



**Methane simulations
for Cape Grim,
Tasmania**

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Simulations of atmospheric methane for Cape Grim, Tasmania, to constrain South East Australian methane emissions

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Abstract

This study uses two climate models and six scenarios of prescribed methane emissions to compare modelled and observed atmospheric methane between 1994 and 2007, for Cape Grim, Australia (40.7° S, 144.7° E). The model simulations follow the TransCom-CH₄ protocol and use the Australian Community Climate and Earth System Simulator (ACCESS) and the CSIRO Conformal-Cubic Atmospheric Model (CCAM). Radon is also simulated and used to reduce the impact of transport differences between the models and observations. Comparisons are made for air samples that have traversed the Australian continent. All six emission scenarios give modelled concentrations that are broadly consistent with those observed. There are three notable mismatches, however. Firstly, scenarios that incorporate interannually varying biomass burning emissions produce anomalously high methane concentrations at Cape Grim at times of large fire events in southeastern Australia, most likely due to the fire methane emissions being unrealistically input into the lowest model level. Secondly, scenarios with wetland methane emissions in the austral winter overestimate methane concentrations at Cape Grim during wintertime while scenarios without winter wetland emissions perform better. Finally, all scenarios fail to represent a methane source in austral spring implied by the observations. It is possible that the timing of wetland emissions in the scenarios is incorrect with recent satellite measurements suggesting an austral spring (September-October-November), rather than winter, maximum for wetland emissions.

1 Introduction

Methane (CH₄) is an important greenhouse gas whose atmospheric concentration has more than doubled since the 18th century (MacFarling Meure et al., 2006), with considerable variations in its growth rate over recent decades (Rigby et al., 2008; Dlugokencky et al., 2009; Sussmann et al., 2012). Methane has both anthropogenic and natural emissions, while the main sink for methane is through reaction with hydroxyl

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radical (OH) in the troposphere and by photolysis in the stratosphere. On a global scale, considerable uncertainty remains about the causes of recent changes in the methane growth rate (Kirschke et al., 2013), and on a regional level, significant discrepancies have been found between “bottom up” inventory estimates of emissions and “top down” atmospheric inverse modelling studies. For example, in Miller et al. (2013), the authors find that the inventories underestimate emissions by up to 2.7 times in the south-central USA.

Atmospheric concentrations of methane are measured approximately weekly at more than 150 flask-sampling sites worldwide and continuously at a smaller number of in situ instrumented sites. Some of the longer in situ records are those of the Advanced Global Atmospheric Gases Experiment (AGAGE) network (Cunnold et al., 2002), including the record from Cape Grim, Australia. A number of global modelling studies have used such methane atmospheric records to constrain the methane budget for example, Fung et al. (1991), Houweling et al. (1999) and Bousquet et al. (2011). Rigby et al. (2008) used the AGAGE in situ methane and CSIRO flask methane data, together with the AGAGE methyl chloroform records to deduce the Northern and Southern Hemisphere changes in methane emissions required to account for the measured increase in methane mole fraction growth rate from 2007.

Recently a model intercomparison, “TransCom-CH₄”, has been run for methane (Patra et al., 2011), with a focus on understanding how transport model differences contributed to variations in the methane simulations. All model simulations used the same prescribed methane emissions and modelled methane loss using prescribed, climatological OH fields. We have used the simulations defined for this intercomparison to compare simulated and measured methane at Cape Grim, with the goal of evaluating the accuracy of the prescribed methane emissions for southeastern Australia. The model intercomparison also simulated radon, sulfur hexafluoride (SF₆) and methyl chloroform (CH₃CCl₃) as additional tests for different components of the transport model. Radon (²²²Rn) is emitted reasonably uniformly in both space and time from land surfaces at much higher rates than from oceans and decays radioactively with a half-life of

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3.8 days. Due to its short lifetime, radon is often used as a tracer of recent contact with land and to explore vertical mixing through the lower atmosphere (Zahorowski et al., 2004; Williams et al., 2011, 2013; Chambers et al., 2011). Since radon fluxes are usually assumed to be better characterized than fluxes of other trace gases, radon has been used, via tracer ratio methods, to estimate regional carbon dioxide, methane and nitrous oxide or other greenhouse gas fluxes (Schmidt et al., 1996; Wilson et al., 1997; Biraud et al., 2000; Zahorowski et al., 2004; Wada et al., 2013).

An Australian methane budget was described by Wang and Bentley (2002), who used 1997 Cape Grim atmospheric methane measurements in an inversion to constrain the methane fluxes from southern Australia. Their study suggested that the methane inventory overestimated southeastern Australian fluxes. A more recent modelling study by Fraser et al. (2011) ran forward model simulations for 2005–2008 and separated methane into different regional and process-based components, focusing on better parameterising methane fluxes from seasonal wetlands in tropical Australia. At Cape Grim, they found their simulated methane was dominated by animal, landfill and ocean fluxes. They also noted that their model reproduced background, baseline methane concentrations at Cape Grim well, but the model was less successful in reproducing concentrations influenced by local emissions. The concentration excursions driven by local emissions are typically large in magnitude and are referred to as “non-baseline” events. These events contain information regarding regional fluxes and are the focus of this study.

To investigate non-baseline events, this study uses both observations at Cape Grim and forward model simulations with prescribed emissions. Sections 2 and 3 describe the observations at Cape Grim and the model simulations, respectively. Section 4 focuses on seasonal scale results, exploring an apparent anomaly between modelled and observed methane in austral spring. The implications for regional methane fluxes are discussed in Sect. 5.

2 Observations

Cape Grim is located at the top of a 90 m cliff on the north-west coast of Tasmania (40.7° S, 144.7° E), which is separated from mainland Australia by Bass Strait (Fig. 1). Cape Grim has been operating since the 1970s and now has the most comprehensive monitoring program in the Southern Hemisphere for greenhouse gases (Langenfelds et al., 2014), ozone-depleting gases (Krummel et al., 2014), and radon (Zahorowski et al., 2014).

Figure 2 shows methane and radon observations made at Cape Grim during 2003. Observations made at Cape Grim are characterised by baseline periods when the wind is from the south-west, (typically 30 % of the time), and air parcels have had long trajectories over the Southern Ocean. During these periods, concentrations are reasonably steady displaying underlying seasonal variations and long-term trends. Baseline periods are punctuated by non-baseline events characterised by higher concentrations that vary rapidly in time, indicative of air parcel trajectories that have travelled over mainland Australia or Tasmania, and consequently have been influenced by regional and local emissions. The seasonal cycles in both methane and radon are apparent in Fig. 2a and b which shows a full year of data. (Unfortunately, there are no radon data available for the month of April in 2003.) In Fig. 2c and d, showing just one month of data, the clear difference between baseline periods and non-baseline events can be more clearly seen and one gets a sense of the degree to which methane and radon are correlated.

Flask measurements of CH₄ began at Cape Grim in 1984, while the record from the current AGAGE GC-MD (multi-gas chromatograph, multi-detector) in situ instrument system, which incorporates a Carle GC fitted with a flame ionisation detector (see Prinn et al., 2000) began late in 1993. Ambient methane measurements are made on discrete air samples every 40 min, taken alternately from a 75 and 10 m inlet for the majority of the study period. Ambient samples are bracketed by analysis of a calibration

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standard, and the resulting CH₄ record is reported on the Tohoku University scale (Aoki et al., 1992; Prinn et al., 2000).

During the period of observations used for this study, Cape Grim radon measurements were made using a number of detectors. From 1994 to 1997, a 9000 L two filter radon detector featuring a particle generator was used, operating at a nominal flow rate of 200 L min⁻¹ and with a response time of approximately 90 min (Whittlestone and Zahorowski, 1995). From 1997, a newly-designed 5000 L dual flow loop, two filter radon detector was commissioned, operating at a nominal flow rate of 285 L min⁻¹ and with a response time of 45 min (Whittlestone and Zahorowski, 1998). Later developments saw this new detector enhanced from a single-head design to two and eventually four heads in 2004, with corresponding improvements to sensitivity and lower limit of detection. Air was sampled from a 75 m inlet, the same height as the upper CH₄ observations. Raw radon counts were collected half-hourly and aggregated to hourly values during post processing. Detector sensitivity ranged from 0.6 to 1.2 counts per second per Bq m⁻³ during the period of measurements. Calibrations were performed monthly using a Pylon flow-through radon source (20.9 ± 0.8 kBq Radium-226), traceable to US National Institute of Standards and Technology (NIST) standards, and instrumental background checks were performed approximately every 3 months. The lower limit of detection for the Cape Grim radon detectors, defined as the radon concentration below which the statistical counting error exceeds 30 %, ranged from 6 to 10 mBq m⁻³.

For this study, CH₄ observations from 1994–2007 have been processed using the following steps:

1. We have linearly interpolated between the discrete measurements of atmospheric CH₄ every 40 min, to generate hourly CH₄ data, to facilitate comparisons with the hourly radon data, and the hourly values from the model simulations.
2. The CH₄ observations were then selected for baseline conditions by excluding all hours when the coincident radon measurement was greater than 100 mBq m⁻³.

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3. A smooth curve was then found through these baseline-selected CH₄ observations using the methodology described in Thoning et al. (1989). Specifically, the baseline data are fitted with a function consisting of a second order polynomial and four harmonics. This function fit is then subtracted from the baseline data and the resulting residuals are then filtered with a band-pass filter with a short term cut-off of 80 days. The original function fit is then added back to the filtered residuals to give a smooth curve fit through the data. This procedure is performed iteratively, and in each iteration, the individual hours that lie outside twice the standard deviation around the fit are excluded until the fit converges.
4. Lastly, the smooth curve fitted to the baseline data is subtracted from all CH₄ observations to give a timeseries of residuals. In most cases it is only the residuals from hours that have been flagged as non-baseline that are used for comparison with the model simulations.

3 Model simulations

This study uses simulation experiments that were run for the TransCom-CH₄ model intercomparison to investigate various methane flux estimates. The TransCom-CH₄ model intercomparison involved running nine tracers in a global atmospheric model for the years 1988–2007. The first six tracers used different methane emission scenarios. The remaining three tracers were radon, sulfur hexafluoride and methyl chloroform. In each methane case, chemical loss of methane was simulated using prescribed OH fields (with seasonal variations but no interannual variations) and prescribed loss rates to represent photolysis in the stratosphere. The emissions are described in detail and a global analysis of the results is presented in Patra et al. (2011). Other details can also be found online in the TransCom-CH₄ protocol (Patra et al., 2010).

3.1 Methane emission scenarios

The six methane scenarios were created by combining various estimates of anthropogenic, rice, biomass burning and wetland components in different ways (Table 1). Details are given in Patra et al. (2011) and we use the same emission scenario labels as used in that paper and across the TransCom-CH₄ project. The control (CTL) scenario uses anthropogenic fluxes as specified in the Emissions Database for Global Atmospheric Research (EDGAR) inventory, version 3.2 (Olivier and Berdowski, 2001) and includes fossil fuel, industrial, animal, fire, waste and biofuel emissions. Added to these fluxes are the flux estimates of biomass burning from the Global Fire Emissions Database (GFED), version 2 (van der Werf et al., 2006), wetland emissions (Matthews and Fung, 1987; Fung et al., 1991) and rice (Yan et al., 2009). Four alternative emissions scenarios change one or more of the CTL component fluxes. CTL_E4 uses EDGAR 4.0 (van Aardenne et al., 2001) for the anthropogenic component: BB (biomass burning) uses interannually varying biomass burning emissions when available (van der Werf et al., 2006); WLBB (wetland and biomass burning) additionally includes interannually varying wetland emissions (Ringeval et al., 2010); EXTRA uses model generated wetlands (as per WLBB) and rice emissions from the VISIT model (Ito and Inatomi, 2012). A final emissions scenario, INV, does not use fluxes from inventories or process models but those estimated by the atmospheric inversion of Bousquet et al. (2006). All six emissions scenarios use the same soil sink. Table 2 details which components of each scenario include interannual variability and in which years of the simulations.

Figure 3a shows the aggregated emissions for the region of southeastern Australia bounded by 135–155° E and 45–30° S which is the region shown in Fig. 1. The emissions show a seasonal cycle that is dominated by the wetland component for CTL, CTL_E4 and BB with higher emissions from May to October. The seasonality is much smaller for the WLBB and EXTRA scenarios with maximum fluxes in December and January due to biomass burning. The INV emissions show similar seasonality to the

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CTL emissions, though with smaller amplitude. It is worth noting that the inversion used to generate these fluxes included only baseline CH₄ data at Cape Grim, so there is no reason to expect the INV fluxes to fit the non-baseline record at Cape Grim better than the other flux scenarios.

Figure 3b shows the interannual variability of the prescribed emissions, again aggregated over the region shown in Fig. 1. The CTL fluxes show almost no change in time, while the CTL_E4 fluxes increase over time. The three fluxes that include interannually varying biomass burning; BB, WLBB and EXTRA, show peaks in 2003 and 2006 associated with significant summer fires in southeastern Australia. This will be discussed in more detail in Sect. 4.1. The INV fluxes show the greatest interannual variability in this region.

The prescribed TransCom-CH₄ methane emissions can be compared with those reported in Wang and Bentley (2002) and Fraser et al. (2011). To approximate our region of interest, from Wang and Bentley (2002) we sum their regions A, C and D, which extend slightly further west and north than our region. This gives total anthropogenic emissions (agriculture including cattle, the energy and transport sectors and waste management) of 3.39 Tg y⁻¹, using the methodology of the Australian National Greenhouse Gas Inventory (NGGI) coupled with statistical data for 1997 to give a spatially explicit representation of methane emissions. By adding in an estimate of methane uptake by Australian soils, the net anthropogenic flux used in Wang and Bentley (2002) across southeastern Australia is 3.09 Tg y⁻¹. From Fraser et al. (2011) we sum their anthropogenic emissions (agriculture including cattle (and rice, which is very small), the energy sector, waste management and a small amount of prescribed burning) from five regions; New South Wales, Australian Capital Territory, Victoria, Tasmania and South Australia, to give total anthropogenic emissions of 2.88 Tg y⁻¹. Fraser et al. (2011) take emissions from the EDGAR 3.2 inventory (Olivier et al., 2005), and scale them to the Australian NGGI. Both inventories have total annual CH₄ emissions which are closer to the annual emissions of the lower set of methane scenarios used here, but the inventories do not include natural fluxes while the TransCom methane scenarios do. In

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southeastern Australia, the major natural flux is from wetlands, but the magnitude of this flux is uncertain. The CTL based emission scenarios (CTL, CTL_E4 and BB) include a large 1.24 Tgy^{-1} component from wetlands for southeastern Australia, taken from Matthews and Fung (1987) and Fung et al. (1991). The WLBB and EXTRA emission scenarios take their estimates of wetland emissions from Ringeval et al. (2010), which for southeastern Australia, are much lower estimates than those of Matthews and Fung (1987) and Fung et al. (1991). Wetland emissions will be discussed further in Sects. 4.2 and 5.

3.2 Atmospheric models

We have run the TransCom-CH₄ simulations with two models: the CSIRO Conformal-Cubic Atmospheric Model (CCAM) (McGregor, 2005; McGregor and Dix, 2008) and the Australian Community Climate and Earth System Simulator (ACCESS) (Corbin and Law, 2011). CCAM has an approximately uniformly spaced conformal cubic grid and was run with a horizontal resolution of approximately 220 km, and 18 levels in the vertical. The horizontal components of the wind were nudged (Thatcher and McGregor, 2009) to NCEP analyses (Kalnay et al., 1996; Collier, 2004). This helps to ensure that simulated atmospheric concentrations of trace gases can be more realistically compared to observations on synoptic timescales.

ACCESS is derived from the UK Met Office Unified Model but has the land surface scheme replaced by the Community Atmosphere Biosphere Land Exchange (CABLE) model. ACCESS was run at 1.875° longitude by 1.25° latitude, with 38 levels in the vertical. Note that the ACCESS case submitted to the TransCom-CH₄ archive was run at lower resolution to meet the TransCom deadlines. ACCESS was run without any nudging, so that the only forcing is through sea surface temperatures. Consequently the output from this model can only be compared to observations on seasonal or longer timescales, or by analysing the statistics of synoptic behaviour. The ACCESS run used a 360 day calendar with 12 months of equal length.

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It is important to consider where the model output is sampled to be most comparable with Cape Grim observations, as well as to minimise differences between the two model simulations. The model sampling locations are shown in Fig. 1. The inset in Fig. 1 shows the spatial extent of each of the grid cells chosen to represent Cape Grim, giving a sense of their relative size. For both models we have sampled slightly to the north of Cape Grim. It is worth noting that in CCAM grid-cells are either all land or all ocean, whereas in ACCESS fractional land area is allowed.

Model timeseries are output hourly. The simulated concentrations are processed in the same manner as for the observations. Firstly coincident radon concentrations are used to select for baseline CH_4 , a smooth curve is fitted to the baseline data, and the baseline fit is removed from the timeseries. The residual concentrations are used for comparison with the observations.

4 Results

Initial analysis of the simulated CH_4 at Cape Grim highlighted two features. The first feature was two periods of 1–2 months duration with very high peak CH_4 concentrations for the flux scenarios that included interannually varying biomass burning. The second feature was a large difference in CH_4 concentrations in winter between different flux scenarios.

Figure 4 shows the monthly mean methane residuals in 2003 for the observations and all emission scenarios for both models. Both features mentioned above are most obvious in the CCAM results (Fig. 4b), where the model predicts exceptionally high methane concentrations in January and February for those tracers that include inter-annually varying biomass burning (BB, WLBB and EXTRA). Over the winter months, the three emission scenarios with default wetland emissions, (CTL, CTL_E4 and BB) all produce markedly higher methane concentrations than the observations or the remaining emission scenarios using modified wetland emissions or those based on the inversion of Bousquet et al. (2006).

4.1 Biomass burning

Figure 5 shows observed and CCAM simulated CH₄ residuals for January 2003 as well as observed H₂ and CO. High H₂ and CO are signatures of air influenced by biomass burning. Two simulated CH₄ tracers are shown, one which includes interannually varying biomass burning emissions (WLBB) and one that does not (CTL). The inclusion of “hot spots” of biomass burning emissions produces very large methane concentrations, much larger than those observed. Significant fires did occur during this period; in the eastern Victorian alpine region starting on 8 January 2003 and burning around 1.3 million hectares over close to two months (<http://www.depi.vic.gov.au/fire-and-emergencies/managing-risk-and-learning-about-managing-fire/bushfire-history/maps-of-past-bushfires>) and around Canberra between 18 and 21 January 2003. The observations indicate that the Victorian fire was likely seen briefly at Cape Grim on 11 January, when CH₄, H₂ and CO all had elevated concentrations. It is less clear what contribution biomass burning makes to other elevated methane events later in the month, when only small CO elevations are seen and H₂ signals do not rise above the instrumental noise (except perhaps around 25 January).

There are a number of reasons why the models may overestimate the impact of this fire at Cape Grim. Firstly, the biomass burning emissions are specified at the middle of each month and interpolated to the middle of the previous and following months. This means that a January fire is spread temporally into December and February. Indeed, WLBB also shows very large CH₄ concentrations in December 2002 and February 2003 (not shown). Secondly, the fire emissions were provided on a 1° × 1° grid and have been regridded to the lower resolutions of the atmospheric models. In reality, the active fire at any given time would have covered a much smaller area. Finally, the fire itself would modify the local circulation, with emissions likely distributed not just near the surface but throughout the entire lower troposphere. This is not captured in our simulations where the emissions are input only to the lowest model level. For instance,

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Sofiev et al. (2013), find that under Australian fire conditions, 90 % of mass is emitted from the surface up to 3 km altitude. We have performed some short tests, running CCAM from December 2002 to February 2003, with only biomass burning emissions and releasing them into different model levels or spreading them through a vertical column. In general we find the same elevated CH₄ events simulated at Cape Grim but with smaller magnitude as the emissions are inserted higher into the atmosphere.

4.2 Relationship between methane and radon: seasonal cycle

Given the variation in simulated winter CH₄ concentrations from the different flux scenarios, we would like to determine which gives a better comparison with the observations. To avoid biasing our analysis of the seasonality of methane by the very large concentrations caused by the interannually varying biomass burning fluxes, we remove the biomass burning contribution from BB, WLBB and EXTRA for the months December 2002 to February 2003 and also November 2006 to January 2007, when another large fire gives unrealistically high simulated concentrations of CH₄ at Cape Grim. This was achieved by subtracting (BB–CTL) from each of the three affected tracers, during the months in question.

To minimise the impact of any errors in modelled atmospheric transport, we consider the ratio of methane concentrations to radon concentrations. The timeseries of methane residuals at Cape Grim is reasonably well correlated with radon, with both showing significantly elevated concentrations when air parcels have travelled over continental Australia. For the period 1994–2007, the correlation coefficient between observed methane residuals and observed radon is 0.65. In the model simulations, correlations between methane and radon vary across methane scenarios between 0.81–0.87 for ACCESS and 0.88–0.89 for CCAM. The higher correlation coefficients for the modelled data set compared to the observed data set presumably reflect the reduced spatial variability of both CH₄ and radon in the gridded models compared to the real world.

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The strategy of using a methane to radon ratio to minimise transport errors relies on the assumption that both sources are similarly distributed. We note that this assumption is only loosely true. The methane emission scenarios we assess here have considerable spatial variability (though doubtless less than the real world), while the TransCom-CH₄ specified radon emissions are uniform over land. Land surface emissions of radon do vary (Griffiths et al., 2010), though much less than for methane. As a check on our sensitivity to radon spatial variability, we ran ACCESS for three years using a series of different radon flux fields taken from Griffiths et al. (2010). In general, neither the spatial variability nor the interannual variability appeared to have a significant impact on our sampled concentrations at Cape Grim. We found that the choice of grid cell was much more influential in the modelled radon results matching the observed radon results, hence our choice of ocean grid cells to the north of Cape Grim. One of the reasons for this is likely to be the coarseness of the grid cells. See Sect. 3.2 and refer to the inset in Fig. 1.

We examine the seasonal relationship between methane residuals and radon by fitting a linear relationship to all hourly methane-radon pairs in a given month across the 14 years of the simulation for which we have observational data for comparison (1994–2007). For clarity, Fig. 6 shows this fit for a single month only, January 2006, for observations and modelled CTL cases. We take this approach in order to compare the observations to both the CCAM and ACCESS simulations. At the time of this work, ACCESS meteorology was forced only with sea surface temperatures and ran on a 360 day calendar, as described in Sect. 3. This means that we do not expect the timing of individual “events” (non-baseline periods) in ACCESS to match the observations well enough for a direct comparison. We do however expect that the seasonal scale meteorology will be realistic enough to provide seasonal fetch changes that are comparable to the real meteorology and fetch patterns at Cape Grim. Averaging results up to a seasonal timescale allows direct comparison of all three datasets (the observations and both models). The observations show more scatter than the model output, and this is reflected in lower R^2 values, in this case 0.61 for the observations and 0.92

and 0.74 for CCAM and ACCESS respectively. The slope of the line gives the methane residual to radon ratio. We determine the ratio this way to preserve the temporal pairing of the methane and radon concentrations. We also calculate linear fits for each individual month as per Fig. 6, and use the standard deviation of the slopes for each of the 14 months as a measure of the uncertainty on the methane-radon ratios.

Figure 7 shows the mean seasonal cycle of the methane residual-radon ratio for each methane scenario, compared to observed ratios. The observed ratios are smallest in winter (June to August) and are largest in spring and summer (October to February). The modelled ratios show three patterns depending on the methane scenario. The CTL, CTL_E4 and BB scenarios show maximum ratios in winter, while the WLBB and EXTRA scenarios show minimum ratios in winter, which is more consistent with the observations. The INV scenario is intermediate between the other cases, also showing somewhat elevated winter ratios. For any given scenario, the ratios for the two different models compare well, much better than if either of the individual trace gases (methane or radon) are directly compared, as was the case in Fig. 4 where the monthly mean methane is shown for both models. This illustrates the benefit of using the radon simulation to account for some of the transport differences between the models.

Overall the WLBB and EXTRA scenarios give ratios that are a better fit to those observed. For the southeastern Australian region, the major difference from the group of CTL-based cases is in the representation of wetlands. These results suggest that the large winter wetland fluxes in the CTL-based scenarios taken from Matthews and Fung (1987) and Fung et al. (1991) are not realistic and annual mean fluxes should be close to the anthropogenic only inventory estimates noted in Sect. 3.1. For all scenarios there is a discrepancy between the observed and modelled ratios in the austral spring (September to November) with the observed ratios being larger than the modelled ones. This suggests that the methane flux scenarios tested here underestimate methane fluxes in spring. We will discuss this further in Sect. 5.

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5 Discussion and concluding remarks

Forward modelling of a range of methane flux scenarios gives us the opportunity to compare those results with the measurement record at Cape Grim for the same period, offering insights into which of the flux scenarios appear to be most representative of southeastern Australian methane fluxes. The WLBB and EXTRA flux scenarios appear to be the best fit to the observed data. Like Fraser et al. (2011), we find that anthropogenic methane emissions taken from inventories for southeastern Australia look quite reasonable in magnitude at around 2.5 Tgy^{-1} . However, there remain questions about the scale and timing of a wetland component to CH_4 emissions. Our analysis reveals that mismatches in the CTL, CTL_E4 and BB scenarios were due to high wetland emissions during the winter, suggesting that the wintertime wetland flux is over-estimated in these scenarios. The Cape Grim observations point to somewhat larger springtime fluxes than are represented in the WLBB or EXTRA emissions scenarios (or indeed any of the other tracers). Although the wintertime maxima of the CTL-style emissions scenarios (driven by a wetland emission component) is clearly not warranted by the observational data, they look to be about the right magnitude to explain the “short-fall” in spring for all the emission scenarios. A shift in wetland emissions from austral winter to spring may be plausible given that wetland methane emissions have both a soil moisture and soil temperature dependence, making it possible that southeastern Australian methane emissions from wetlands are highest in springtime when there is available moisture and warmer temperatures. Indeed, Bloom et al. (2012) use satellite column observations of CH_4 from the SCanning Imaging Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY) coupled with a measure of equivalent water height from the Gravity Recovery and Climate Experiment (GRACE) to model seasonal variability in wetland methane emissions. For our region of interest across the years 2003–2008, they find the minimum in methane emissions from southeastern Australian wetlands occurs in late autumn and winter with a rapid rise through spring giving a maximum in October and November, (A. Fraser, personal communication, 2012) in

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accordance with the Cape Grim observations. However, the magnitude they predict for this *wetland* flux is around 2.5 Tg y^{-1} . This seems larger than indicated by the Cape Grim data; of the six emission scenarios considered in this work, annual means for *total* CH_4 flux (including all anthropogenic emissions) in our defined region range between 2.4 (WLBB) and 4 (CTL) Tg y^{-1} (Fig. 3b). Moreover, the additional 1.6 Tg y^{-1} in the CTL emission scenario comes from a wintertime wetland flux that we find no evidence for in the observations. It should be noted however that in Fraser et al. (2011) the GRACE data used were scaled to match a prior emissions estimate. We therefore find that the GRACE data have plausible seasonality for southeastern Australia but that the magnitude is too large to offer a realistic assessment of the scale of southeastern Australian wetland emissions given the Cape Grim observations. Nevertheless, we believe the seasonality of the GRACE data lends credibility to the idea that a springtime wetland emission of around the magnitude represented in the CTL-style emissions scenarios for winter may be responsible for the discrepancy between our modelled WLBB results and the observations.

Although our hypothesis that the timing of wetland methane emissions in the inventories may be off by 2–3 months is plausible and supported by other data, we cannot entirely rule out another source for the additional austral spring methane emissions. For instance, ruminant emissions from cattle are the single biggest contributor to Australia's anthropogenic methane emissions. Seasonality in ruminant emissions that is not captured by the inventories, due to changes in feed or cattle number might also be responsible for the austral springtime maximum observed. This explanation is offered in Wang and Bentley (2002) to account for the spike in emissions from their region D (roughly equivalent to Victoria and NSW) during the springtime. However, although Wang and Bentley find an austral springtime peak in CH_4 that coincides with the timing of the maximum noted in this work, they find overall a significant reduction in the CH_4 inventory for southeastern Australia is required to reconcile it to the Cape Grim observations for 1997. The reduction proposed by Wang and Bentley of around 40% in the largest region, D is at odds with Fraser et al. (2011) and this work, which both

has been undertaken as part of the Australian Climate Change Science Program, funded jointly by the Department of the Environment, the Bureau of Meteorology and CSIRO. Model simulations were undertaken on the NCI National Facility in Canberra, Australia, which is supported by the Australian Commonwealth Government.

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Table 1. The broad components of the methane emission scenarios. The anthropogenic category includes fossil fuel, industrial, animal, fire, waste and biofuel emissions. See Patra et al. (2011) for further details of how the emission scenarios were constructed.

Code	Anthropogenic	Rice	Biomass burning	Wetlands
CTL	EDGAR3.2	Yan et al. (2009)	van der Werf et al. (2006)	Matthews and Fung (1987); Fung et al. (1991)
CTL_E4	EDGAR4.0	Yan et al. (2009)	van der Werf et al. (2006)	Matthews and Fung (1987); Fung et al. (1991)
BB	EDGAR3.2	Yan et al. (2009)	van der Werf et al. (2006)	Matthews and Fung (1987); Fung et al. (1991)
WLBB	EDGAR3.2	Yan et al. (2009)	van der Werf et al. (2006)	Ringeval et al. (2010)
EXTRA	EDGAR3.2	Ito and Inatomi (2012)	van der Werf et al. (2006)	Ringeval et al. (2010)
INV			LSCE Inversion Bousquet et al. (2006)	

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Table 2. Methane emission scenarios, indicating which components have interannual variations over different periods. A mean seasonal cycle is used outside the listed periods.

Code	Anthropogenic	Rice	Biomass burning	Wetlands
CTL	1990 : 1995 : 2000	No	No	No
CTL_E4	1990–2005	No	No	No
BB	As CTL	No	1996–2008	No
WLBB	As CTL	No	As BB	1994–2000
EXTRA	As CTL	1988–2008	As BB	1988–2008
INV		1988–2005		

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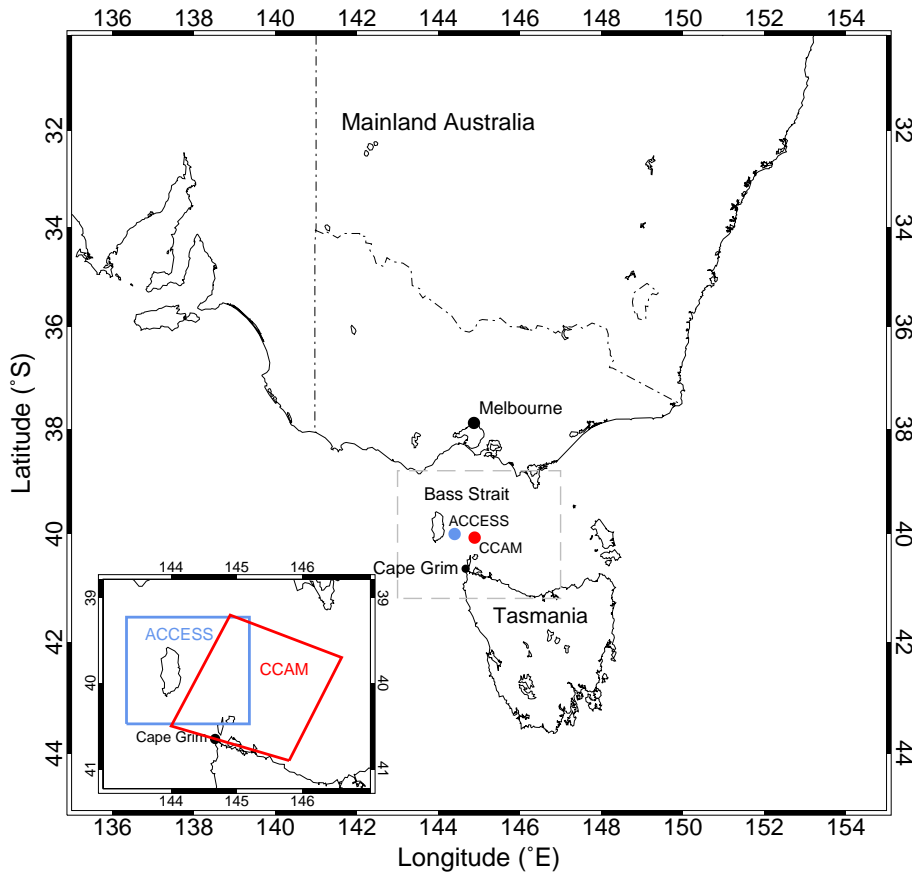


Figure 1. Map of South East Australia, showing the location of the Cape Grim Baseline Air Pollution Station, along with the ACCESS (blue) and CCAM (red) grid points selected to best represent Cape Grim. The choice of these grid points is discussed in Sect. 4.2. The inset shows the extent of each of the grid cells.

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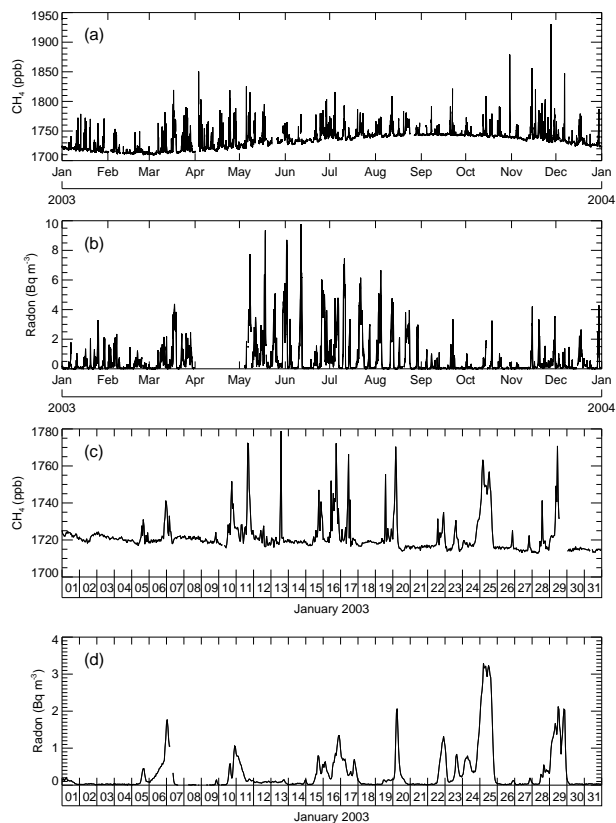


Figure 2. Cape Grim observations **(a)** 2003 methane data **(b)** 2003 radon data **(c)** January 2003 methane data and **(d)** January 2003 radon data.

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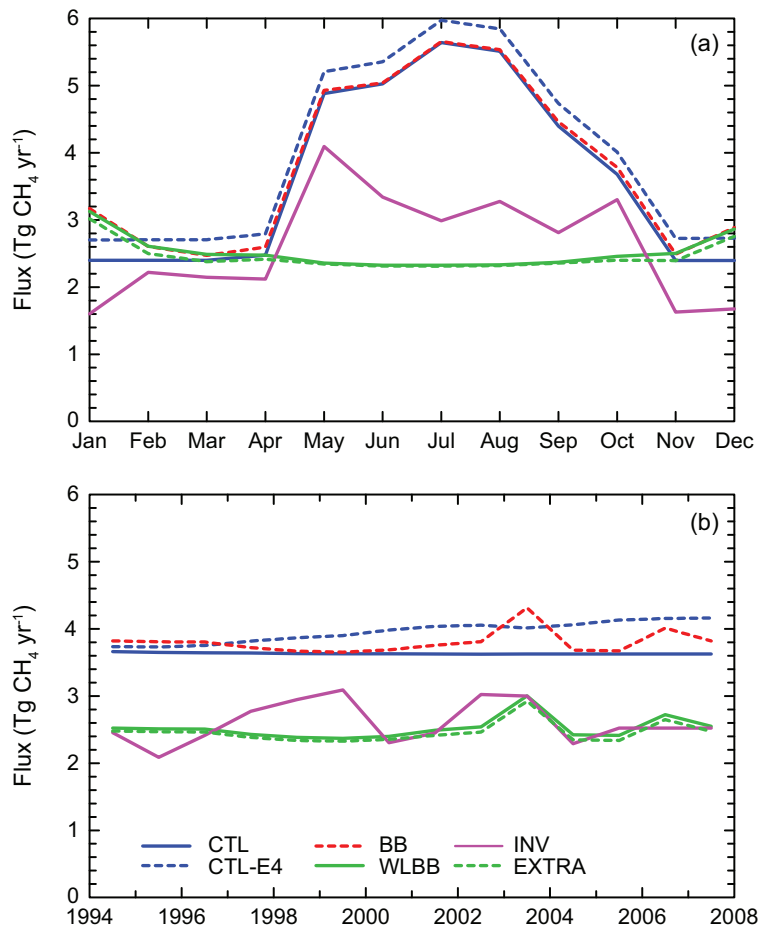


Figure 3. Seasonal cycle (a) and annual mean (b) methane fluxes for 1994–2007 integrated over the SE Australian region (135–155° E, 30–45° S). Six flux scenarios are shown as listed in the key.

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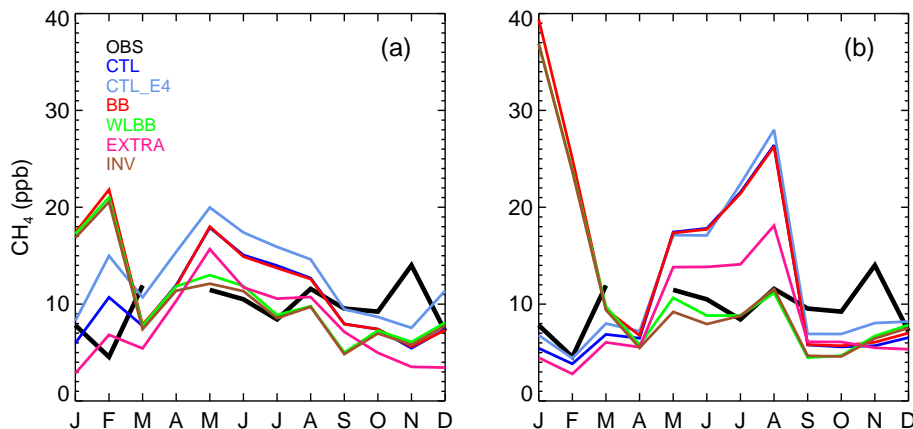


Figure 4. Monthly mean methane residuals at Cape Grim for the observations (black) and six emission scenarios (colours shown in key) **(a)** ACCESS **(b)** CCAM. Note that the observational mean residual for April is missing, due to lack of radon data with which to define the baseline threshold (rather than a lack of methane data). Refer Fig. 2.

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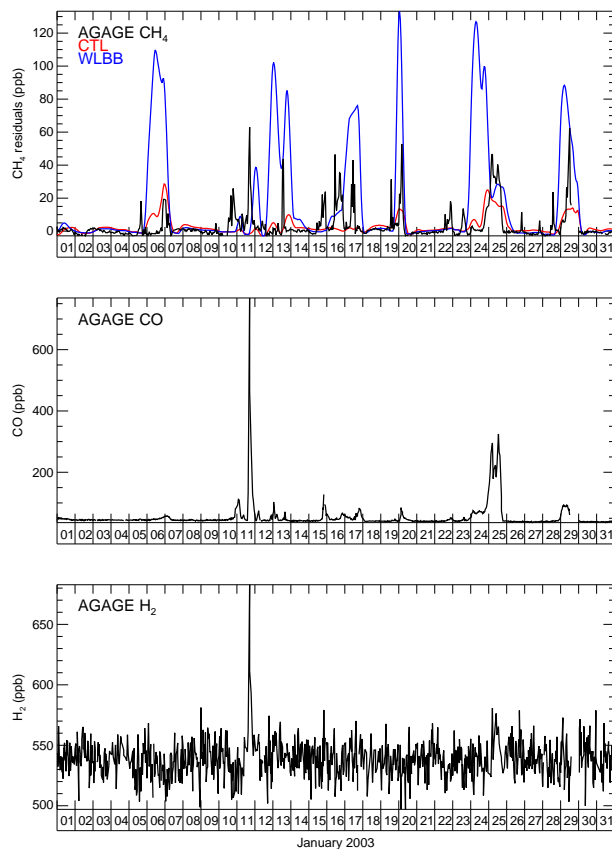


Figure 5. Cape Grim data for January 2003. Upper panel: observed (black) CH_4 residuals; CCAM simulated residuals for tracers CTL (red) and WLBB (blue). Middle panel: observed CO . Lower panel: observed H_2 .

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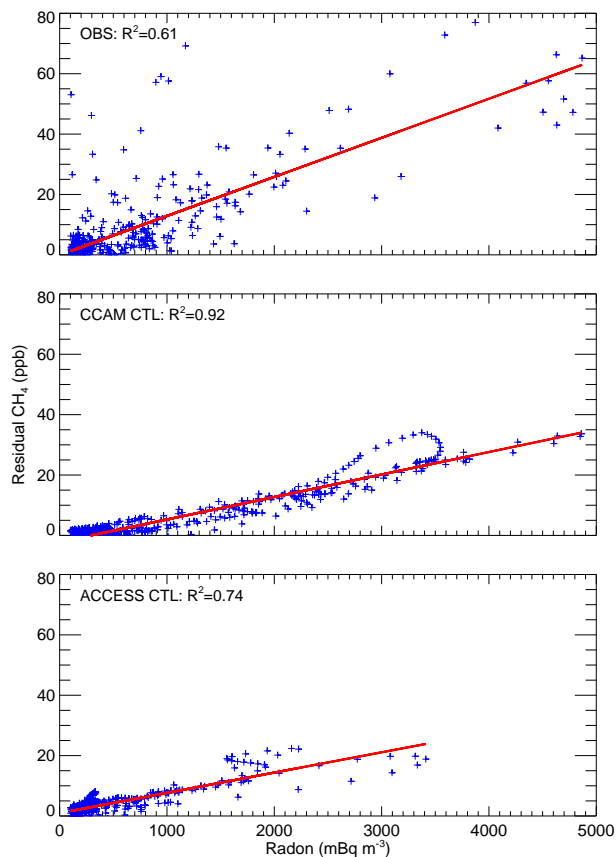


Figure 6. Scatter plots of methane to radon, with linear fits for January 2006. The upper panel shows the observational data, the middle panel the CCAM data and the lower panel the ACCESS data.

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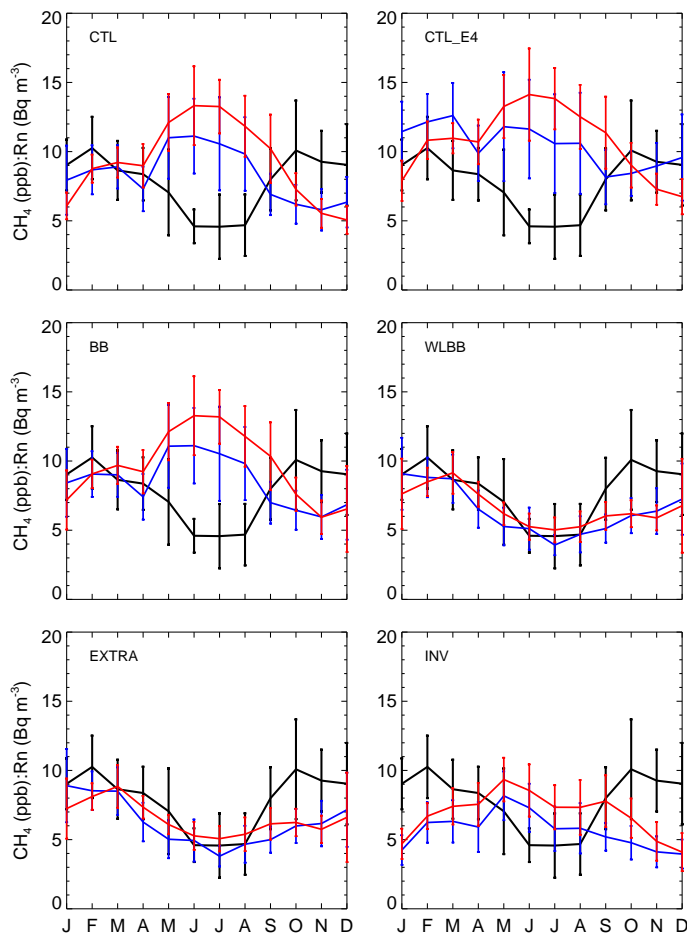


Figure 7. Mean seasonal cycle of methane to radon ratios for each of the six methane tracers. Observations are shown in black, ACCESS results in blue and CCAM results in red.

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