“Detecting long-term changes in point source fossil CO_2 emissions with tree ring archives”

E.D. Keller et al.

Response to Referee Comments

Thank you to both referees for your comments. Each comment is copied below in red, and our response appears immediately after in black. Changes made to the text appear in italicised blue.

Comments from Anonymous Referee #1:

1. The authors describe the site to be located in a flat terrain dominated by Mount Taranaki. They also point out that the wind direction and speed can be very different at sites only a few kilometers apart due to the mountain influence on atmospheric flow. Hawera is considered to be representative of Kapuni in the manuscript, but comparisons of wind speed and direction were made for a very short time interval ” 14 August-26 October 2012, with some significant data gaps” as specified by the authors (page 9, line 1). I doubt that two months of measurements (with significant gaps) are sufficient to consider the two locations similar from this prospect. Also, the wind speed at Hawera is approximately double compared to the wind speed at Kapuni (not “slightly higher speeds” as mentioned on page 9, line 10). I wonder how much does this affect the model results? It is not clear to me what data is used in Fig. 2 for the wind rose “Kapuni 2013”.

Our response also addresses the following comment from Anonymous Referee #2:

Comments: I have questions about the meteorology used for the modeling. You compare the limited data set at Kapuni, close to the sampling site, with the much more complete set at Hawera, 20 km southwest of the sampling site. You state that the correlations between wind speed and wind direction between the two sites are consistent enough to warrant using the complete Hawera data set, as shown in a direction comparison for limited dates during August-October 2012 (Fig. S2). But is the limited period in 2012 adequate for evaluating whether Hawera data are appropriate for modelling wind transport at the Kapuni site? Moreover, Figures 2 and S1 show that the wind speed at Hawera (6-7 m/s) averages on the order of twice that at Kapuni (2-3 m/s). Have you done any sensitivity calculations to see how this difference in wind speed affects the modeling? The wind directions seem to be fairly consistent at the two sites.

Thank you to both referees for raising this issue. We have re-examined all of the wind data that is available to us from Kapuni, and in particular the data from 2013-14 that is shown in Fig. 2 and Fig. S1. This data came from a temporary automated weather station (AWS) that was installed in Sep 2013 at the Shell Todd Oil Services (STOS) gas production station, adjacent to the north side of the

Vector Gas Plant. It was hired from and maintained by a third party and removed in Dec 2014, and we unfortunately do not have any record of its calibration or maintenance. We compared several other independent datasets (the 10-minute wind speed and direction from the temporary weather station installed by the authors between 14 Aug – 26 Oct 2012 that is already mentioned in the manuscript, daily mean wind speeds available through New Zealand's Virtual Climate Station Network (VCSN; Tait et al., 2006), 10-minute wind speed from an AWS installed by Vector on-site at the Kapuni Gas Plant covering Aug 2012, Oct 2012, Nov 2013, and Sep-Dec 2014, and two weeks of measurements from a sonic anemometer installed by the authors in a nearby paddock at Kapuni in Oct 2014), in which the mean wind speeds at Kapuni are on average only 80-90% lower than those at Hawera and, where there is overlap, are in disagreement with the wind speeds in the STOS dataset. Consequently, we believe that the wind speeds from the STOS AWS are biased low, and the true relationship between the wind speeds at Hawera and Kapuni is close to that derived from our data measured between Aug and Oct 2012. (We are confident of the quality of this data because we installed the weather station and verified the instrument calibration ourselves.) The wind direction is for the most part consistent in all data sources.

The low wind speeds measured at the STOS AWS could be due to either poor instrument calibration or the placement of the station itself. We emphasize that we did not use the STOS dataset in our modelling, but only for general comparison with the data from Hawera. Because we now doubt the accuracy of the wind speeds from the STOS dataset, we have replaced the data in Fig. 2b with our dataset from Aug-Oct 2012, and also added a wind rose from Hawera during the same time period for direct comparison. As more evidence of the similarity of Kapuni and Hawera, we have added a comparison of daily mean wind speeds from the VCSN, and have included a histogram from this dataset as Fig. S1. The text has been edited to remove all mention of the STOS dataset. We acknowledge that the correlation is still based on a very limited dataset, and that this is a potential source of error in our results. However, we have no reason to think that these months were atypical of conditions at Kapuni; other months and seasons for which we have data follow the same general patterns.

We did perform a sensitivity test of the effect of wind speed on the modelled results. There is an inverse proportional relationship between wind speed and modelled concentrations, so that halving the wind speed approximately doubles the modelled CO_2ff concentrations. If the wind speeds at Kapuni are significantly lower than those at Hawera, our results would be an under-prediction. The statement "slightly higher speeds" to which Referee #1 referred is based on model II linear regression performed on the overlapping dataset between 14 Aug – 26 Oct 2012. This equation is $y = 0.90x - 0.32$, where x is wind speed at Hawera and y is wind speed at Kapuni (calculated using R package lmodel2 major axis regression). This equation is now mentioned in the text and the caption of Fig. S2. The additional data sources that we have examined show a similar relationship, and we maintain that wind speed and direction at Hawera is similar enough to Kapuni for this study, which does not depend on exact point-by-point correlation. The revised text reads (p. 9 line 8 - p. 10 line 11):

The area to the northwest of Hawera and Kapuni is dominated by Mount Taranaki, a 2518m volcanic cone that rises steeply from relatively flat surrounding terrain. Wind direction and speed can be very different at sites only a few kilometres apart because of the local impact of the mountain on atmospheric flow. Thus we compared Hawera and Kapuni meteorological datasets to ensure that Hawera is representative of Kapuni over long (~1 year) time periods and the wind speed and direction distributions as a whole are similar at both locations. ~~A wind rose for the eight years (2004-2011) of data at Hawera is shown in Fig. 2, together with a wind rose for one year (2013) of data at Kapuni.~~ Daily mean wind speeds were compared using the Virtual Climate Station Network (VCSN; Tait et al., 2006). This is a set of "virtual" weather stations that uses re-analysis interpolation techniques to provide historical daily weather variables on a 5 x 5 km grid across New Zealand. The mean wind speed at Hawera over the modelled time period, 5.0 m s^{-1}, is only slightly higher than that at Kapuni, 4.6 m s^{-1}. Histograms comparing the wind speed distributions at both sites are in Fig. S1. ~~Wind speeds are on average higher at Hawera, but the distribution in direction is very similar, with a small overrepresentation of northerlies at Hawera. The wind speed and direction distributions at both locations are shown in more detail in Fig. S1.~~

~~We demonstrate correlation between the two sites using the only~~ Only one overlapping dataset with sub-daily time intervals ~~that~~ was available for direct comparison at the time of ~~the~~ our study. We collected data at a temporary meteorological station situated in a paddock at Kapuni at 10-minute intervals during the period 14 August – 26 October 2012, with some ~~significant~~ data gaps (Turnbull et al., 2014). These were averaged to hourly intervals and compared with the corresponding set of measurements at the Hawera AWS. Only daylight hours were included for consistency with the model simulations. Wind roses for the Kapuni dataset and the corresponding time period at Hawera are shown in Figs. 2b and 2c. The distribution in direction is similar to the north, but there are more southerlies and fewer westerlies at Hawera. Using these datasets, correlation in wind speed is good, with R^2 = 0.82, and correlation in wind direction is moderate (R^2 = 0.61). Because wind direction is an angular measurement, correlation in wind direction was performed using the circular package v0.4-7 in R v3.0.2 (Lund and Agostinelli, 2013; R Core Team, 2013) rather than the standard linear correlation function. Scatter plots comparing wind speed and direction at Kapuni and Hawera directly at each time step are in Fig. S2. Wind speed is a good match, with Hawera on average having slightly

higher speeds than Kapuni. *When wind speed at Hawera is linearly regressed against wind speed at Kapuni, the resulting equation is y = 0.90x – 0.32. (Model II regression was performed with the lmodel2 v1.7-2 package in R v3.0.2 (Legendre, 2014)).* *With wind direction, most points are close to the 1:1 line or slightly below, indicating a small rotation in direction between the sites. Approximately 67% of data points (one sigma) are within 30° of each other, and 85% are within 45°. For the purpose of our simulation in which we focus on integrated averages rather than particular points in time, the Hawera dataset is sufficiently representative of typical conditions at Kapuni.* *We note, however, that the dataset from Kapuni spans a very limited time period, and this is a potential source of error in our results.*

References added:

Tait, A., Henderson, R., Turner, R., and Zheng, X. G.: Thin plate smoothing spline interpolation of daily rainfall for New Zealand using a climatological rainfall surface, Int. J. Climatol., 26, 2097-2115, 2006.

Revised Figure 2:

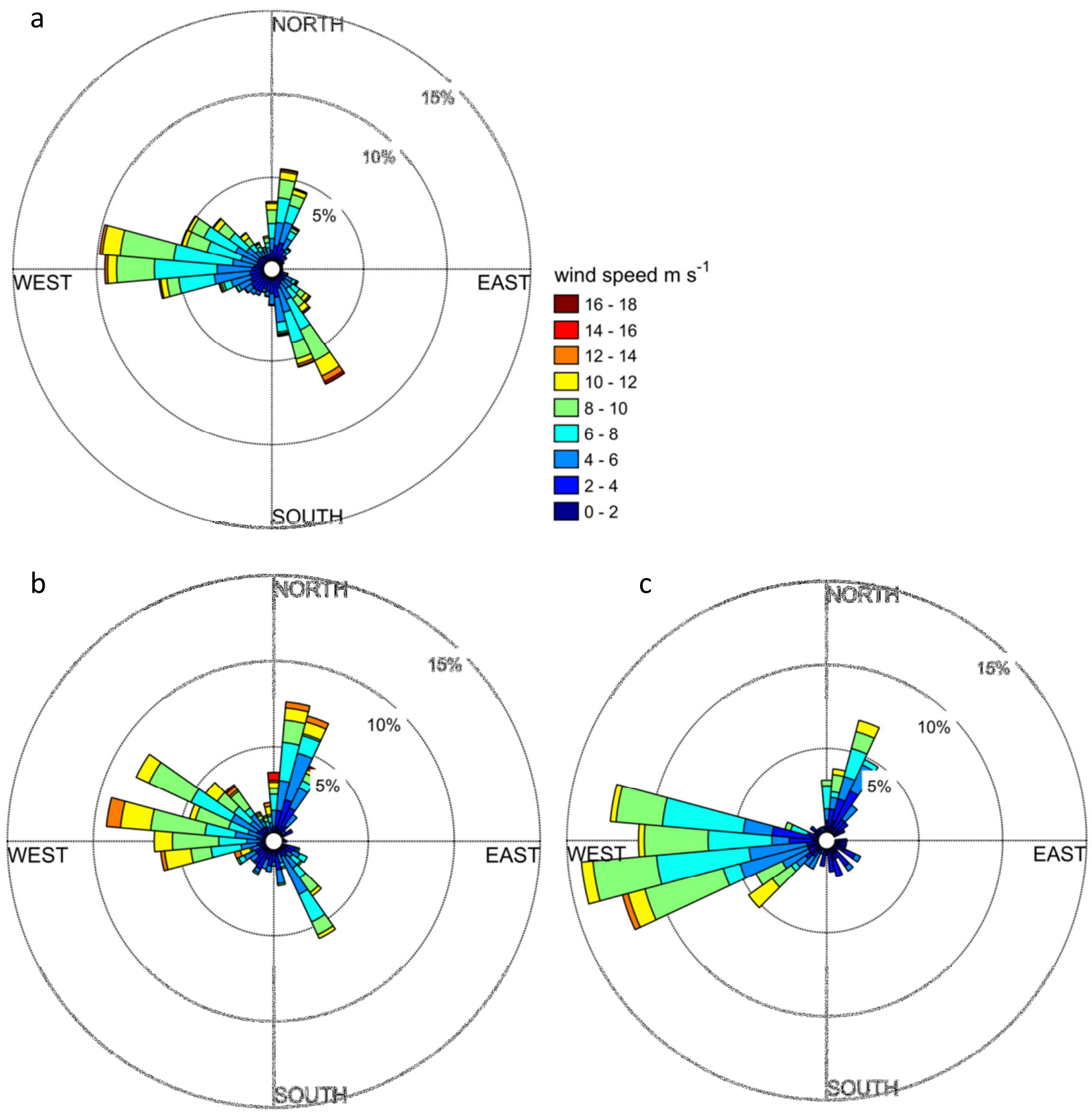


Figure 2. Wind roses at hourly intervals a) at Hawera during the eight growing season (Sep-Apr) between 2004-2011, b) Hawera 14 Aug – 26 Oct 2012, and c) Kapuni 14 Aug – 26 Oct 2012, all showing daylight hours only (8:00am – 6:00pm). Wind speed is in m s^{-1}. Data at Kapuni was collected at 10-minute intervals and averaged to hourly intervals to match Hawera data.

Revised Figure S1:

a

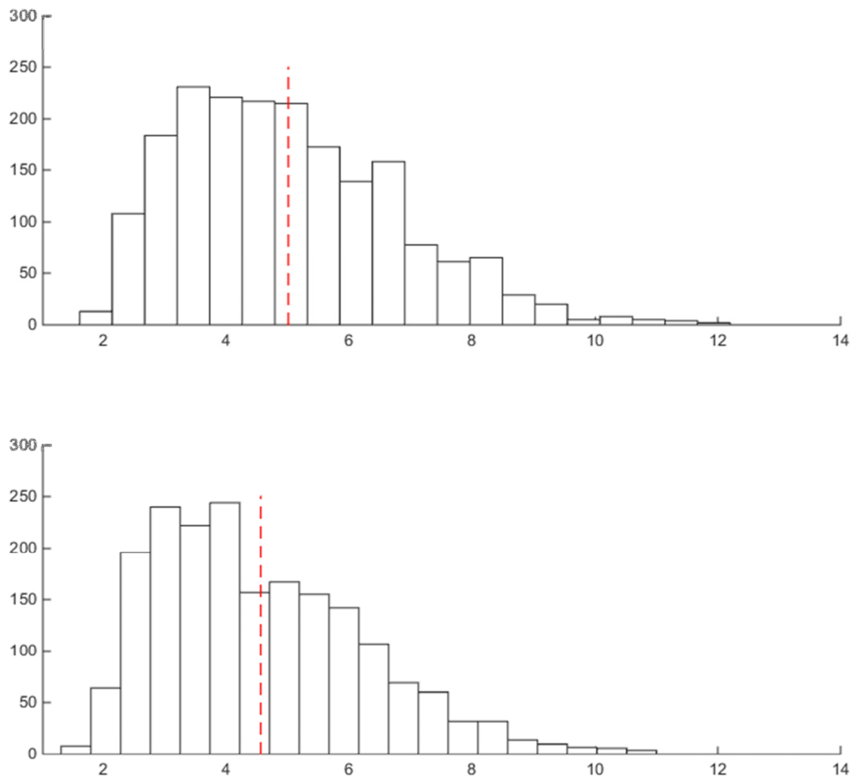
b


Figure S1. Histograms of daily mean wind speeds (m s^{-1}) at Hawera (a) and Kapuni (b) for the eight growing seasons 2004-2011 from the VCSN. Dashed red line shows the mean over the entire period (5.0 and 4.6 m s^{-1} for Hawera and Kapuni, respectively).

2. It is not clear from the text how many trees were sampled from each species. How many replicates were used and how was the data analyzed? Please specify and expand this paragraph (page 6, lines 9-24) rather than pointing to papers only. The reader should have a clear idea about the tree ring observation methodology without reading Norris, 2015 and Turnbull et al, 2014.

Additional details about the tree ring measurements have been added. The equation in Turnbull et al., 2014 used to derive CO_2ff from the measurements has also been added. The revised text reads (p. 6 line 12 – p.7 line 10):

In summary, wood was sampled from the trees using a Haglöff incremental borer. Four cores were extracted per tree at equidistant points at a height of approximately 1.2m from the base of the tree. One core from each tree was used to create a historic record of CO_2 emissions from commission of the Kapuni plant in 1971 to the outermost ring at the time of sampling in 2012. Replicates were taken from a second core to validate ring counting and ^{14}C results. Alpha cellulose was extracted *from individual rings using a method modified from Hua et al. (2000), combusted with a Europa ANCA elemental analyser (EA), reduced to graphite and measured by accelerator mass spectrometry at GNS Science laboratories in Lower Hutt, New Zealand (Baisden et al., 2013; Zondervan et al., 2015; Turnbull et al., 2015).*

CO_2ff was determined following Turnbull et al. (2014) from the isotopic difference between the measured tree ring and clean air background CO_2 measured at Baring Head, Wellington (41.4167°S, 174.8667°E; Currie et al., 2011; extended with unpublished data ~~dataset to 2015 will be presented in an upcoming publication~~). Baring Head, located at the southern end of New Zealand's North Island and approximately 300 km south of Kapuni, was chosen as the background for this study over more local sites because it provides a long-term record of background CO_2 and ^{14}C, dating back to the early 1970s. The following equation was used:

$$C_{ff} = \frac{C_{obs}\left(\Delta_{obs} - \Delta_{bg}\right)}{\left(\Delta_{ff} - \Delta_{bg}\right)} - \beta \qquad (1)$$

where C_{ff} is CO_2ff, C_{obs} is the CO_2 mole fraction in the observed sample, Δ_{obs} and Δ_{bg} are the $\Delta^{14}C$ of the observed sample and background sample, respectively. Δ_{ff} is the $\Delta^{14}C$ of CO_2ff, and is assigned to be -1000‰. Δ_{bg} is from the summer season average from the long-term Wellington $^{14}CO_2$ record at Baring Head. Comparison of this record with tree rings collected 3 km upwind of our source showed no difference from the Wellington record. β is a small correction to account for the fact that the $\Delta^{14}C$ of CO_2 from other sources may be slightly different from that of the atmosphere; in our case we set β to zero since the proximity to the coast and consistent winds suggest that CO_2other is negligible in this location (Turnbull et al., 2014). ~~Baring Head, located at the southern end of New Zealand's North Island and approximately 220 km southeast of Kapuni, was chosen as the background for this study over more local sites because it provides a long-term record of background CO2 and 14C, dating back to the early 1970s. Background levels in tree rings measured at a site in Kapuni 2km upwind of the Vector plant are close to those measured at Baring Head in the same time period, justifying the use of the Baring Head dataset (Norris, 2015).~~ Uncertainty in CO_2ff is dominated by $\Delta^{14}C$ measurement uncertainty in both background and the observed sample and is typically ~1ppm for this dataset.

References added:

Baisden,W. T., Prior, C. A., Chambers, D., Canessa, S., Phillips, A., Bertrand, C., Zondervan, A., and Turnbull, J. C.: Radiocarbon sample preparation and data flow at Rafter: accommodating enhanced throughput and precision, Nucl. Instrum. Meth. B, 294, 194–198, 2013.

Hua, Q., Barbetti, M., Jacobsen, G.E., Zoppi, U. and Lawson, E.M.: Bomb radiocarbon in annual tree rings from Thailand and Australia, Nucl. Instrum. Meth. B, 172, 359-365, 2000.

Zondervan, A., Hauser, T.M., Kaiser, J., Kitchen, R.L., Turnbull, J.C. and West, J.G.: XCAMS: The compact 14 C accelerator mass spectrometer extended for 10 Be and 26 Al at GNS Science, New Zealand, Nucl. Instrum. Meth. B, 361, 25-33, 2015.

3. When describing the model, the authors state that this is appropriate for estimating emission rates from a source over short distances (page 7, line 4). They also show that the time interval recommended for the meteorological observations used for the model is 10-30 min. How reliable are the model results for these simulations given that one hour time-step was used for wind speed/direction?

This information comes from Flesch et al., 2004, which provides detailed analysis of the effect of averaging time on WindTrax model results. The model is built on a traditional Monin–Obukhov similarity theory (MOST) description of the atmosphere and relationships derived from 15-60 min wind statistics. WindTrax is limited by the fact that large-scale atmospheric dispersion fluctuations are not incorporated in the model structure. As the time interval increases, large-scale motions become more important, and Flesch et al., 2004 states that applying the model to “time averaging periods greatly different from 15-60 min carries a risk” and an increase in error. While the preferred choice of time step is given as 10-30 minutes, 60-min time intervals are still in the range considered valid for application of MOST statistics. We believe that using a one-hour time step in this context does not make the model unreliable. We have edited the text to clarify this point and more accurately reflect the language in the original reference (p. 8 lines 7-12):

It assumes wind and other meteorological observations are averaged over a suitable time interval representing a stable, mean atmospheric state (model relationships are built from wind statistics over 15-60 minute intervals; ~~10-30 minute intervals are recommended~~ using model time steps greatly outside of this range is not recommended). Intervals longer than one hour ~~have been shown to~~ can be problematic (Flesch et al., 2004) because at these time intervals, large-scale fluctuations not described by MOST statistics become important.

Also, the chestnut tree is located at the limit of the simulation capability, 1 km. How does this influence the result?

The referee correctly points out that the chestnut tree is located at the limit of WindTrax’s capability. We discuss this in sections 3.1 and 3.3, attributing the large errors and high detection thresholds at least in part to the tree’s distance from the point source. The small concentrations combined with the large model error make this distance impractical for detecting changes in CO_2 emissions. We have not made any changes to the text.

4. The authors present this method to be useful for verifying emission changes at other locations where the point sources are much stronger, mentioning that there are approximately 800 power plants worldwide that emit more than 10 times the annual total CO_2ff at Kapuni (page 18, line 17). They also explain that "WindTrax is not applicable to complex terrain or larger distance scales and caution is urged when applying our methodology to other sites". I have a feeling that Kapuni site is very specific and I am not sure that there are so many other sites with flat terrain, trees within 300-600m of the point source located downwind, and consistent winds through time. What other model would then be most suitable for complex terrain and larger distances? Add suggestions for other model(s) that would be suitable in this case.

We acknowledge that the Kapuni site is somewhat unique in this respect. As requested, we have added a paragraph at the end of section 3.4 discussing the advantages that the Kapuni site offers with regards to atmospheric transport modelling and have listed several other models that are applicable at larger distance scales and with more complex terrain and that would be more appropriate for regional-scale studies. The added text reads (p. 19 lines 23-30):

The Kapuni site has several advantages that simplify the modelling component of this method: the terrain is flat, and there are trees conveniently located close to the CO_2ff sources. With larger distance scales and/or more complex terrain, WindTrax might not be an appropriate choice of model. Alternative atmospheric transport models that are applicable to larger distances (hundreds of kilometres and/or regional scales) and more complicated geographic features include CALPUFF (Scire et al., 2000), WRF-CHEM (Grell et al., 2005), and AERMOD (Cimorelli et al., 2005). While these models would need to be tested in the context of our method, the same general principles would apply.

References added:

Cimorelli, A. J., Perry, S. G., Venkatram, A., Weil, J. C., Paine, R. J., Wilson, R. B., Lee, R. F., Peters, W. D., and Brode, R. W.: AERMOD: A Dispersion Model for Industrial Source Applications. Part I: General Model Formulation and Boundary Layer Characterization, J. Appl. Meteor., 44, 682–693, doi: http://dx.doi.org/10.1175/JAM2227.1, 2005.

Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmos. Environ., 39, 6957-6975, 2000.

Scire, J. S., Strimaitis, D. G., and Yamartino, R. J.: A user's guide for the CALPUFF dispersion model, Earth Tech, Inc, Concord, Massachusetts, USA, 2000.

Specific comments:

Check the table captions. Information is missing (e.g. Table 2 – column 4 not explained).

All table captions have been expanded and the requested information has been added to the Table 2 caption:

Table 2. ~~Eight-year~~ Modelled mean CO_2ff and standard deviation (SD) of eight hypothetical sensors ~~for~~ simulated over the eight years 2004-2011 with ~~of~~ constant emissions. Measurement uncertainty (MU) of 1.0ppm is added to the standard deviation in the fourth column. ~~simulation and~~ Columns 5-10 show the detection limits calculated at the two-sigma (95%) and one-sigma (68%) confidence level (CL) for samples representing an average of one, two, or four years. Measurement uncertainty (MU) of 1.0ppm is added in quadrature to the standard deviation of modelled CO_2ff before limits are calculated.

Check figure captions. The reader should understand what those figures represent without reading the text.

Figure captions have been expanded where possible.

Figure 2: name the two panels a) and b) and refer to them in text accordingly. Expand the caption.

Done.

Figure 4: Same as for Fig. 2.

Done.

Page 5, line 18: 2008 should be 2007.

Thank you for catching this error.

Page 17, lines 26-28: "Indeed, looking at the results in Fig. 4, there is no significant decline at the chestnut tree in 2007; there is a small decline in CO_2ff at the pine tree but it is too small to conclude that emissions have changed. " As I estimate from the figure, the observed value is smaller in 2007 than in 2006 at the chestnut tree by 1.3 ppm, and by 0.3 ppm at the pine tree. Isn't the former significant?

The referee is correct that the 2007 observed value at the chestnut tree is lower than the previous year by 1.3 ppm. However, the meaning of the words "significant decline" in this context refers to the decline relative to the long-term mean (which is 2.1 ppm for the chestnut tree). With respect to the long-term mean, the decline in 2007 is only 0.4 ppm (or 19% of the mean), which is not enough to declare it statistically significant. We have added specific numbers to the text to clarify this point (p. 17 lines 20-25):

For a one-year observation from the pine tree, this is 18%; for the chestnut, it is 92%. The largest change in emissions in any single year at the Vector plant is in 2007, with a decline of 14% relative to the long-term mean, still below the detection limit. Indeed, looking at the results in Fig. 4, ~~there is no significant~~ the decline (0.4ppm, or 19% of the mean) at the chestnut tree in 2007 is not significant; there is also a small decline (0.7ppm, or 13% of the mean) in CO_2ff at the pine tree but it is again too small to conclude that emissions have changed.

I recommend using the same scale for the two graphs.

Done.

Comments from Anonymous Referee #2:

Comments: I have questions about the meteorology used for the modeling. You compare the limited data set at Kapuni, close to the sampling site, with the much more complete set at Hawera, 20 km southwest of the sampling site. You state that the correlations between wind speed and wind direction between the two sites are consistent enough to warrant using the complete Hawera data set, as shown in a direction comparison for limited dates during August-October 2012 (Fig. S2). But is the limited period in 2012 adequate for evaluating whether Hawera data are appropriate for modelling wind transport at the Kapuni site? Moreover, Figures 2 and S1 show that the wind speed at Hawera (6-7 m/s) averages on the order of twice that at Kapuni (2-3 m/s). Have you done any sensitivity calculations to see how this difference in wind speed affects the modeling? The wind directions seem to be fairly consistent at the two sites.

See response to first comment from referee #1.

p.1, line 25: change "lowers" to "is reduced"

Done.

p.2, line 9: rearrange "reduction targets are commonly agreed as" to "commonly agreed upon reduction targets are"

Done.

p. 3, lines 16-17: You mention here and again later "the [photosynthesis] process faithfully recording the 14C content in new plant material", but you only reference the work showing this significantly after the mention on p. 11. It might help the reader to have this discussion earlier, since it is critical to the method.

We believe the referee is referring to the references and full description on p. 6 (rather than p. 11). The text in the introduction has been rearranged, but we have chosen to leave the detailed discussion and references for section 2.3 (p. 3 lines 15-23):

Plant material can be used as a proxy for atmospheric CO_2ff because plants assimilate carbon from the atmosphere during photosynthesis, in the process faithfully recording the ^{14}C content in new plant material. ~~The radiocarbon content in tree rings has been well established as a tracer for fossil CO2 emissions (Suess, 1955; Tans et al., 1979; Djuricin et al., 2012; Rakowski et al., 2013) and as a method to detect leaks from CO2 geosequestration (Donders et al., 2013).~~ Tree rings represent an integrated average of daytime CO_2 atmospheric mole fractions and ^{14}C content over the tree's annual growth period, <u>and can be independently dated using dendrochronology methods</u>. This allows for a retroactive analysis of CO_2ff mole fractions over many years, including any trends in emissions that occurred during the life of the tree. <u>The radiocarbon content in tree rings has been well established as a tracer for fossil CO_2 emissions (Suess, 1955; Tans et al., 1979; Djuricin et al., 2012; Rakowski et al., 2013) and as a method to detect leaks from CO_2 geosequestration (Donders et al., 2013).</u>

p. 5, line 18: "2008" should probably be "2007".

Thank you for noticing this error.

Figures: In general, increase font sizes for labels. Label panels within figures "a", "b", "c" to make it easier to refer to them in the text.

Done.

Figure 1: Can you add a large-scale location map locating Taranaki in New Zealand, as well as Hawera and Mount Taranaki? Add a label for Kapuni stream.

Done.

Figure 2: Font sizes. Label the legend (m/s).

Done.

Figure 4: The bottom axis of the top panel is missing. Increase the font size of the axis tick labels in all panels. The dates don't line up between the top two panels and the bottom panel. Increase all font sizes for the bottom panel. You use a subscript for CO2 in the bottom panel, but not in the top two. In the caption: "Dotted and dashed lines show modeled and observed six-year means, respectively."

Done.

Figure 5: What do the different colors for the circles indicate? The legend only shows the purple color.

The colours indicated the model sensor and were redundant (because the x-axis indicates the sensor as well). We have changed the colour of all of the circles to a single colour to avoid confusion.