 4,  $7.3 \times 10^{14}\text{‰}$  was used as the  $\Delta_{\text{nuc}}$ . To calculate the mole fraction of  $\text{CO}_2$  derived from the nuclear industry ( $\text{CO}_{2\text{nuc}}$  in Equation 4) atmospheric back trajectories were multiplied with a  $^{14}\text{CO}_2$  emission map of reported nuclear industry emissions that was especially created for this study. This  $^{14}\text{CO}_2$  emissions map was created with the highest frequency data available from each nuclear site. Monthly atmospheric emission data were provided by the two operators of the ten UK nuclear power plants; EDF and Magnox Ltd. Data for the of other seventeen UK nuclear sites were taken from the annual Radioactivity in Food and the Environment RIFE, 1995-2016 (Environment Agency, Natural Resources Wales, 2017). The emissions from other European nuclear power plants were sourced from annual environmental reports if available (France,

Germany) otherwise parameterized emissions were calculated according to (Graven and Gruber, 2011b). The largest emitter of  $^{14}\text{C}$  during the study period was the nuclear fuel reprocessing site in La Hague, Northern France (49.68 °N, 1.88 °W). For La Hague, monthly emission data reported on their website were utilised and is included in the supplementary material (S3).”

*Reviewers comment:* Is Figure 4 unnecessary due to the inclusion of Figure 3? If so, remove it!

Authors response: The Authors believe both figures add to the value of the work. Figure 3 (now Figure 2) investigates how the  $^{14}\text{CO}_2$  simulations established in section 3.2 compare with the  $\Delta^{14}\text{CO}_2$  observations taken at TAC. This gives an indication on whether the modelling framework that was chosen in section 3.2 is appropriate. While Figure 4 compares the  $\text{ffCO}_2$  in TAC with the emissions from the EDGAR inventory. Figure 4 shows not only that there is no significant difference between the calculated  $\text{ffCO}_2$  and the emission inventory but also illustrates that the measurement uncertainty in the  $\text{ffCO}_2$  calculation clearly limits the use of the usefulness of the radiocarbon method at TAC. We have aimed to make this distinction between the two figures clearer by expanding description of them in the text.

Changes to manuscript:

“For this work  $^{12}\text{CO}_2$ ,  $\delta^{13}\text{CO}_2$  and  $\Delta^{14}\text{CO}_2$  were simulated using Equation 1, 2 and 3 at TAC and are compared with observations in Figure 2. Daily mean values are displayed for both the modelled (blue line) and the observed data (black line, points). The uncertainty estimate (light blue area) includes the baseline uncertainty as well as the emission inventory uncertainty.”

“In Figure 4 we present the results  $\text{ffCO}_2$  calculated with Equation 4 from  $\Delta^{14}\text{CO}_2$  observations at TAC station ( $\text{ffCO}_2^{\text{observed}}$ ) and compare it with modelled emissions obtained from the simulations performed in Section 3.1 ( $\text{ffCO}_2^{\text{simulated}}$ ). 1 ppm of  $\text{ffCO}_2$  causes a depletion of approximately 2.5 ‰ in  $\Delta^{14}\text{CO}_2$ . Figure 4 shows that most observed values are not significantly different from the modelled values. This implies that the  $\text{ffCO}_2$  derived from  $\Delta^{14}\text{CO}_2$  observations at TAC agrees well with the values simulated using emissions inventories (EDGAR 2010) and an atmospheric model (Section 3.2). However, the uncertainties associated with the observed  $\text{ffCO}_2$  is relatively large, while the  $\text{ffCO}_2$  emissions from the UK are comparatively low. This means that in the UK only very large deviations from the reported emissions in bottom up inventories would be captured by  $\text{ffCO}_2$  derived from  $\Delta^{14}\text{CO}_2$ .”

*Reviewers comment:* For Figure 5, it might be clearer to colour the winter measurements differently from those made during the rest of the year, as these are specifically referred to in the text.

Authors response: The reviewers idea to change the figure to visually distinguish the observations made in winter from the other season is very good and has been implemented. The CO ratio discussion has now also been divided in to a separate subsection (4.2.3). The discussion about the two very depleted samples in November 2014 has also been removed from this section. As those two samples in November 2014 have previously been excluded from analysis in section 4.2.2. This was done as including them in the discussion did not add much scientific value and only increased the complexity of the text.

“Figure 5 shows the CO<sub>enh</sub> in TAC versus the observed ffCO<sub>2</sub> from the **radiocarbon method**, a list of the results can be found in Table 2. The median CO<sub>enh</sub>/ ffCO<sub>2</sub> ratio was 5.7 (2.4-8.9) ppb ppm<sup>-1</sup>, with a median R<sup>2</sup> correlation coefficient of 0.50. The CO<sub>enh</sub>/ ffCO<sub>2</sub> ratio is often described as more robust in winter because the fossil fuel fluxes are larger, minimising the influence of CO from biogenic sources. Restricting the analysis to include only samples taken in winter results in a the CO<sub>enh</sub>/ ffCO<sub>2</sub> ratio of 4.7 (1.0-10.1) ppb ppm<sup>-1</sup>, with a median R<sup>2</sup> of 0.7 (0.1-1.0). It is assumed that the higher variability in the CO<sub>enh</sub>/ ffCO<sub>2</sub> ratio calculated from samples taken in winter only compared to the ratio obtained from all values is due to the lower amount of data points taken in winter rather than a genuinely higher variability of the CO<sub>enh</sub>/ ffCO<sub>2</sub> ratio at TAC in winter. The CO<sub>enh</sub>/ ffCO<sub>2</sub> ratio where all data points are used (5.7 ppb ppm<sup>-1</sup>) is similar to the ratio obtained by the model (5.1 ppb ppm<sup>-1</sup>) for the TAC site.”

*Reviewers comment:* All equations to be checked for consistency and fully explained.

Authors response: We have aimed to ensure a consistent labelling of the equations. All key equations used in the work have been included in the main text. The equations that are used to derive those key equations were moved in to the supplementary material to improve the structure and clarity of the main text.

Changes to the manuscript:

“We separate CO<sub>2</sub> mole fractions at time t (CO<sub>2,t</sub>) into a background concentration (CO<sub>2,bg,t</sub>) and a contribution from each source i:

$$CO_{2,t} = CO_{2,bg,t} + \sum_i CO_{2,i,t} \quad (1)$$

A basic mass balance (Equation 1) was used as the basis of the modelling. Where the observed atmospheric mole fraction of CO<sub>2,obs</sub> can be described as the sum of CO<sub>2</sub> from individual sectors (CO<sub>2,obs</sub>) and a background contribution. This simple concept was adapted to the different CO<sub>2</sub> isotopes, by using the definition of the small delta (δ) value for <sup>13</sup>CO<sub>2</sub> and the definition of the large delta (Δ) <sup>14</sup>CO<sub>2</sub> as defined in Stuiver & Polach (1977). The simulated <sup>13</sup>CO<sub>2</sub> was calculated with Equation 2 and the Δ<sup>14</sup>CO<sub>2</sub> with Equation 3. . A detailed description on how Equation 2 and Equation 3 were derived can be found in the supplementary material.

$$\delta^{13}CO_2 = \left( \frac{\sum \left( \left( \frac{\delta^{13}CO_{2,i} + 1}{1000} \right) \times {}^{12}CO_{2,i} \times {}^{13}R_{ref} \right) + {}^{13}CO_{2,bg}}{{}^{12}CO_2} \right) \frac{{}^{13}R_{ref}}{{}^{13}R_{ref}} - 1 \times 1000 \quad (2)$$

Here, δ<sup>13</sup>CO<sub>2,i</sub> is the <sup>13</sup>CO<sub>2</sub> signature of sector i [‰], <sup>13</sup>CO<sub>2,bg</sub> is the background <sup>13</sup>CO<sub>2</sub> abundance from the rolling (± 30 days) median values of the MHD observations, CO<sub>2,i</sub> = abundance CO<sub>2</sub> from sector i [mol mol<sup>-1</sup>] as simulated in TAC (Equation 1), <sup>13</sup>R<sub>ref</sub> is the ratio of reference standard [(mol mol<sup>-1</sup>)/(mol mol<sup>-1</sup>)] and CO<sub>2</sub> is the total abundance CO<sub>2</sub> enhancement [mol mol<sup>-1</sup>] from Equation 1.

$$\Delta^{14}CO_2 = \left( \frac{\sum \left( \frac{\left( \frac{\Delta^{14}CO_{2i} + 1}{1000} \right) \times {}^{14}R_{ref}}{1 - 2 \times \frac{25 + \delta^{13}C_i}{1000}} \right) \times CO_{2i}}{CO_2} \times \left( 1 - 2 \times \frac{25 + \delta^{13}CO_2}{1000} \right) - 1 \right) \times 1000 \quad (3)$$

Where,  $\Delta^{14}CO_{2i}$  is the  $^{14}CO_2$  signature of sector  $i$  [‰],  $^{12}CO_{2i}$  is the abundance  $CO_2$  from sector  $i$  [mol mol<sup>-1</sup>] from Equation 1,  ${}^{14}R_{ref}$  is the ratio of reference standard [(mol mol<sup>-1</sup>)/(mol mol<sup>-1</sup>)],  $^{12}CO_2$  is the total abundance  $CO_2$  enhancement [mol mol<sup>-1</sup>] from Equation 1 and  $\delta^{13}CO_2$  is the  $^{13}CO_2$  signature [‰] from Equation 2.