Replies to Referee #1, Jocelyn Turnbull

We wish to thank Jocelyn Turnbull for her comments and suggestions for changes; we have revised the manuscript as follows (our answers are given in blue in the text below)

This paper describes a new modelling study that evaluates the influence of local (<100 km distant) nuclear power plant 14C emissions on 14CO2 measurements at Heidelberg, Germany. They transport detailed reported emissions from the nearby power plants using the HySPLIT model at several different meteorology resolutions. They identify which power plants contribute significantly to 14CO2 at Heidelberg, and how that varies through time. The results show that higher resolution meteorological fields are helpful in evaluating the influence of point source emissions such as these. More importantly, they show that when looking at individual sites with nearby nuclear 14CThis paper has a well-defined topic that is clearly explained, it is well-written, and the results are clear. It is a nice contribution to the literature and will be particularly relevant to the atmospheric 14C community. I have only a few extremely minor comments to clarify particular points, and recommend that this paper be accepted with these very minor changes.

Specific comments: Pg 2 line 3 and Pg 3 line 1-2. You say “in order to quantify the 14CO2 signal”, but I think you mean to say “in order to quantify the fossil fuel CO2 signal”. The 14CO2 signal naturally includes all sources including nuclear contributions, it is the fossil fuel CO2 calculation that needs to be adjusted to account for nuclear emissions.

This is absolutely correct, we have changed the wording correspondingly.

Pg 2 line 14. Naegler and Levin 2009 and Graven 2016 are not in the reference list. Please check referencing throughout. Also, please use hanging indents or numbering for the reference list to make it easier to scan through.

Thank you for pointing this out. We have added the references. Concerning formatting, it is not our choice but the Copernicus word template, which asks for this formatting, which I also find very unpractical …

Pg 2 line 22. I am not sure that “contaminate” is the right word, “influence” would be better.

From our point of view it is a “contamination”, and we would like to keep this expression, as “influence” is very unspecific.

Pg 2 line 22-24. There are a number of studies that have looked at 14C emissions from nuclear power plants, please reference some from research groups other than your own. For example:

Povinec, P.P., ChudÃ¡, M., Šivo, A., Šimon, J., HolÃ¡, K., Richtáriková, M. Forty years of atmospheric radiocarbon monitoring around Bohunice nuclear power plant, Slovakia (2009) Journal of Environmental Radioactivity, 100 (2), pp. 125-130.

Dias, C.M., Santos, R.V., Stenström, K., Nícoli, I.G., Skog, G., da Silveira Corrêa, R. 14C content in vegetation in the vicinities of Brazilian nuclear power reactors (2008) Journal of Environmental Radioactivity, 99 (7), pp. 1095-1101.

Koarashi, J., Akiyama, K., Asano, T., Kobayashi, H. Chemical composition of 14C in airborne release from the Tokai reprocessing plant, Japan (2005) Radiation Protection Dosimetry, 114 (4), pp. 551-555.

Stenström, K., Erlandsson, B., Hellborg, R., Wiebert, A., Skog, S., Vesanen, R., Alpsten, M., Bjurman, B. A one-year study of the total air-borne14C effluents from two Swedish light-water reactors, one

boiling water- and one pressurized water reactor (1995) Journal of Radioanalytical and Nuclear Chemistry Articles, 198 (1), pp. 203-213.

Uchrin, G., Hertelendi, E., Volent, G., Slavik, O., Morávek, J., Kobal, I., Vokal, B. 14C measurements at PWR-type nuclear power plants in three middle European countries(1998) Radiocarbon, 40 (1), pp. 439-446.

We added as references Uchrin et al., 1998 and Povinec et at., 2009.

Pg 4 line 5-8. Please include references to back the statement that BWR reactors mostly emit 14CO2 whereas others emit 14CH4.

We added the original reference from Kunz, 1985

Pg 8 line 21-28. Are there previous studies that examined the performance of HySPLIT with met data at different resolutions? What did they conclude?

There have been earlier studies using HYSPLIT with differently resolved meteorological data, such as the one cited (Su et al., 2015, Science of the Total Environment 506-507, 527-537) however, their findings were not directly applicable to our problem.

Pg 11 line 5 and throughout. Through most of the paper, the nuclear facilities are identified by their names – "Phillipsburg", etc. Here they are identified by the 3 letter codes, which are particularly confusing since KPP is not obviously the same place as Phillipsburg. Choose either the names or 3 letter codes and stick with them throughout the text.

We have removed the 3 letter codes in the text and use now only real names of the facilities

Pg 14 lines 1-10. I agree that the detailed emissions and LaGrangian model used in this paper give more detail (and more variability) than Graven and Gruber showed in their earlier paper. Yet a little more nuance in this paragraph would be helpful. In cases where nuclear facilities are nearby and have a strong influence, the detailed studies such as this one will be necessary. But for continental-scale studies looking at monthly or annual resolution, the gridded datasets provided by Graven and Gruber will likely be sufficient – and in many cases, it may be difficult to get more detailed information, so the Graven and Gruber dataset may still be the best choice.

We do not fully agree to the reviewer: We rather think that a coarse-resolution Eulerian model, similar to that used by Graven and Gruber, is not able to provide reliable results, neither in the near (10s of km) nor in the far field (few 100s of km), simply because - with a spatial resolution of 1.8° x 1.8° - it is principally not suited to simulate properly dispersion from a point source. It may be valuable to estimate the (very diluted) signal at the scale of 1000 km or so. Therefore, we think that for a reliable correction for nearby NPP contamination either a simple ("high-resolution") Gaussian plume approach (up to 10 km) or a high-resolution Lagrangian model is needed, preferably with higher resolution wind fields than used in the current study.

Replies to anonymous Referee #2

We wish to thank Referee #2 for her/his comments and suggestions for changes; we have revised the manuscript as follows (our answers are given in blue in the text below)

Kuderer et al. present an analysis of nuclear power plant influences on radiocarbon measurements in CO2 at Heidelberg using emissions data and the Hysplit model at three resolutions. Their main conclusions are that the nuclear correction decreased after the shutdown of Philippsburg BWR, the corrections they estimate are sensitive to model resolution, and nuclear corrections require careful consideration. The authors' work is useful and important to the community. However, some revisions are needed to clarify the details of their study and to expand the conclusions drawn from their results. The methods for model simulations are not very clear and there appear to be several different simulations used that are rather hard to follow.

A table describing the different simulations run for each nuclear site would be helpful.

We added a table (Tab. 2) with the respective information

Details about how the Hysplit runs were conducted, such as the number of particles and release times should be added.

The authors should also clarify that Hysplit was run in forward mode from the locations of the nuclear sites rather in backward mode from the observation site in Heidelberg. There appears to be some details described in the results section 3.2 that would fit better in the methods section.

The requested information is now added in the methods section 2.4 with some technical information moved here from the results section 3.2

The authors report in the abstract that "The mean correction for the period from 1986- 2014, if based on the 0.5◦ x 0.5◦ wind field, which we assume as the most accurate, is 2.3 ‰''. However, it appears high resolution 0.5◦ winds were only used in simulations for 2009 and 2011 – 2014, so it is not correct to say the 1986-2014 correction is from the 0.5◦ x 0.5◦ wind field. The other years were estimated from the coarser 2.5◦ resolution simulations with a correction factor based on comparisons for the years where 2.5◦ and 0.5◦ simulations were run for two reactors.

This is correct, and we changed the abstract as follows:

The finally applied mean $\Delta^{14}CO_2$ correction for the period from 1986-2014 is 2.3 ‰ with a standard deviation of 2.1 ‰ and maximum values up to 15.2 ‰. These results are based on the 0.5° x 0.5° wind field simulations in years when these fields were available (2009, 2011-2014) and, for the other years, they are based on 2.5° x 2.5° wind field simulations, corrected with a factor of 0.43.

Since Fig 4 shows the difference between simulated corrections at different resolution for individual samples is sometimes very small and sometimes very large (even with fixed emissions), is it valid to apply a mean correction to the data before 2009? Particularly if a main argument the authors are making is that the correction is highly variable in time? The authors argue that, since the correction is highly variable in time, monthly emissions data must be used and average emissions cannot be used, but then seem to contradict themselves by saying an average correction can be applied to account for model resolution, when actually this can be highly variable as well.

The referee is absolutely correct, however, we simply do not see an alternative possibility to correct for that obvious bias when using the coarse resolution wind field. We added in section 3.2 the expression … factor of 0.43, "as an attempt" to account for …

Another point is that 0.5◦ is still rather coarse compared to some regional modelling currently being done at 0.1◦ or finer resolution.

Yes, correct, however, higher resolution wind fields than those used here have not been available to us. We would be happy to work together with modelers in the future to refine our corrections …

Why do the authors use fixed emissions in the simulations shown in Fig 4?

We wanted to investigate the sole influence of the different wind fields on the correction (displayed in the more intuitive ‰ units). Further, including variable emissions would have led to a bias for high emission months.

If the authors have simulations with both fixed and monthly-varying emissions, can they include a comparison of these two simulations to quantify variability due to varying emissions vs variability due to varying transport? This comparison would be very useful.

All simulations are done with fixed emissions, which are later scaled. Sole emission variability is displayed in Fig. 2b, sole transport variability in Fig. 4a, and total scaled variability in Fig. 5b.

The authors note the previous estimate of the average nuclear correction by Levin using the plume model is higher than their estimate. Although the plume model is simpler, it might be considered to be at finer resolution than 0.5◦, and therefore a better estimate.

Levin et al. (2003) simply used one single dilution factor taken from the Turner (1970) workbook, and the mean wind statistics at the Philippsburg facility. The estimated factor, especially at the relatively large distance of 25 km from the emission point, may easily be wrong by a factor of two. (See also last comment to Referee #1.)

Can the authors make any inference on the detectability of the Philippsburg shutdown based on the Heidelberg Delta14CO2 data?

We have looked at that, however, as the average NPP contamination is of the same size as the individual measurement uncertainty, and as the variable fossil fuel signal is generally one order of magnitude larger, this is difficult or impossible.

The authors should discuss the impact the Philippsburg shutdown would have on the inferred fossil fuel CO2 at Heidelberg, if the change in the nuclear correction after 2011 was not accounted for. How does the change in the nuclear correction compare to the average fossil fuel signal in Delta14CO2 at Heidelberg?

(2.3 – 0.4 ‰)/(1.8‰/ppm) ≈ 1 ppm, this corresponds to ca. 10% of the total fossil fuel signal.

We added a respective remark at the end of the conclusions.

Section 3.3 Uncertainty in estimated nuclear correction – this needs more detail and seems rather too qualitative. The authors do not seem to include model transport uncertainty also for the high-resolution case.

Yes, but we are, unfortunately, not able to add more quantitative uncertainty estimates here, particularly not of model transport errors.

Do the authors have an estimate for the magnitude of sub-monthly variation in emissions?

No, the measurements in the exhaust air of Philippsburg reactors are integrated monthly values, and we are not aware of high-resolution exhaust data of $^{14}CO_2$. During revisions or fuel element change, there may occur short-term activity maxima, smeared out in the monthly means.

Do the authors think that monthly resolution in emissions data is sufficient, in general? Would this depend on the sampling integration time?

Emission data should ideally be as highly resolved as temporal changes of the meteorology occur, this may be hours or days. However, we think that for ^{14}C emissions we will probably not get higher than monthly or, at best, weekly resolved data, because ^{14}C measurements are (currently) not made continuously in situ, but rather on grab samples. We do not think that the required resolution would depend on the sample integration time, because the contamination at the measurement site varies most with meteorological conditions.

Could Fig 2b show the time series of emissions rather than yearly boxplots? It would be interesting to know if there is any pattern to the emissions over the year – for example, are emissions typically higher in summer potentially related to more maintenance undertaken in summer?

There is no apparent seasonality in the monthly emission data and no clear relationship between emissions and maintenance intervals. We added a respective sentence in the revised manuscript.

The authors should consider the Cattenom reactor in France, to the west and upwind of Heidelberg.

The estimated $^{14}CO_2$ emissions of the 4 PWR blocks of Cattenom (ca. 5.4 GWe) is about 0.2 TBq/year, i.e. half of the emissions from Philippsburg I. These reactors are located around 170 km west of Heidelberg. Therefore, we do expect at least one order of magnitude lower contamination from these facilities compared to those estimated for Philippsburg I.

Shouldn't Eq. 3 have a factor of 1000 for per mil units? What is used for XCO2 in this C3 calculation?

We have added the factor 1000. We used individually measured two-weekly mean CO_2 mole fractions.

First sentence in abstract – last word "fluxes" should be deleted. Radiocarbon measurements quantify fossil fuel derived CO2, but not fluxes.

We have changed "fluxes" into "component"

Also here the Delta notation is used without describing it. The phrase "14CO2 signal" is unclear – do you mean nuclear Delta14CO2 signal? Why are Delta14CO2 and 14CO2 both used in the abstract?

Isn't the Heidelberg site monitoring Delta14CO2 rather than 14CO2?

We think we can use in qualitative cases $\Delta^{14}CO_2$ and $^{14}CO_2$ more or less synonymously; however, we have made it clearer now that we give all numbers on the Δ-scale

Last sentence should be revised to "After operations at the Philippsburg boiling water reactor ceased in 2011, the" . . .

We changed this sentence accordingly

P2, L22: Delete "well".

Not sure why - the signals can really be large close to the facilities, i.e. a few km away !

L24 Comment about "normally quickly disperse" needs reference or should be deleted.

References were added.

P10, L21-24 – Please show some quantitative evidence from the simulations to support these statements.

Statements: “Therefore, and considering the high month-to-month variability of emissions (Fig. 2, lower panel), it is important to use monthly resolved emission data to estimate the Δ14Cnuclear signals originating from KKP I & II. The other four nuclear installations are secondary contributors permitting the use of annual average 14CO2 emission rates in absence of higher temporally resolved emission data.”

As the variability in model transport and NPP emissions are independent, we have to assume that both variabilities contribute to the total variability.

Figure 4b directly shows the low contamination from Neckarwestheim, which is only about 20% of that from Philippsburg (due to the ca. 25% lower emission rate and the larger distance of this NPP from Heidelberg).

The influence of $^{14}CO_2$ releases from regional nuclear facilities at the Heidelberg $^{14}CO_2$ sampling site (1986 - 2014)

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Abstract. Atmospheric $\Delta^{14}CO_2$ measurements are a well-established tool to estimate the regional fossil fuel-derived CO_2 component. However, emissions from nuclear facilities can significantly alter the regional $\Delta^{14}CO_2$ level. In order to accurately quantify the signal originating from fossil CO_2 emissions, a correction term for anthropogenic $^{14}CO_2$ sources has to be determined. In this study, the HYSPLIT atmospheric dispersion model has been applied to calculate this correction for the long-term $\Delta^{14}CO_2$ monitoring site in Heidelberg. Wind fields with a spatial resolution of 2.5° x 2.5°, 1° x 1° and 0.5° x 0.5° show systematic deviations, with coarser resolved wind fields leading to higher mean values for the correction. The finally applied mean $\Delta^{14}CO_2$ correction for the period from 1986-2014 is 2.3 ‰ with a standard deviation of 2.1 ‰ and maximum values up to 15.2 ‰. These results are based on the 0.5° x 0.5° wind field simulations in years when these fields were available (2009, 2011-2014) and, for the other years, they are based on 2.5° x 2.5° wind field simulations, corrected with a factor of 0.43. After operations at the Philippsburg boiling water reactor ceased in 2011, the monthly nuclear correction terms decreased to less than 2 ‰, with a mean value of (0.44 ± 0.32) ‰ from 2012 to 2014.

1 Introduction

Evaluation of the perturbation of atmospheric $^{14}CO_2$ by nuclear bomb tests in the middle of the last century has given very useful insight into carbon cycle dynamics (e.g. Levin and Hesshaimer, 2000). Today this artificial spike has almost equilibrated with the fast exchanging carbon reservoirs, and the currently observed global $\Delta^{14}CO_2$ trend ($\Delta^{14}CO_2$ being the relative deviation of the $^{14}C/C$ ratio in atmospheric carbon dioxide from standard material in permil (Stuiver and Polach, 1977)) is almost exclusively due to the ongoing input of ^{14}C-free fossil fuel CO_2 into the atmosphere (~~Naegler and Levin, 2009;~~ Levin et al., 2010; Graven, 2015). This long-term trend can potentially be used to estimate the global input of fossil fuel CO_2 into the atmosphere. However, the uncertainty of this estimate is still large (ca. 30%, Levin et al., 2010) due to the uncertainty of the large $^{14}CO_2$ disequilibrium fluxes from biosphere and ocean, as well as artificial ^{14}C sources. On the continental scale, however, atmospheric $\Delta^{14}CO_2$ measurements provide a powerful and the only direct and quantitative tool for estimating the regional fossil fuel component. $\Delta^{14}CO_2$ measurements at a polluted station allow separating fossil fuel-derived regional CO_2 enhancements relative to a clean reference level from those originating from biospheric fluxes if also the $\Delta^{14}CO_2$ level at the reference site is known (Levin et al., 2003; Turnbull et al., 2009). However, on that local to regional scale (several 10 km), $^{14}CO_2$ emissions from nuclear facilities, such as boiling water reactors, can significantly contaminate atmospheric $\Delta^{14}CO_2$. The ^{14}C signals from such point sources are well detectable in their immediate neighborhood in atmospheric CO_2 (and CH_4, e.g. Levin et al., 1992; Uchrin et al., 1998; Povinec et al., 2009) but also in plant samples (Levin et al., 1988). $^{14}CO_2$ "plumes" from point sources normally quickly disperse at distances of some tens of kilometers (Pasquill, 1961; Turner, 1970). But if a sampling station is located in the catchment of such $^{14}CO_2$ point sources, special care is required to accurately quantify the $\Delta^{14}CO_2$ contamination and correct for it to estimate reliable fossil fuel CO_2 values (e.g. Levin et al., 2003).

Here we present results from HYSPLIT dispersion modelling (Draxler and Hess, 1998) of $^{14}CO_2$ emissions from five nuclear installations in the < 60 km neighborhood of our long-term $\Delta^{14}CO_2$ monitoring site in Heidelberg. We apply the HYSPLIT model for the period of 1986-2014 with available wind fields of 2.5° x 2.5°, 1° x 1° and 0.5° x 0.5° resolution. Using reported $^{14}CO_2$ emission rates, these model estimates for the Heidelberg sampling site allow us to correct for the local $^{14}CO_2$ contaminations from nuclear facilities (Kuderer, 2016). Our model results, however, turned out to strongly depend on the resolution of the wind field used for the calculation. We discuss this important finding and present the currently most reliable corrections of our long-term $\Delta^{14}CO_2$ measurements.

2 Methods

2.1 Site description

The Heidelberg monitoring site is located on the University campus in the outskirts of Heidelberg, a medium size city in the upper Rhine valley in southwestern Germany (49° 25' N, 8°41'E, 116 m a.s.l., and see Figure 1). From 1986-2001, $^{14}CO_2$ samples for $\Delta^{14}C$ analysis have been collected from the roof of the former building of the Institute (INF 366) and from 2001 to present, from the new building about 500 m to the east (INF 229). At both locations, air was sampled from about 25 – 30 m a.g.l. The small difference in location of the two sampling sites is not relevant when estimating the nuclear $^{14}CO_2$ contamination with HYSPLIT.

Five nuclear installations with reported $^{14}CO_2$ emissions are found at distances between 25 km and 55 km to the Heidelberg station. Figure 1 shows their locations; details of reactor type, installed electrical output, period of operation, distance from the Heidelberg station and mean reported $^{14}CO_2$ emission during their operation up to 2014 are listed in Tab. 1. As the prevailing winds in the Upper Rhine valley are from south-west, Philippsburg (KKP I & II) is the most important source of potential $^{14}CO_2$ contamination in Heidelberg. Philippsburg I is the only boiling water reactor (BWR) with its major ^{14}C emissions being $^{14}CO_2$, whereas pressurized water reactors (PWR) emit ^{14}C mainly as $^{14}CH_4$ (Kunz, 1985). All the other nuclear installations except for Neckarwestheim II (GKN II) emit less than 15 % of Philippsburg I. Neckarwestheim is, however, located to the southeast of Heidelberg in the Neckar valley at a distance of 55 km, so that its relative contribution to the total $^{14}CO_2$ contamination is only less than 10 % (see Table ~~2~~3).

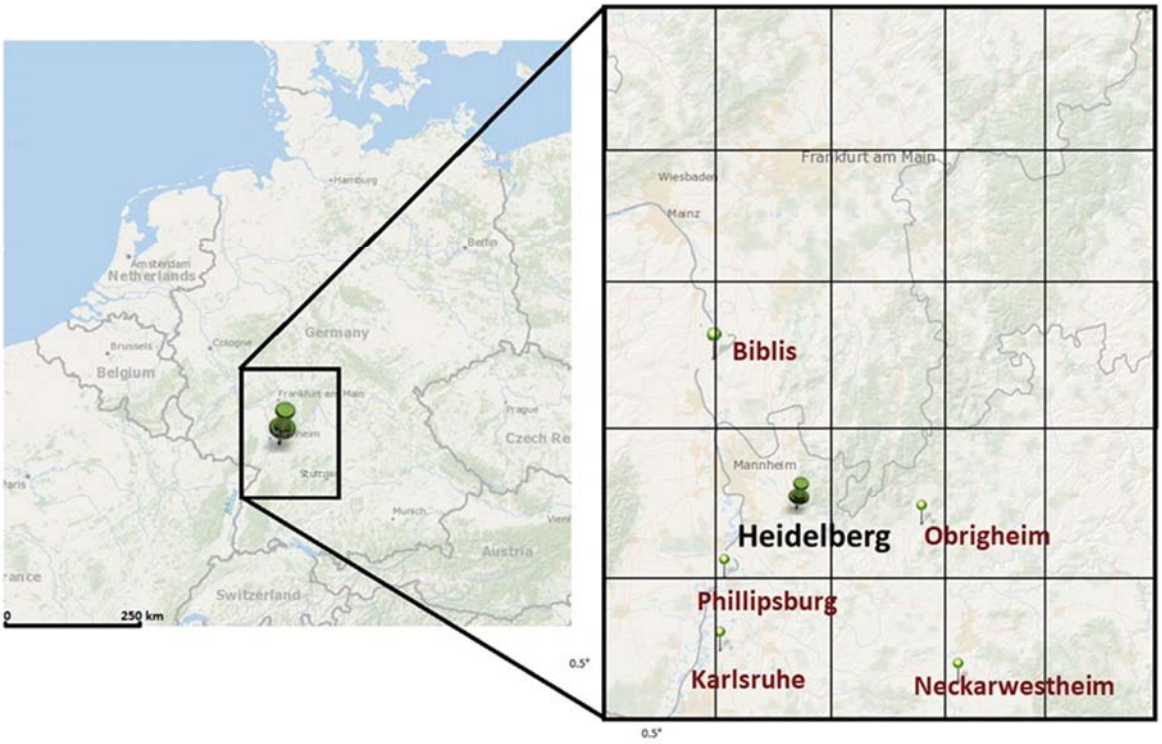

Figure 1: Map of the Heidelberg sampling site in southwest Germany. The locations of the five nearest nuclear facilities are shown in the enlargement. This enlargement corresponds to the size of the 2.5° x 2.5° wind field grid. The 0.5° x 0.5° wind field resolution is indicated by the grid in the enlargement.

2.2 $^{14}CO_2$ sampling and analysis

Two- and, for limited periods, also one-week integrated large volume samples of atmospheric CO_2 were collected from the roof of the Institute's buildings by quantitative chemical absorption in basic sodium hydroxide (NaOH) solution, as described by Levin et al. (1980). Except for the first few years, samples were collected only during night (from 19:00 to 7:00 Central European Winter Time), in order to avoid CO_2 contamination from local traffic. Moving the Institute to a new building in the year 2000 required parallel CO_2 sampling at both, the old and the new sampling locations on the Heidelberg University campus, in order to quantify possible differences and then allow combining the data sets from the two locations about 500 m apart. As the new building is located closer to the Heidelberg city center, slightly lower $\Delta^{14}C$ values (by on average 0.8 ‰) were found at the new location over the more than one-year overlapping period from late 2000 to early 2002. The results obtained from samples collected until 2002 at INF 366 at about 25 m a.g.l. were adjusted accordingly, and are now comparable with those

obtained at the current sampling location at INF 229 at about 30 m a.g.l. (for details of this comparison and correction, see Levin et al. (2008)).

$^{14}CO_2$ samples were processed in the Heidelberg ^{14}C laboratory by acidification of the NaOH solution in a vacuum system. The extracted CO_2 was subsequently purified over charcoal. The $^{14}C/C$ ratio was then measured by low level counting (Kromer and Münnich, 1992). All results are presented here as ^{13}C-corrected $\Delta^{14}C$ deviations from the international reference standard (Oxalic acid) in permil. They are corrected for decay to the date of CO_2 sampling (Stuiver and Polach, 1977). Note that Stuiver and Polach (1977) refer to this ^{14}C notation as Δ not $\Delta^{14}C$, however in order to be consistent with other atmospheric radiocarbon literature we stick to using $\Delta^{14}C$ instead of Δ. Precision of $\Delta^{14}C$ values was of order 4-5 ‰ in the 1980s and 1990s, of 3-4 ‰ in the 2000s and of 2-3 ‰ thereafter.

2.3 Reported $^{14}CO_2$ emissions from nuclear facilities in the surroundings of Heidelberg

According to the German Atomic Energy Act (Strahlenschutzverordnung, 2001), emissions of radioactive substances from nuclear facilities with the exhaust air must be monitored and reported quarterly to regional and federal authorities. The Bundesamt für Strahlenschutz (BfS, German Federal Office for Radiation Protection), releases yearly reports on radioactive emissions from all German reactors and research facilities; here the $^{14}CO_2$ emissions are reported separately from other radioactive substances. These BfS reports are available for the years 1986 – 2014 (BfS, 1986 – 2015). For Philippsburg I and II higher resolution, i.e. monthly emission data are available (Kernkraftwerk Philippsburg, pers. comm.); these monthly data were used in this work to estimate the $^{14}CO_2$ contamination in Heidelberg.

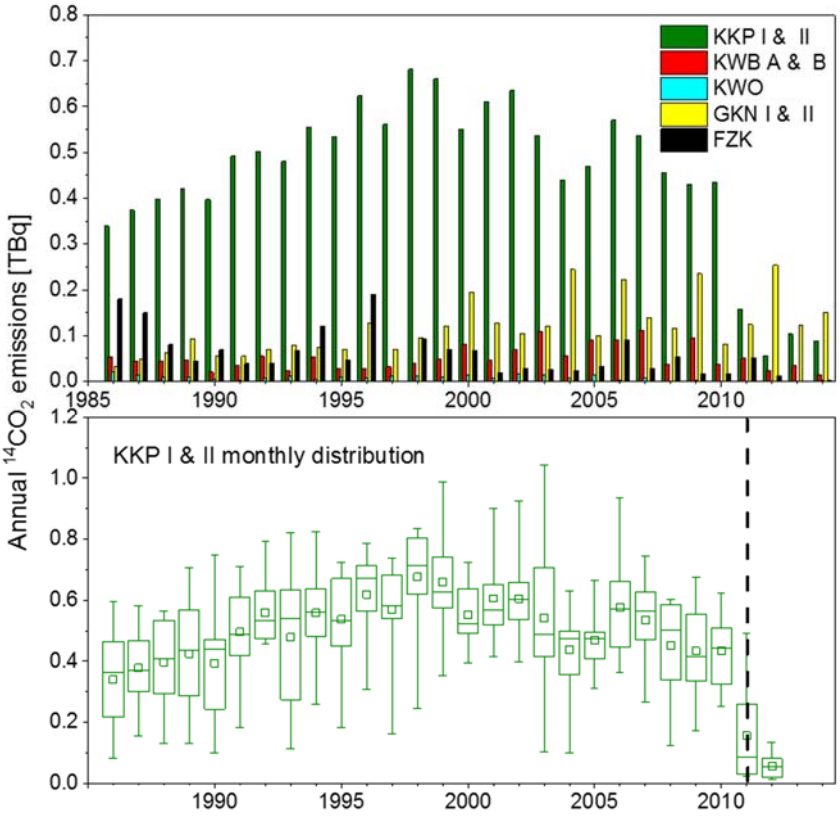

Figure 2: $^{14}CO_2$ emissions from nuclear facilities: Annual mean emissions from all facilities (upper panel) and box plots of the distribution of monthly values from Philippsburg (KKP I & II, (lower panel); the boxes include 50% of all months of the year with the horizontal bar indicating the mean and the square indicating the median value of the year. The whiskers show the minimum and maximum monthly values of the individual years. The dashed line indicates the shutdown of Philippsburg I shortly after the Fukushima accident.

Figure 2 (upper panel) shows annual $^{14}CO_2$ emissions from 1986 – 2014 for all five facilities listed in Tab. 1, while Fig. 2 (lower panel) shows the distribution of monthly emissions from Philippsburg I and II for the years 1986 - 2012. Note the huge variability of monthly emissions, which can differ from month to month by more than a factor of two. No seasonal variation nor any relation to particular maintenance activities was observed. Graven and Gruber (2011) estimated mean emission factors of 0.06 TBq $^{14}CO_2$ GWa^{-1} for PWRs and 0.51 TBq $^{14}CO_2$ GWa^{-1} for BWRs. From our emission data and corresponding power production reports, we do see, however, large differences from these emission factors and for PWRs no correlation at all, as displayed in Fig. 3. Moreover, keeping in mind the huge month-to-month variability of $^{14}CO_2$ emissions from Philippsburg

(Fig. 2, lower panel), underlines the necessity of reliable high-resolution $^{14}CO_2$ emission data from nuclear installations, if accurate corrections shall be applied to atmospheric $\Delta^{14}CO_2$ observations for fossil fuel CO_2 estimates.

Table 1: Nuclear facilities in the surroundings of Heidelberg. Reactor type (BWR: boiling water reactor, PWR: pressurized water reactor), installed electrical power and the average annual $^{14}CO_2$ emissions during their respective period of operation up to 2014 as well as the distance to the Heidelberg sampling site are given. Different reactor blocks are separated by slash. RR are research reactors and RP is the research reprocessing plant (WAK) of the Karlsruhe Research Center (FZK). After the operation period, further emissions occur during the decommissioning of the facilities (data taken from BfS (1986-2015)).

Nuclear facility	Installed electric capacity (MWe)	Type	Operation period	Mean $^{14}CO_2$ emission (TBq/yr)	Distance from Heidelberg
Philippsburg (KKP) I/II	926/1468	BWR /PWR	1980-2011/ 1984-2019	0.414/0.055	25 km
Obrigheim (KWO)	357	PWR/PWR	1969-2005	0.008	30 km
Biblis (KWB) A/B	1225/1300	PWR/PWR	1975-2011	0.025/0.037	37 km
Karlsruhe FZK/WAK	-	RR/RP	1971-1991	0.036	39 km
Neckarwestheim (GKN) I/II	840/1400	PWR/PWR	1976-2011/ 1989-2022	0.008/0.135	55 km

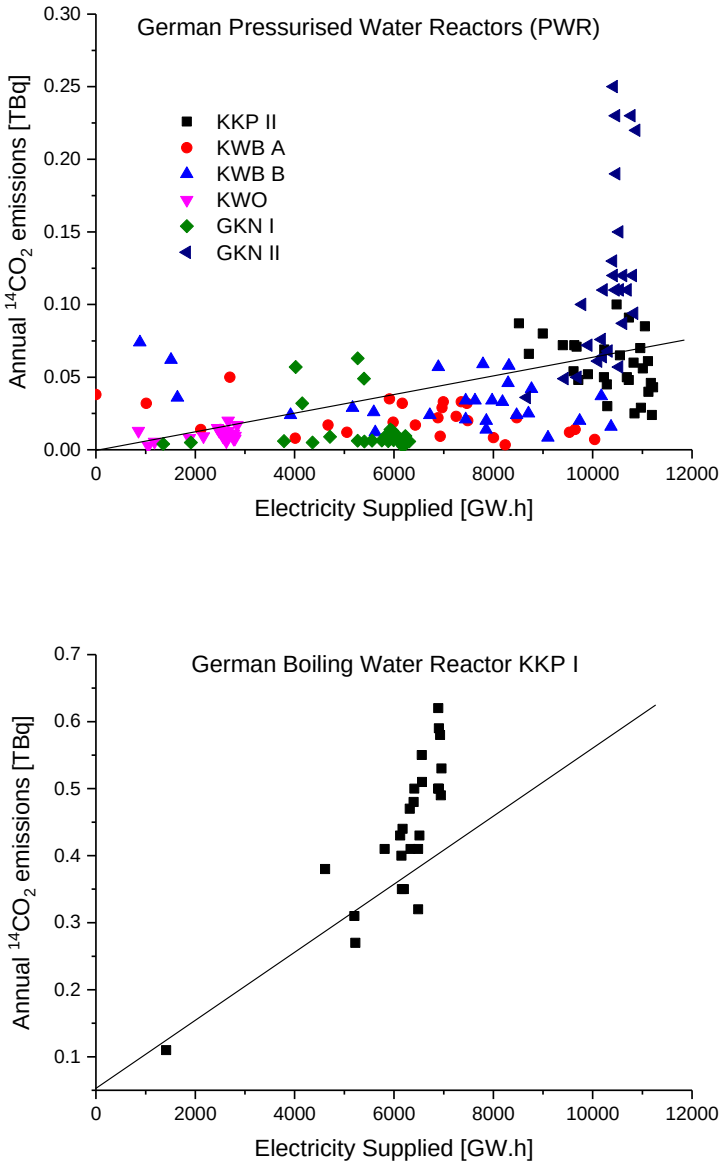

Figure 3: Relationship between annual $^{14}CO_2$ emissions from Pressurized Water Reactors (upper panel) and the Boiling Water Reactor Philippsburg I (lower panel) and their annual electricity supplied. The solid lines show the specific emission factors reported by Graven and Gruber (2011).

2.4 The HYSPLIT model

The Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) from NOAA offers a variety of services ranging from computing simple air parcel trajectories up to complex dispersion simulations (Draxler and Hess 1998). During the simulations, virtual particles are emitted at the source location and advected to the new particle position, described by the position vector **P**, using the input wind velocity vector field **V**:

$$\mathbf{P}(t + \Delta t)\_advection = \mathbf{P}(t) + 0.5 \cdot [\mathbf{V}(P, t) + \mathbf{V}(\mathbf{P'}(t+\Delta t), t+\Delta t)] \cdot \Delta t. \quad (1)$$

The advection equation is solved with a dynamic time step Δt, demanding that the advective displacement is smaller than the size of a grid cell (Draxler, 1999). Equation 1 is solved numerically by integrating the velocity vector over time, making use of the trapezoidal rule, i.e. averaging the velocity vectors at the initial position **V**(**P**, t) and first-guess position **V**(**P**'(t+Δt), t+Δt) = **V** {(**P**(t) + **V** (**P**, t) · Δt),(t + Δt)} of the particle. To account for atmospheric dispersion, the particles are displaced stochastically (Eq. 2a & b):

$$X\_final\ (t + \Delta t) = X(t + \Delta t)\_advection + U'\_dispersion\ (t + \Delta t) \cdot \Delta t \quad (2a)$$

$$Y\_final\ (t + \Delta t) = Y(t + \Delta t)\_advection + W'\_dispersion\ (t + \Delta t) \cdot \Delta t \quad (2b)$$

where the turbulent velocity components U', W' are estimated from the standard deviations σ of the horizontal or respective vertical velocity components (Fay et al., 1995). For more details, see Stein et al. (2015) and references therein.

The HYSPLIT model was run here in the forward mode with an internal spatial resolution of 0.05° x 0.05° and an internal time step fixed by the stability ratio 0.75, i.e. the timestep is chosen such that the maximal advective displacement is smaller than 0.75 times the grid size. For every nuclear facility location, a separate run has been conducted with a constant emission rate. Due to the small distance between ^{14}C sources and the measurement station Heidelberg, simulations were limited to 48 hours, where each run consisted of a 24-hour period, with 2500 particles being emitted every hour, followed by 24 hours of sole propagation of the particles. Thus, for each day the simulated nuclear ^{14}C activity included the actual emissions of this day arriving at the sampling site and the propagated emissions from the day before. This could potentially lead to loss of particles, which arrive at the measurement site more than 24-48 hours after the release, but for an extended reference period only a minor effect has been observed. Note that typical travel times from the nuclear power plants to Heidelberg are of order 6-12 hours. The HYSPLIT model computes for every hour the particle concentration in every grid box, which gives a dilution factor f (see Eq. 3), describing how much the point source emissions are diluted over the respective grid. This dilution factor is strongly depending on the prevailing meteorological conditions. All relevant control parameters of the different runs are listed in Table 2.

2.5 Wind fields

Previous studies have shown that HYSPLIT calculations are sensitive to the meteorological input data (e.g., Cabello et al., 2008; Lin et al., 2015). Here we used three different wind velocity fields that have a horizontal resolution of 2.5° x 2.5°, 1° x 1° and 0.5° x 0.5°. The GDAS (Global Data Assimilation System) assimilates meteorological observations in numerical weather prediction models and archives the results. The one degree fields GDAS1 are available since 2005 and the half degree fields GDAS0p5 since 2008. GDAS1 and GDAS0p5 differ besides the horizontal also in the vertical resolution (Lin et al., 2015). The NCEP/NCAR (National Centre for Environmental Prediction/National Centre for Atmospheric Research) reanalysis provides atmospheric analyses with a spatial resolution of 2.5° x 2.5°, using historical data from 1948 onwards. All three wind fields are readily available at ftp://arlftp.arlhq.noaa.gov/pub/archives/.

2.6 Estimation of $\Delta^{14}C_{nuclear}$

The ^{14}C signal at the sampling site $\Delta^{14}C_{nuclear}$ originating from $^{14}CO_2$ emissions from each nuclear facility is calculated by scaling the meteorological dilution factor f (s m^{-3}) at the measurement station obtained from the HYSPLIT simulation with the time-varying emission strength Q (Bq s^{-1}) of the source. This specific ^{14}C activity is converted (according to its definition from Stuiver and Polach (1977)) into $\Delta^{14}C_{nuclear}$ in ‰ according to Eq. 3

$$\Delta^{14}C_{nuclear} = f \cdot Q \cdot X_{CO2} / (M_C \cdot V_m \cdot a) \ (\cdot 1000\ ‰), \quad (3)$$

with the molar volume at standard atmospheric temperature and pressure (STP) V_m = 24.465 mole m^{-3}, molar mass of carbon M_C = 12 g $mole^{-1}$, mole fraction of CO_2, X_{CO2}, and specific activity of the ^{14}C standard a = 0.238 Bq gC^{-1}.

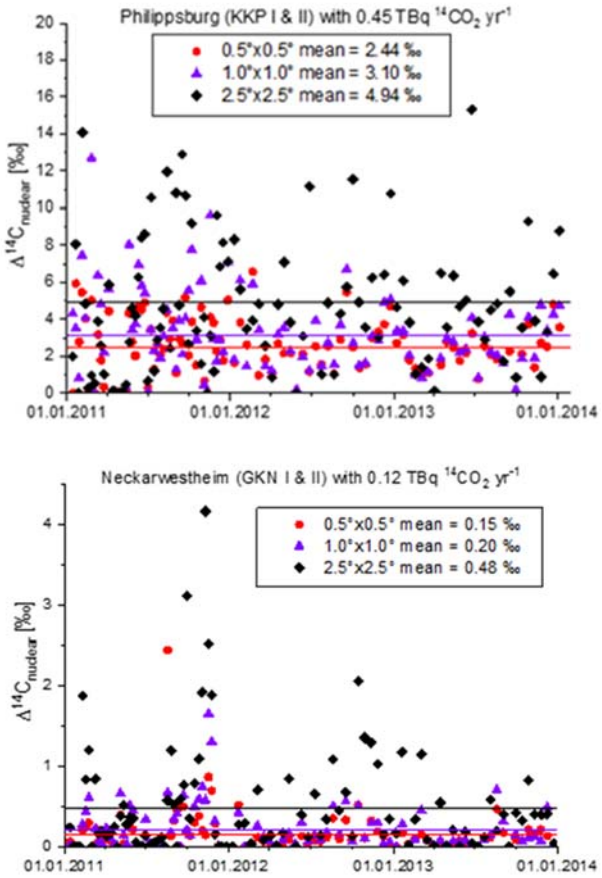

Figure 4: Upper panel: Calculated $\Delta^{14}C_{nuclear}$ contributions from Philippsburg with assumed constant $^{14}CO_2$ emissions using the three wind fields with different resolution. Lower panel: Same as upper panel, showing the contributions from Neckarwestheim.

3 Results

3.1 $\Delta^{14}C_{nuclear}$ estimates using wind fields of different resolution

Figure 4 (upper panel) shows two-weekly (i.e. sampling period) integrated HYSPLIT-estimated $\Delta^{14}C_{nuclear}$ contributions in Heidelberg for 2011 – 2013, originating from assumed constant $^{14}CO_2$ emissions from Philippsburg of 0.45 TBq yr^{-1} (corresponding to the long-term average emission from this facility). The different symbols distinguish the results when using the three different wind fields, i.e. with resolution of 2.5° x 2.5° (black diamonds), of 1° x 1° (blue triangles) and of the highest resolution of 0.5° x 0.5° (red circles). The two-week integrated $\Delta^{14}C_{nuclear}$ signals vary between 0‰ and 16 ‰ for the coarse

resolution wind field, and show on average lower signals when using the higher resolved wind fields. There are, however, also situations when we obtain lower contamination signals with the coarse resolution wind field than with the higher resolved fields. The 1° x 1° wind field also yields, on average, slightly higher $\Delta^{14}C_{nuclear}$ signals from Philippsburg than the highest resolution 0.5° x 0.5° wind field, but the differences between those two are often only marginal. Looking at the contributions from the Neckarwestheim reactors (GKN I & II) (Figure 4 lower panel), we also estimate the largest $\Delta^{14}C_{nuclear}$ signals with the low-resolution wind field, while the highest resolution wind field yields the smallest signals. The mean ratio between the contamination signals estimated with the highest resolution wind field and those estimated with the 2.5° x 2.5° resolution field is 0.43. We consider the results from the higher-resolution wind fields more reliable to calculate $\Delta^{14}C_{nuclear}$ than those with the coarse resolution field (see discussion below). We can further see that the contributions from Neckarwestheim $^{14}CO_2$ emissions on the Heidelberg $\Delta^{14}CO_2$ signal are, on average, about one order of magnitude smaller than those from Philippsburg and, thus, with an average $\Delta^{14}C_{nuclear}$ of less than 0.2 ‰, almost negligible.

3.2 Estimation of $\Delta^{14}C_{nuclear}$ in Heidelberg from all five nuclear installations

Owing to its source strength and proximity to Heidelberg, Philippsburg I is the dominant contributor to the nuclear contamination at our sampling site. Therefore, and considering the high month-to-month variability of emissions (Fig. 2, lower panel), it is important to use monthly resolved emission data to estimate the $\Delta^{14}C_{nuclear}$ signals originating from this facility. The other four nuclear installations are secondary contributors permitting the use of annual average $^{14}CO_2$ emission rates in absence of higher temporally resolved emission data. For each source location, the HYSPLIT model was run for every calendar day separately covering the period 1986 - 2014.

Table 2: Control parameters of the HYSPLIT runs and used wind field data for $^{14}CO_2$ contamination estimates for the different nuclear facilities

Internal spatial resolution	0.05° x 0.05°
Internal temporal resolution	fixed internally by stability criterion (0.75)
Direction of the run	forward
Number of source locations per run	1
Number of runs with different source locations	5
Emission rate (per hour)	1
Hours of emission	24
Total run time (hours)	48
Particles released per cycle	2 500
Maximum number of particles	50 000

Wind field resolution:	
Philippsburg I & II	1986-2008, 2010: 2.5° x 2.5°*; 2009, 2011-2014: 0.5° x 0.5°
Obrigheim	1986-2014: 2.5° x 2.5°*
Biblis A & B	1986-2014: 2.5° x 2.5°*
Neckarwestheim I & II	1986-2014: 2.5° x 2.5°*
Karlsruhe	1986-2014: 2.5° x 2.5°*

*The HYSPLIT results obtained with 2.5° x 2.5° wind fields have been corrected with a factor of 0.43

Table 3: Relative average $\Delta^{14}C_{nuclear}$ contribution in Heidelberg from 1986 to spring 2011 (shutdown of Philippsburg I)

	Obrigheim	Biblis A & B	Neckarwestheim I & II	Philippsburg I & II	Karlsruhe
%	1.05	1.39	6.80	88.13	2.63

For the Philippsburg reactor site, the following meteorological data has been used (Tab. 2): For 1986 – 2008 and 2010, we used the 2.5° x 2.5° fields, for 2009 and 2011 – 2014 the 0.5° x 0.5° fields. For the other four source locations (Obrigheim, Biblis A & B, Neckarwestheim 1 & 2 and Karlsruhe), the 2.5° x 2.5° wind field data have been used for the entire period 1986 – 2014, in order to save computing time. All coarse grid dilution factors were then corrected with a factor of 0.43 as an attempt to account for the effect of under-estimating atmospheric dispersion in coarse grid simulations. This factor was obtained from the comparison made for the 3-year period 2011-2013 at Philippsburg and Neckarwestheim (Fig. 4). The average relative contributions to the total $\Delta^{14}C_{nuclear}$ signal for all facilities are listed in Tab. ~~2~~3. The largest correction terms for a two-week sampling period originating from Philippsburg I & II were 15.2 ‰, from Neckarwestheim I & II, it was 3.3 ‰ and from Biblis A & B it was 1.1 ‰. From the other two facilities, they were always smaller than 1 ‰.

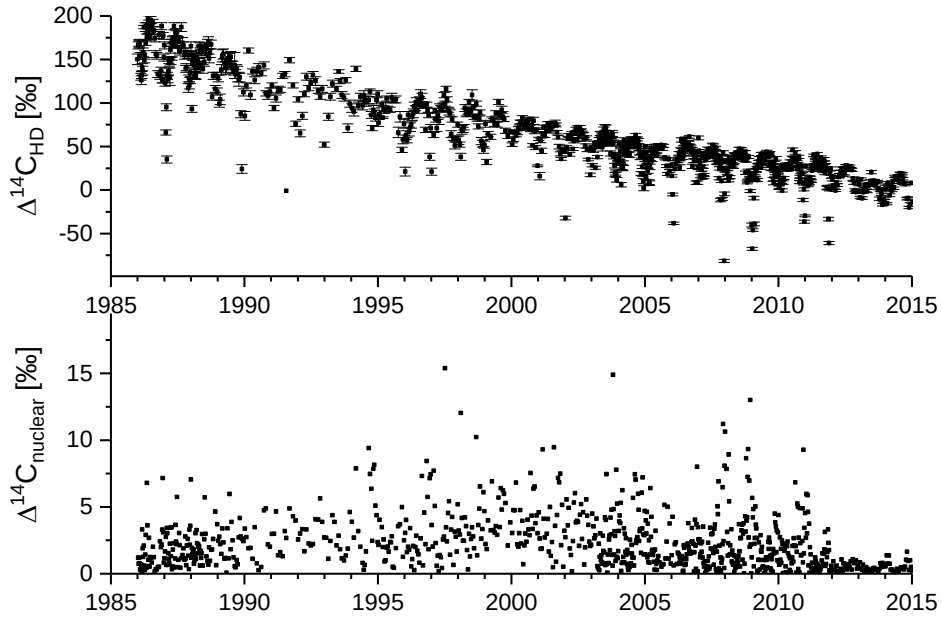

Figure 5: Upper panel: Results of $\Delta^{14}CO_2$ measurements in Heidelberg (uncorrected); lower panel: nuclear contribution from all installations in Heidelberg (note expanded $\Delta^{14}C$ scale)

The individual uncorrected Heidelberg $\Delta^{14}CO_2$ data are displayed in Fig. 5 (upper panel) together with the individual total $\Delta^{14}C_{nuclear}$ corrections (lower panel). In the years before the Philippsburg I shutdown, about 1 % of all corrections were above 10 ‰ and less than 2 % above 5 ‰. The mean correction was 2.3 ‰ with a standard deviation of 2.1 ‰. After the shutdown of the BWR Philippsburg I, the largest $^{14}CO_2$ source before 2011, $\Delta^{14}C_{nuclear}$ decreased to less than 2 ‰, with a mean value of (0.44 ± 0.32) ‰ from 2012 to 2014. It is therefore feasible to apply only an average correction of this size to the Heidelberg measurements of all subsequent years.

3.3 Uncertainty of estimated $\Delta^{14}C_{nuclear}$

The uncertainty of our $\Delta^{14}C_{nuclear}$ estimates originates from uncertainties in emission data and uncertainties in the HYSPLIT model transport. From comparison of results based on the differently resolved wind fields (Fig. 4), we find the largest deviations between the 2.5° x 2.5° and the 1° x 1° fields while the average differences between the two finer resolved wind fields are of order 30 %, they can, however, be as large as a factor of two for individual two-week periods. The uncertainty of the measured monthly emission data is probably less than 10-20 % and thus small if compared to the uncertainty of the model

transport (although sub-monthly variability in the emissions may also contribute to the uncertainty of the $\Delta^{14}C_{nuclear}$ estimates). For the contributions from nuclear installations where only annual average emission data were available to us, the uncertainty of emissions is estimated to 30 %. As the contribution from all four installations except Philippsburg contribute on average only 12 % (Tab. ~~2~~3) this uncertainty is small compared to the transport uncertainty of the contributions from Philippsburg. We, therefore, estimate the typical uncertainty of individual total $\Delta^{14}C_{nuclear}$ signals to less than 35 %. It is worth noting from Fig. 4 a & b that the variability of $\Delta^{14}C_{nuclear}$ is larger for the 2.5° x 2.5° wind field calculations than would be expected from the mean differences between the fine and the coarse resolution wind field simulations. Applying a simple correction factor of 0.43 on all values estimated for the years 1986-2008 and 2010 with the 2.5° x 2.5° wind field, therefore, adds variability and uncertainty to the $\Delta^{14}C_{nuclear}$ corrections, which is, however, not possible to quantify with the currently available information.

4 Discussion and Conclusions

Our HYSPLIT estimates of $^{14}CO_2$ contaminations from nuclear facilities in the catchment area of Heidelberg showed large differences when using wind fields of different resolution. The calculated mean contamination was approximately twice as large when using the coarse resolution 2.5° x 2.5° wind field compared to the two higher resolution fields. Previous studies have shown, that meteorological coarse grid re-analyses can be well suited to capture synoptic-scale dynamical processes, but biases in surface wind speeds may be introduced as re-analysis data are not well adapted to reproduce transient strong wind events occurring at the mesoscale and generating a large sub-grid scale variability (Largeron et al., 2015). These can arise in HYSPLIT trajectory calculations, which are the basis for concentration simulations, when the air mass passes through areas with complicated topography and meteorological patterns that are on a smaller scale than the data resolution (Su et al., 2015). Another and possibly more important factor is that atmospheric dispersion is included in the model by using the standard deviation of the interpolated velocity field. Linearly interpolating the coarse wind field to the internal HYSPLIT grid (here 0.05° x 0.05°) leads to a less variable velocity field compared to initially starting with a fine grid. This generates more distinct plume shapes in coarse grid simulations (Kuderer, 2016). Therefore, using the coarse wind field may underestimate the effect of atmospheric dispersion, leading to high values when the plume directly passes the measurement point. We expect this to occur frequently in the case of the Philippsburg $^{14}CO_2$ plume, where the source lies in the main wind direction at rather short distance from the measurement point. This effect may explain the occasionally high $\Delta^{14}C_{nuclear}$ values estimated for a number of sampling periods before 2009 (Fig. 5b), which are not seen in the measured uncorrected data (Fig. 5a). In the case of Neckarwestheim, this explanation does not hold. However, also here we consider the results obtained with the finest resolution wind field as more accurate. Neckarwestheim lies in the hilly Neckar valley with a complex topography, which is probably better represented by the finer resolution wind fields. Overall, we expect the HYSPLIT estimates that are based on higher resolution wind fields to provide more realistic results, in particular as the topography around Heidelberg is not flat. We therefore correct the HYSPLIT results obtained with the 2.5° x 2.5° wind fields for the earlier years when high-resolution wind

fields (0.5° x 0.5°) are not available. Note, however, that this first rough correction comes with additional uncertainty and variability (see above).

In an earlier study by Levin et al. (2003), Philippsburg I & II were considered as the sole sources for the nuclear contamination at the Heidelberg sampling site. A Gaussian plume model (Turner, 1970) with a constant mean dispersion factor had been applied there to calculate $\Delta^{14}C_{nuclear}$ as a first approximation, but using the same monthly $^{14}CO_2$ emissions as in the present study. The mean nuclear signal estimated by Levin et al. (2003) was $\Delta^{14}C_{nuclear}$ = (4.8 ± 2.0) ‰ ranging from 0.2 ‰ to 10 ‰ for monthly mean values. This earlier estimate of $^{14}CO_2$ contamination is approximately twice the value obtained with the HYSPLIT model and the high-resolution wind fields. Graven and Gruber (2011) used the TM3 model with a spatial resolution of 1.8° x 1.8° and estimated for 2005 a total $\Delta^{14}C_{nuclear}$ of 2.1 (1.1 - 3.7) ‰ for the Heidelberg grid cell. Their estimate is in agreement with our results for that year ((2.1 ± 1.6) ‰) obtained with the 2.5° x 2.5° resolution wind field corrected by the factor of 0.43. As in the present study, Graven and Gruber (2011) also included ^{14}C contributions from other nuclear installations in their estimates. However, their assumed emissions from the Philippsburg I reactor were estimated with the average emission factor for BWR, which is about 20 % smaller than the measured value for 2005 used in our estimate. They also mention that their Eulerian model may have under-estimated the true contamination due to its coarse resolution, which would dilute point source emissions over a large grid in an Eulerian approach.

These comparisons with earlier studies indicate that more work and higher resolution models and wind fields are needed to reduce the uncertainty of the $^{14}CO_2$ contamination estimates from nuclear installations at measurement sites where $\Delta^{14}CO_2$ observations shall be used to precisely determine the regional fossil fuel CO_2 component. Currently, we have to take into account a model transport uncertainty of about 1-2 ‰ in the estimated $\Delta^{14}C_{nuclear}$ contamination, if the measurement site is located closer than about 30 km downwind from a nuclear facility, which has a $^{14}CO_2$ emission rate of about 0.5 TBq yr^{-1} similar to the Philippsburg I boiling water reactor with 1 MWe power production. Other reactor types, such as the Canadian CANDU reactors may have significantly larger emission rates (Graven and Gruber, 2011; Vogel et al., 2013); the uncertainty of corresponding $\Delta^{14}C_{nuclear}$ estimates in their close neighbourhood may then be considerably larger.

The limited temporal resolution of $^{14}CO_2$ emission rates from nuclear installations cause additional uncertainty on the $\Delta^{14}C_{nuclear}$ estimates, as generally only annual mean emissions are reported. Graven and Gruber (2011) assume that $^{14}CO_2$ emissions are proportional to the annual power production. However, the present study on the influence from German reactors on the Heidelberg measurement site does not fully support this finding. Figure 3 does not show significant correlations between annual $^{14}CO_2$ emissions and corresponding electricity supply. Therefore, assuming emission factors as suggested by Graven and Gruber (2011) will add considerable uncertainty to the $\Delta^{14}C_{nuclear}$ estimates, which may be as large as the uncertainties estimated here for the wind field-based model transport error.

Overall, we conclude that careful investigation of potential $^{14}CO_2$ emissions in the catchment of sampling sites is required when using $\Delta^{14}CO_2$ observations for fossil fuel CO_2 estimates. The differences of our HYSPLIT modelling results, when based on differently resolved wind fields, together with the findings from earlier studies suggest that current $\Delta^{14}C_{nuclear}$ estimates may be wrong by a factor of two. Therefore, careful investigations with high-resolution models must be performed at all stations where ^{14}C-based fossil fuel CO_2 measurements are conducted. Based on our simulations, the shutdown of Philippsburg I in 2011, if not accounted for in the $\Delta^{14}C_{nuclear}$ correction, would have masked a fossil fuel CO_2 signal of 1 ppm, corresponding to 10% of the average total fossil fuel CO_2 signal in Heidelberg. We, therefore, plan similar studies for the European ICOS atmospheric station network (https://www.icos-ri.eu/icos-stations-network). The basis must be high-resolution $^{14}CO_2$ emissions data from nuclear facilities, which need to be made available for these investigations, if contamination estimates shall be accurate.

Data availability

Data will be available from the Heidelberg University data depository under
https://heidata.uni-heidelberg.de/dataverse/carbon

Author contributions

I.L. and S.H. have designed the study. M.K. made the HYSPLIT model calculations and evaluated the data. I.L. prepared the manuscript with support from M.K. and S.H.

Acknowledgements

The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for providing the HYSPLIT transport and dispersion model used in this publication. We especially like to thank Bernd Kromer and the staff of the Heidelberg ^{14}C laboratory for their careful work analysing the $^{14}CO_2$ samples, and Ute Karstens for helpful discussions on the manuscript. Financial support came from a number of agencies in Germany and Europe. These are the Heidelberg Academy of Sciences, the Ministry of Education and Science, Baden-Württemberg, Germany, the German Science Foundation, the German Ministries for the Environment, of Education and Science, and of Transportation and digital Infrastructure, the German Umweltbundesamt and the European Commission, Brussels.

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