Author response to reviewer comments of "Detecting changes in Arctic methane emissions: limitations of the inter-polar difference of atmospheric mole fractions," by Dimdore-Miles et al

First, we thank the two reviewers for providing their comments on our analysis. Their comments have helped us to clarify the motivation and derivation of our analytical model. They have also helped us to put our work in the context of the wider challenges associated with detecting changes in Arctic emissions of methane. We apologize for the unnecessarily large number of typos, which are an egregious oversight. Below, we respond to individual reviewer comments (italics).

\subtitle{\underline{Reviewer #1}}

{\em This paper addresses the inter polar difference in methane. This concept was introduced as a "data-only" method to study changes in the methane emissions. Most notably, the observed step-like drop in 1991 was attributed to sudden changes in the emissions from the former Soviet Union. Likewise, it could be a useful metric to signal increases in Arctic methane emissions resulting from climate-related thawing of permafrost. The second part of the paper addresses the detectability of emission changes in the Arctic, and claims that changes and variability in emissions elsewhere (mid-latitude Northern Hemisphere, tropics) would blur the Arctic signal. Although not very surprising, this is a message that could deserve a paper. In the first part of the paper, the authors introduce an "analytic" model, mainly for illustration purposes. This part is not well thought of, unclear, contains errors, and does not contribute in any positive way to the paper. There is absolutely no link to the analytical model and the 3D model simulations with perturbed methane sources. The two points made around line 80 can be made without the analytical model, so I recommend (and this is expressed mildly) to remove this part entirely from the paper. }

We included the analytical model because it illustrated that the IPD as originally defined would unlikely be able to achieve what it claimed. It also helped to motivate the numerical experiments. Admittedly, we only included a summary of this model description, and did not include a link between the first and second equation. Because we strongly believe this model provides pedagogical value to the paper we have expanded our description of this model, and in doing so we show more clearly why accounting for atmospheric transport prevents the IPD from effectively isolating Arctic emissions of methane.

{\em Detailed argumentation: Method is simple, but contains a number of flaws. On equation (1): 1. IPD has units ppb, which clearly differs from L (mass per time) and B (also mass per time, but unit in the integral should be different. add: per degree latitude).}

This reviewer is correct. The IPD could equally well be described in terms of the mass or the mixing ratio of methane. For consistency with the original definition we have clarified this point in our derivation.

{\em 2. Now it looks there is a hard cut-of at r, where a unit of emissions decides to flow either to the NP or to the SP. In reality there is probably a latitude where emissions do not contribute to an IPD. North and South of this break-even point, an emission progressively contributes to setting an IPD. This issue is not reflected in the formula. }

This is a continuous integral in which air masses are free to flow either northward or southward. The point r can be evaluated anywhere between the North (r=0) and South (r=R) pole. If the role of atmospheric transport was negligible (equation 3) then the original definition of the IPD would be able to isolate $L(t)$. Evaluating the importance of these atmospheric transport terms is the main point of the study. Otherwise, we do not understand this reviewer's comment.

{\em The limits of the integral are not clear. Where is "r"? Somewhere between the North pole and the South pole?}

Correct, as per the definition in the paper (line 68 of the ACPD manuscript) and in the accompanying Figure 1.

{\em On equation (2): This now adds more confusion: I do not see how equation 2 follows from equation 1 (this relationship reduces to. . .?). Even more surprising is the change of sign, which suggests that an increase in emissions North of r would reduce the IPD. I really wonder if this analytical model has been tested using a realistic latitudinal emission distribution. I guess not!}

We have now provided a more thorough version of the derivation. Our interpretation of equation is different to this reviewer. We hope the revised derivation will remedy this.

{\em Further remarks and corrections to the manuscript are in the annotated pdf. Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2017-1041/acp-2017-1041-RC1- supplement.pdf}

Typos have been addressed. The main points about the equations are addressed above.

We have fixed a bug in the IPD error propagation. We thank the reviewer for pointing this out.

This reviewer raises the point about the sine weighting of the measurements (equation 3). To test the original definition of the IPD we have faithfully followed the definition provided by Dlugokencky.

\subtitle{\underline{Reviewer #2}}

{\em Detecting changes in Arctic emissions is critical in the framework of fast regional warming in the poles. CH4 emissions are expected to rise dramatically in the next decades with the timing, amplitude and localization remaining largely unknown. The inter-polar difference was proposed in the past to detect emission changes with a simple metric. The authors evaluate the relevancy and robustness of such method. }

{\em The IPD is known to have weaknesses and flaws. Only highlighting them with two simulations computed with a GCM clearly would not make a valuable scientific contribution to the community.}

We use the (revised) analytic model to highlight the main flaws of the original IPD approach, which allowed us to pinpoint the necessary computational experiments to run. Additional experiments (not shown) only serve to repeat our message. We hope this paper will dissuade future studies from using this metric for the purpose of isolating Arctic emissions.

{\em The only valuable content of the manuscript is to show that in idealistic (hence unlikely) conditions it would require >15 years to detect changes in Arctic emissions (hence forever under-realistic global emission scenario).}

We agree this is very valuable content. But this was only achieved by developing the analytical model that highlighted the numerical experiments we needed to run that subsequently allowed us to determine the timescales over which we could detect changes in Arctic emissions.

{\em A useful contribution would require (i) a thorough evaluation of the limitations of the method as defined by Dlugokencky et al., and}

We could not find any specific recommendations put forward by Dlugokencky. Nevertheless, our analytical model and accompanying numerical experiments show a fatal flaw in the use of the IPD that negates any further evaluation of this metric.

{\em (ii) quantified recommandations towards a less "simple-minded" but usable metrics. The poor scientific and presentation quality of the submitted manuscript hardly contribute to (i), and (ii) is fully missing.}

We believe there is no such usable metric and that is one of our points. This paper was about the role of atmospheric transport in the real IPD. Bayesian inference of Arctic emissions lead to uncertain estimates that reflect data sparsity of ground-based coverage and seasonal-bias of SWIR satellite observation (coverage is non-existent during winter months when the solar zenith angle precludes useful retrievals of column methane). The advent of active satellite sensors (e.g. MERLIN) capable of observing during day and night in all seasonal may open up the possibility of developing a more robust observational capability for the Arctic. We would then not require any data-led metric.

{\em Plus, the introduction and conclusion seem to be written independently of the main body of the manuscript: it is obvious that we need a long-term accurate network in the Arctic, and that we must analyze the data with some Bayesian inversions to get valuable insights, event though the manuscript discussion does not really proved it explicitly...}

We agree with some of this comment. We explicitly mention (abstract and main text) that we need to maintain that network of measurements in the Arctic. But there is no quick data-led metric that can isolate Arctic emissions of methane. Bayesian inversions do provide valuable insights but the data are not particularly dense over and upwind of the Arctic. Warming of the Arctic due to changes in climate is uneven so that could easily envisage a situation where methane emissions are far upwind of any

measurement. If anything we would argue for a high density of measurements over the Arctic if and only if someone was determined to develop a data-led metric.

{\em The authors are recommended to drastically improve the quality of the manuscript and to complement its scientific content. Here are suggestions: 1) propose combinations of emissions that would explain the observed IPD of the 30 past years, but would be unrealistic, hence proving the weaknesses of the IPD; 2) comment and analyze the observed IPD in term of real emissions as represented by your model; 3) assess the impact of the choice of stations (number, location) on the IPD; 4) suggest an improved IPD (probably including some transport but still being simple enough) that might point to changes with some confidence interval; 5) what impact TROPOMI and MERLIN could have on the IPD estimates? Having idealistic polar satellite coverage could make a comparison with the idealized IPD as defined in equations 1 and 2.}

We thank this reviewer for their suggestions. Suggestion 1) is the reverse of what we have studied. But we do not believe this approach would necessarily (dis)prove the IPD. The forward approach we have taken is cleaner and more straightforward to understand. The inverse approach, because it is a under-determined and ill-posed inverse problem due to poor data coverage and transport model errors, results in a range of possible outcomes. A similar argument can be made for suggestion #2. Re suggestion #3, choosing the stations to determine the IPD is not a useful line of questioning given our results. Without explicitly accounting for atmospheric transport a data-led metric will not work. Suggestion #4 is a Bayesian inversion but the data are too sparse. Ensemble model runs show a range of possible emissions for the Arctic (AMAP 2015 ref from main text). Suggestion #5 is mentioned in section 4 of the paper, although not in the context of the IPD. We have expanded on our description of satellite data based on our responses to reviewer comments.

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Detecting changes in Arctic methane emissions: limitations of the inter-polar difference of atmospheric mole fractions

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Abstract. We consider the utility of the annual inter-polar difference (IPD) as a metric for changes in Arctic ~~emission~~ emissions of methane (CH_4). The IPD has been previously defined as the difference between weighted annual means of CH_4 mole fraction data collected at polar stations (-53° >latitude>53°). This subtraction approach (IPD_Δ) implicitly assumes that extra-polar CH_4 emissions arrive within the same calendar year at both poles. Using an analytic approach we show that a more comprehensive description of the IPD includes terms corresponding to the atmospheric transport of air masses from/to lower latitudes to/from the polar regions. We show the importance of these ~~transport flux~~ atmospheric transport terms in understanding the IPD using idealized numerical experiments with the TM5 global 3-D atmospheric chemistry transport model that is run from 1980 to 2010. A northern mid-latitude pulse in January 1990, which increases prior emission distributions, arrives at the Arctic with a higher ~~mixing ratio~~ mole fraction and $\simeq$12 months earlier than at the Antarctic. The perturbation at the poles subsequently decays with an e-folding lifetime of $\simeq$4 years. A similarly timed pulse emitted from the tropics arrives with a higher value at the Antarctic $\simeq$11 months earlier than at the Arctic. This perturbation decays with an e-folding lifetime of $\simeq$7 years. These simulations demonstrate that the assumption of symmetric transport of extra-polar emissions to the poles is not realistic, resulting in considerable IPD_Δ variations due to variations in emissions and atmospheric transport. We assess how well the annual IPD can detect a constant annual growth rate of Arctic emissions for three scenarios, 0.5%, 1%, and 2%, superimposed on signals from lower latitudes, including random noise. We find that it can take up to 16 years to detect the smallest prescribed trend in Arctic emissions at the 95% confidence level. Scenarios with higher, but likely unrealistic, growth in Arctic emissions are detected in less than a decade. We argue that a more reliable measurement-driven ~~IPD metric would include~~ approach would require data collected from

all latitudes, emphasizing the importance of maintaining a global monitoring network to observe decadal changes in atmospheric greenhouse gases.

1 Introduction

Atmospheric methane (CH$_4$) is the second most important contributor to anthropogenic radiative forcing after carbon dioxide. Observed large-scale variations of atmospheric CH$_4$ (Nisbet et al., 2014) have evaded a definitive explanation due to ~~sparcity in data (Rigby et al., 2016; Turner et al., 2016; Schaefer et al., 2016; Kirsc~~ sparseness of data (Kirschke et al., 2013; Rigby et al., 2016; Turner et al., 2016; Schaefer et al., 2016; Saunois et al., 2016).

Atmospheric CH$_4$ is determined by anthropogenic and natural sources, and by loss from oxidation by the hydroxyl radical (OH) with smaller loss terms from soil microbes and oxidation by Cl. This results in an atmospheric lifetime of $\simeq$10 years. ~~Industrial anthropogenic~~ Anthropogenic CH$_4$ sources include leakage from the production and transport of oil and gas, coal mining, and biomass burning associated with agricultural practices and land use change. Microbial anthropogenic sources include ruminants, landfills, ~~rice cultivation, and biomass burning~~ and rice cultivation. The largest natural source is microbial emissions from wetlands, with smaller but significant contributions from wild ruminants, termites, wildfires, landfills, and geologic emissions (Kirschke et al., 2013; Saunois et al., 2016). Here, we focus on our ability to quantify changes in Arctic emissions using polar atmospheric mole fraction data.

Warming trends over the Arctic~~are~~, approximately twice the global mean (AMAP, 2015), ~~which is~~ are eventually expected to result in thawing of permafrost. Observational evidence shows that permafrost coverage has begun to shrink (Christensen et al., 2004; Reagan and Moridis, 2007). Arctic soils store an estimated 1700 GtC (Tarnocai et al., 2009). As the soil organic material thaws and decomposes it is expected that some fraction of this carbon will be released to the atmosphere as CH$_4$, depending on soil hydrology. Current understanding is that permafrost carbon will enter the atmosphere slowly over the next century, reaching a cumulative emission of 130–160 PgC (Schuur et al., 2015). If only 2% of this carbon is emitted as CH$_4$, annual Arctic emissions could approximately double by the end of the century from current estimates of 25 Tg CH$_4$/yr inferred from atmospheric inversions (AMAP, 2015). At present, using data from the current observing network there is no strong evidence to suggest large-scale changes in Arctic emissions (Sweeney et al., 2016).

The inter-polar difference (IPD) has been proposed as a sensitive indicator of changes in Arctic emissions that can be derived directly from network observations of atmospheric CH$_4$ mole fraction. The IPD, as previously defined (Dlugokencky et al., 2003), is the difference between weighted annual means of CH$_4$ mole fraction data collected at polar stations (-53° >latitude>53°) such as those from the NOAA Earth System Research Laboratory (ESRL) network (https://www.esrl.noaa.gov/gmd/dv/site/site_table2.php). Data from individual sites are weighted inversely by the sine of the station latitude and by the standard deviation of the data at a particular site. Hereafter, we denote this

subtraction method as IPD_Δ to distinguish it from the full description of the IPD, as described below. Dlugokencky et al. (2003) reported an abrupt drop in IPD_Δ during the early 1990s. They suggested

60   this magnitude of change was indicative of a 10 Tg CH_4/yr reduction, which they attribute to the collapse of fossil fuel production in Russia following the 1991 breakup of the Soviet Union (Dlugo-kencky et al., 2011). In more recent work, Dlugokencky et al. (2011) proposed that the IPD_Δ metric is potentially sensitive to changes in Arctic emissions as small as 3 Tg CH_4/yr, representing a value of 10% of northern wetland emissions. However, studies have reported little or no increase in IPD_Δ

65   between 1995 and 2010 (Figure 1, Dlugokencky et al. (2011, 2003)), a period during which rising Arctic temperatures were expected to lead to an increase in emissions (Mauritsen, 2016; McGuire et al., 2017). In this work, we examine how sensitive the IPD is to changing CH_4 emissions by using model simulations ~~as well as a simple~~ guided by results from an analytical approach.

~~To describe the full IPD, we define~~ First, we introduce an analytic model of the atmosphere to

70   demonstrate the atmospheric transport terms involved with the full IPD. We divide the world into three contiguous regions: Arctic, background, and Antarctica (Figure 2); the inter-polar meridional length, defined from the Arctic to the Antarctic, is given by R. We have a local Arctic source $L(t)$ (mass CH_4 per unit time) and an isolated inter-polar source B ~~emitted at position r and time t~~ (mass CH_4 per unit time) emitted at position r and time t. For simplicity, we assume that there is only one

75   measurement site in the Arctic and one in Antarctica, and have neglected the transport of $L(t)$ to the Antarctic.

The IPD in the general sense is then given by:

$$\mathrm{IPD}(t) = k \left(\underbrace{L(t) + \int_r^0 B(t - \tau_N(r'), r')dr'}_{\text{Signal measured at the Arctic site}} - \underbrace{\int_r^R B(t - \tau_S(r'), r')dr'}_{\text{Signal measured at the Antarctic site}} \right), \tag{1}$$

where τ_N and τ_S denote the time taken for source B at latitude r to reach the Arctic and Antarctic,

80   respectively. The variable k describes the mass to mole fraction conversion, following the mole fraction definition for IPD_Δ. The dummy variable r' has been introduced to avoid confusion in the integral. This relationship ~~reduces to:~~

$$\mathrm{IPD}(t) = L(t) - \int_r^0 \frac{dB}{dt}\Big|_{t-\tau_N} \tau_N \ dr' + \int_R^r \frac{dB}{dt}\Big|_{t-\tau_S} \tau_S \ dr'.$$

The purpose of this simple-minded derivation is to introduce the two independent atmospheric transport terms, which are not included in the definition of can be expanded (Appendix A):

$$
IPD(t) \; \widetilde{\overline{\overline{=}}} \; kL(t) + k \left(\int\limits_{r}^{R} \frac{dB(t,r')}{dt} \Big|_{t-\tau_S(r')} \tau_S(r') \; dr' - \int\limits_{r}^{0} \frac{dB(t,r')}{dt} \Big|_{t-\tau_N(r')} \tau_N(r') \; dr' \right) +
$$

$$
k \left(\int\limits_{r}^{0} B(t,r') \; dr' - \int\limits_{r}^{R} B(t,r') \; dr' \right). \tag{2}
$$

The analytical model highlights two grouped terms: the first term represents the integrated response of B as it travels northward and southward to the poles, and the second term represents differences in the response functions. The IPD_Δ . A consequence of these terms, is that there are only definition implicitly assumes these grouped terms are zero. Under these circumstances, there are two limiting cases in which the IPD_Δ can isolate $L(t)$: 1) emissions from B are constant in time(second and third R.H.S. terms become zero), and 2) B arrives at both poles simultaneously (second and third R. H.S. terms cancel out). with symmetric transport histories. Neither case is realistic on any time scale. Even point sources are often time-dependent. The characteristic timescale for inter-hemispheric transport of an air mass is $\simeq 1$ year (Holzer and Waugh, 2015). Using the annual IPD_Δ (Dlugokencky et al., 2003) we show that only a fortuitous set of circumstances would allow this metric to isolate $L(t)$.

In the next section, we describe the data and methods used previously to define IPD_Δ, and the model calculations we use to explore the importance of the transport flux termsatmospheric transport terms, as illustrated in equation 2. In section 3, we report the results from our numerical experiments. We conclude in section 4.

2 Data and Methods

2.1 Observed and Model IPD_Δ

To calculate the IPD_Δ, following Dlugokencky et al. (2011), we first group together a subset of NOAA ESRL global monitoring measurement sites that are located $-53° >$latitude$>53°$ (Table 2), and assign them as the North and South polar regions. For each polar region we calculate mean biweekly mole fractions across the stations, weighted inversely by station latitude and the standard deviation about the biweekly mean CH_4 mole fraction. Biweekly values of IPD_Δ are then averaged over a calendar year to determine the annual IPD_Δ, which has been used in previous studies.

We use biweekly CH_4 values determined from measurements of discrete air samples collected in flasks from the NOAA Cooperative Global Air Sampling Network (NOAA CGASN). Air samples (flasks) are collected at the sites and analysed for CH_4 at NOAA ESRL in Boulder, Colorado using a gas chromatograph with flame 220 ionization detection. Each sample aliquot is referenced to the WMO X2004 CH4 standard scale (Dlugokencky et al., 2005). Individual measurement uncertainties

115 are calculated based on analytical repeatability and the uncertainty in propagating the WMO CH$_4$ mole fraction standard scale. Analytical repeatability ~~has varied~~ varies between 0.8 to 2.3 ppb, ~~but~~ and has a mean value of approximately 2 ppb averaged over the measurement record~~is approximately 2 ppb~~. Uncertainty in scale propagation is based on a comparison of discrete flask-air and continuous measurements at the MLO and BRW observatories and has a fixed value 0.7 ppb. These two

120 values are added in quadrature to estimate the total measurement uncertainty, equivalent to a $\simeq 68\%$ confidence interval.

Five northern and two southern polar stations (Table 2) have data that cover the period discussed in previous studies (approximately 1986–2010) and a weekly resolution to calculate biweekly averages. We impute missing data filled using a two-stage approach. We use linear interpolation to

125 replace missing measurements from a given week and year with the average of the measurement values from the same week of the three preceding and subsequent years (to provide a climatological value but preserve long term trends in the data). If corresponding weekly measurements for the six ~~neighboring~~ neighbouring years are incomplete, we use a cubic spline interpolation. We calculate the uncertainties on the biweekly weighted concentration means from the polar regions using the

130 formula for the standard error $-\sigma_{\bar{x}}$ of a weighted mean μ (Taylor, 1997)~~:~~

$$\sigma_{\bar{x}}^2(\mu) = \frac{1}{\sum_i \left(\frac{1}{\sigma_i + sin(\phi_i)}\right)^2},$$

$, \sigma_{\bar{x}}^2(\mu) = 1/\sum_i \left(\frac{1}{\sigma_i + sin(\phi_i)}\right)^2,$ where the denominator represents weights assigned to each station i as a function of biweekly mole fraction standard deviation σ_i and the latitude ϕ_i of the station. We propagate these errors to determine the error on the annual IPD, following Dlugokencky et al. (2011).

135 We calculate the corresponding model IPD$_\Delta$ values by sampling TM5 (described below) at the time and location of each NOAA ESRL observation and processing the values as ~~describe~~ described above for the observations.

2.2 Numerical Experiments

~~We~~ Building on the terms evaluated using our analytical model (equation 2) we use the TM5 at-

140 mospheric transport model (Krol et al., 2005) to ~~examine the~~ 1) examine the atmospheric transport fluxes of emissions to the polar regions~~(equation 2), and to~~, and 2) determine the sensitivity of the IPD$_\Delta$ to different emission distributions. ~~For our~~

For our numerical experiments, we run the TM5 model using a horizontal spatial resolution of $2°$ (latitude) and $3°$ (longitude), ~~forced with~~ driven by meteorological fields from the European Center

145 for Medium Range Weather Forecast (ECMWF) ERA-Interim reanalysis. Fossil fuel and agricultural emission estimates are taken from the EDGAR3.2 inventory (Olivier et al., 2005) with modifications (Schwietzke et al., 2016). Natural emissions are based on the prior values used by CarbonTracker-CH$_4$ (Bergamaschi et al., 2005; Bruhwiler et al., 2014). Bruhwiler et al. (2014) reported posterior CH$_4$ emission estimates for high northern latitudes that were 20–30% smaller than prior values,

which we use in our current experiments. An important consequence of our using these prior values is that the model IPD_Δ values have a positive bias compared to values determined by CH_4 mole fraction measurements.

We ~~ran a set~~ run a suite of targeted numerical experiments to test the sensitivity of the IPD_Δ to pulsed and noisy variations from mid-latitude and tropical emission sources. ~~We also considered~~ The noise-variation experiments mostly explore the role of the first grouped term in the IPD (equation 2) and the single-pulse experiments mostly explore the role of the second grouped term in the IPD (equation 2). In practice, both sets of experiments integrate information from both grouped atmospheric terms. We also consider experiments that included Arctic emissions with different constant growth rates and realistic variations in lower latitude emissions. As a control we ~~ran~~ use a simulation with constant emissions. Appendix B includes a presentation of the time series used to calculate the IPD_Δ from our experiments.

We ~~initialized~~ initialize our TM5 numerical experiments from 1980 using initial conditions defined by the observed North-South distribution of CH_4 in the early 1980s. ~~We ran each experiment~~ Each experiment is run from 1980 to 2010, ~~and sampled mole fractions~~ with mole fractions sampled at the time and location of the network observations.

Control Run

Figure 1 shows that the ~~simulated~~ model IPD_Δ for the control run is higher than observed values, as explained above. The model IPD_Δ also shows less variability than observed values~~, particularly~~. Variations of IPD_Δ in the early 1990s ~~that is~~ have been attributed to a rapid decline in fossil fuel production following the 1991 breakup of the Soviet Union (Dlugokencky et al., 2011). We determine the model response to changes in emissions (as described below) by subtracting the control run from the perturbed emissions runs.

Pulsed Emission Runs

To investigate the impact of a sustained continental-scale change in emissions on the weighted polar means and the IPD_Δ metric, we run the control experiment configuration but during 1990 we increase emissions by an amount that is evenly distributed throughout the year. In the first pulse experiment, we increase existing mid-latitude emissions over the contiguous USA by 10 Tg CH_4. In the second experiment, we increase existing tropical land sources (within $\pm 30°$) by 20 Tg CH_4. We present polar mole fraction time series produced using the control and pulsed experiments shown in Appendix B.

Random Noise Emission Runs

To investigate the role of intra- and inter- annual variations of emission sources on the IPD_Δ we re-run the two pulse experiments but superimpose standard uniform distribution noise $\mathcal{U}(0,1)$ on

the emissions. We conduct two runs of TM5: one with a noise function of amplitude 10 Tg on US
emissions and another with a function of amplitude 20 Tg on tropical sources. These experiments
help us to determine the observability of changes in mid-latitude and tropical sources at the poles
and whether the IPD$_\Delta$ can isolate $L(t)$.

Arctic Emission Variation

To investigate the ability of IPD$_\Delta$ to detect a constant annual growth rate of Arctic emissions, we use
the control experiment configuration but in three separate experiments we increase Arctic emission
by 0.5%, 1% and 2% on an annual basis. Emissions are mostly limited to summer months (June–
August) when the soil surface is typically not frozen.

3   Results

Figure 3 summarizes the results from our pulsed emission experiments. The model response at both
poles to the 1990 pulse peaks rapidly and then falls off approximately exponentially over several
years. The Northern Region tracer represents the sum of $L(t)$ and the ~~first atmospheric transport term~~
second and third atmospheric transport terms in equation 2, and the Southern Region tracer repre-
sents the ~~second atmospheric transport~~ first and fourth atmospheric transport terms in that equation.
   Figure 3A shows that the mid-latitude pulse of 10 Tg CH$_4$ results in a larger change at the northern
polar stations (7.3 ppb peak) than at the southern polar stations (3.0 ppb peak). This reflects the
longer transport time for the pulse to reach the southern stations during which time the pulse becomes
more diffuse. More importantly, for the interpretation of the IPD$_\Delta$ we find that the northern polar
stations experience the majority of the pulse 0.96 years before the southern polar stations. After 1991
the pulse responses decay with e-folding lifetimes of 4.43 years and 8.94 years in the Northern and
Southern polar stations, respectively. Figure 3C shows that the difference in pulse response at the
poles decays from a maximum value in 1992 with an e-folding time of approximately 0.36 years.
   Figure 3B shows that the peak of the 20 Tg CH$_4$ tropical pulse reaches the southern polar region
0.92 years earlier than the northern polar region. This results in a larger change in southern polar CH$_4$
mole fractions (8.3 ppb peak) compared to corresponding values over the northern polar regions.
The earlier transit of the tropical pulse to the southern polar region reflects that much of the prior
tropical CH$_4$ fluxes that we perturb lie in the southern hemisphere. Responses to the tropical pulse
decay after 1992 with e-folding lifetimes of 8.65 years and 7.07 years for the northern and southern
regions, respectively. The significant transport delay and disparity in responses means that an annual
mean subtraction of northern and southern polar stations (IPD$_\Delta$) will not remove the influence of
the mid-latitude pulse and isolate $L(t)$ as previously assumed.
   Figure 4A shows that signal variations that we might expect from the atmospheric transport of
intra- and inter- annual ~~variations~~ changes in ~~emissions sources can dominate any signal that might~~

~~exist in~~ emission sources (first grouped term in equation 2 can dominate the IPD$_\Delta$ signal. In response
to noise superimposed on mid-latitude USA emissions, changes in biweekly IPD$_\Delta$ values have a
mean value of 3.0 ppb (range ~~6.0~~ -0.1–6.0 ppb). The corresponding changes in the annual IPD$_\Delta$
has a mean value of 3.0 ppb (range ~~5.4–0.3~~0.3–5.4 ppb). The response of the biweekly IPD$_\Delta$ to noise
on tropical emissions have a mean value of -2.8 ppb (range -12.8–5.6 ppb) and the corresponding
response to the annual IPD$_\Delta$ has a mean value of -2.7 ppb (range -4.7–0.6 ppb). These experiments
show that the IPD$_\Delta$ is susceptible to variations in inter-polar sources.

Figure 4B shows that IPD$_\Delta$ is sensitive to changes in $L(t)$, as expected, with a near-perfect cor-
relation. We find only a modest response of IPD$_\Delta$ to large percentage increases in Arctic emissions:
annual increases of 0.5%, 1%, and 2% in Arctic emissions result in changes of 0.09, 0.17 and 0.35
ppb per year in IPD$_\Delta$. IPD$_\Delta$ ~~variation that might~~ variations that might be expected from intra- and
inter- annual variations in mid-latitude and tropical ~~source~~ sources are typically much larger than the
signal associated with changes in $L(t)$. We find that the IPD$_\Delta$ in the presence of a constant Arctic
annual growth rate and intra- and inter- variations in mid-latitude and tropical emissions can detect
a 0.5% annual growth rate within 11–16 years to 95% confidence level (Weatherhead et al., 1998).
Table 1 summarizes our results for different growth rates but generally the larger the Arctic growth
rate the shorter it takes to detect the signal, as expected. The IPD$_\Delta$ is more susceptible to variations
in northern mid-latitude sources than tropical sources, as described above. These results represent
a best-case scenario for the IPD$_\Delta$. In practice, there are also intra- and inter- annual variations as-
sociated with $L(t)$ that will complicate the interpretation of the IPD$_\Delta$ and likely increase the time
necessary to detect a statistical significant signal.

4   Concluding Remarks

We critically assessed the inter-polar difference (IPD) as a robust metric for changes in Arctic emis-
sions. The IPD has been previously defined as the difference between weighted means of atmo-
spheric CH$_4$ time series collected in the northern and southern polar regions (IPD$_\Delta$). A compre-
hensive definition of the IPD includes at least two additional terms associated with atmospheric
transport. Using the TM5 atmospheric transport model we highlighted the importance of these atmo-
spheric transport terms. We showed that IPD$_\Delta$ has a limited capacity to isolate ~~any change~~ changes
in Arctic emissions.

We show that an inter-polar emission (here, we have evaluated emissions from midlatitudes and
the tropics) generally arrives at one ~~polar~~ pole earlier the other pole by approximately one year,
invalidating a key assumption of the IPD$_\Delta$. We also show that a small amount of noise on prior mid-
latitude or tropical sources that might be expected due to intra- and inter- annual source variations is
not removed in the calculation of the IPD$_\Delta$. While the IPD$_\Delta$ can detect ~~an unrealistic~~ a constant Arc-

tic annual growth rate of emissions, any additional variation due to mid-latitude or tropical sources can delay detection of a statistical significant signal by up to 16 years.

Our study highlights the need for sustaining a spatially distributed and intercalibrated observation
255 network for the early detection of changes in Arctic CH$_4$ emissions. The ability to detect and quantify trends in these emissions directly from observations is attractive, but in reality we need to account for variations in extra-polar fluxes and differential atmospheric transport rates to the poles. This effectively demands the use of a model of atmospheric transport, which must be assessed using global distributed observations.

260 A Bayesian inference method that integrates information from prior knowledge and measurements is an ideal approach for quantifying changes in Arctic CH$_4$ emissions, but assumes a) reliable characterization of model error and b) measurements that are sensitive to all major sources. Model error characterization is an ongoing process. Estimating CH$_4$ emissions from atmospheric measurements is an undetermined (i.e., number of fluxes to be estimated
265 $\gg$ number of observations available) and an ill-posed (i.e., several different solutions exist that are equally consistent with the available measurements) inverse problem. Prior emissions are required to regularize the inverse problem, allowing posterior fluxes to be determined that are consistent with prior knowledge and atmospheric CH$_4$ measurements, and their respective uncertainties. Ground-based measurements represent invaluable information to determine atmospheric variations of CH$_4$, but the
270 spatial density of these data limits the resolution of corresponding posterior emission estimates to long temporal and large spatial scales. Column observations from satellites represent new, finer-scale information about atmospheric CH$_4$, but they are generally less sensitive to surface processes than ground-based data. Daily global observations of atmospheric CH$_4$ from the latest of these satellite instruments, TROPOMI aboard Sentinel-5P (launched in late
275 2017), promise to confront current understanding about Arctic emissions of CH$_4$ described by land-surface models and bottom-up emission inventories. Passive satellite sensors, such as TROPOMI, rely on reflected sunlight so they are limited by cloudy scenes and by low-light conditions during boreal winter months. Active space-borne sensors (e.g. Methane Remote Sensing Lidar Mission, MERLIN, due for launch in 2021) that employ onboard lasers to make mea-
280 surements of atmospheric CH$_4$ have the potential to provide useful observations day and night and throughout the year over the Arctic. The sensitivity of MERLIN to projected changes in Arctic emissions of CH$_4$ is still to be determined. Another major challenge associated with satellite observations is cross-calibrating sensors to develop self-consistent timeseries than can be used to study trends over timescales longer than the expected
285 lifetime of a satellite instrument (nominally <5 years). Even with access to all these data, it is clear that no simple, robust data metric exists without integrating the effects of atmospheric transport, but data-led analyses remains critical for underpinning knowledge of current and future changes in Arctic CH$_4$ emissions.

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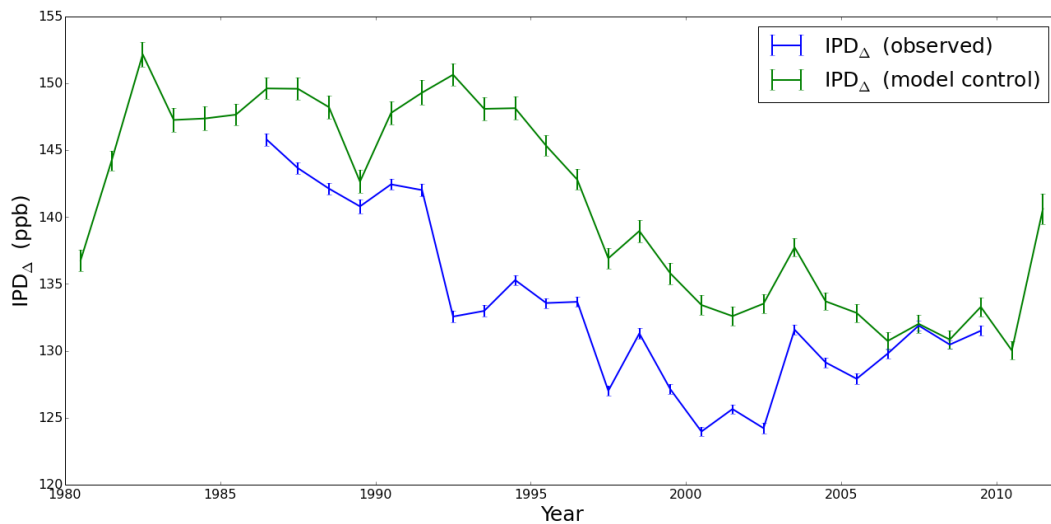
Figure 1. Annual mean IPD$_\Delta$ values (ppb) determined by NOAA ESRL and TM5 model atmospheric CH$_4$ mole fractions using data collected at seven geographical locations (Table 2). Vertical bars denote the one standard deviation associated with the annual mean.
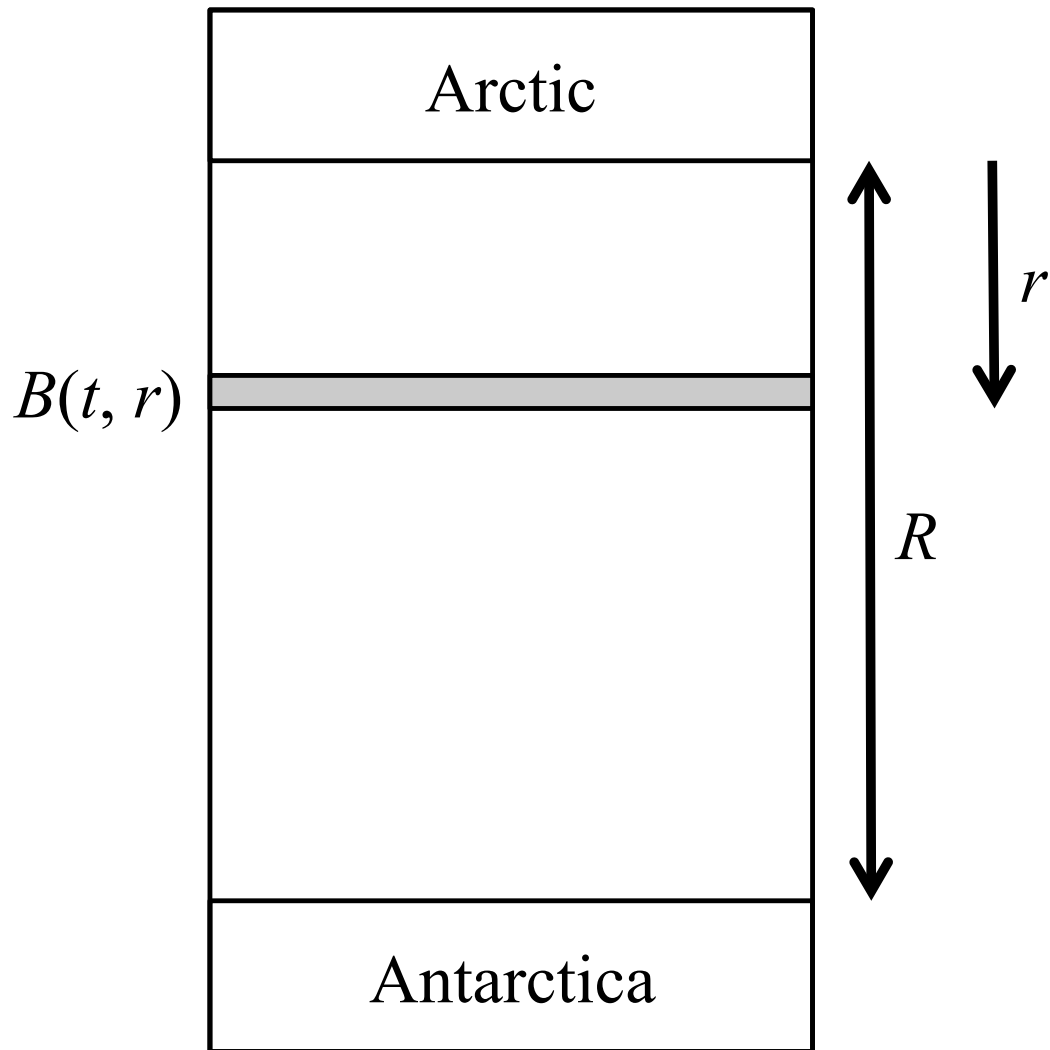
Figure 2. Schematic to describe how an inter-polar source $B(t,r)$ would be viewed at measurement sites in the Arctic and Antarctic.
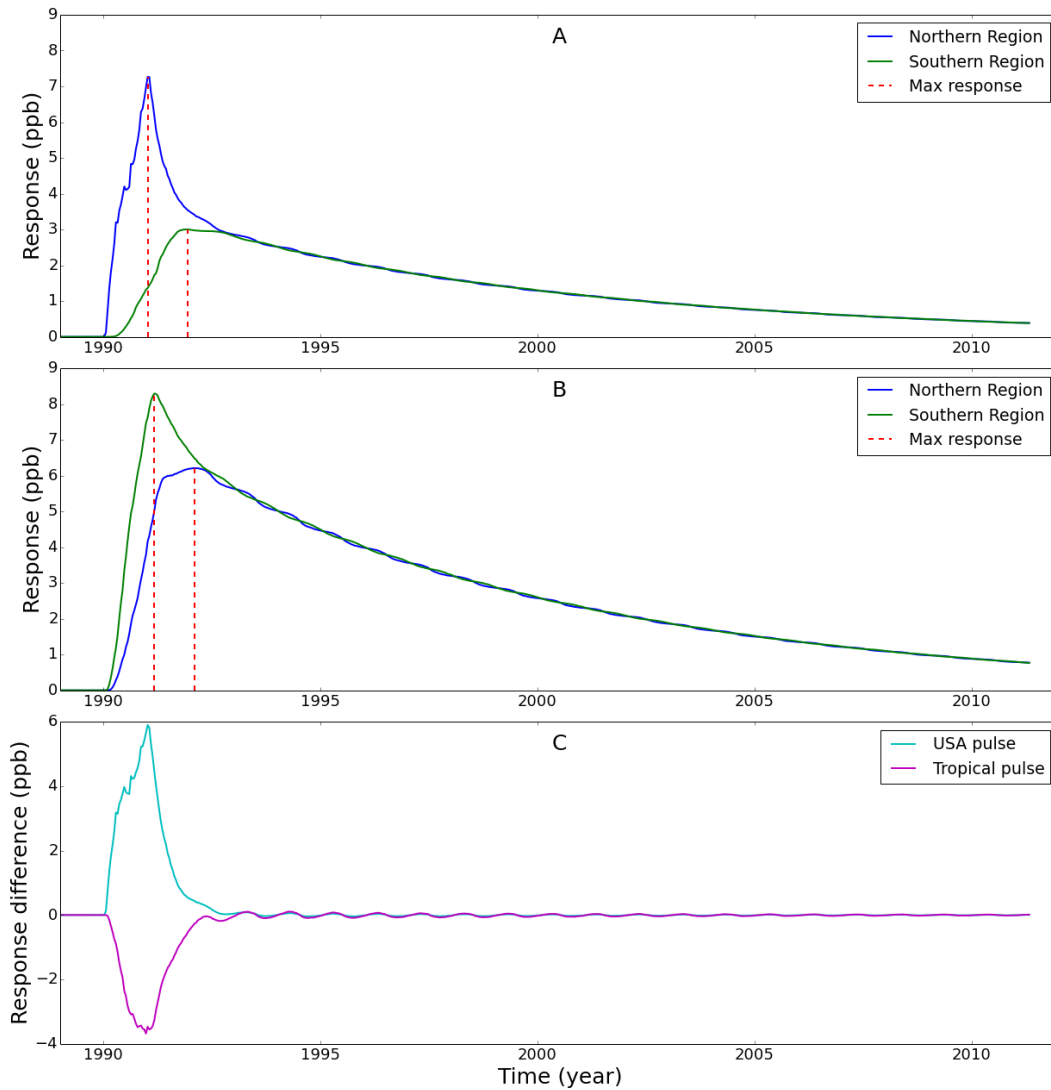
Figure 3. The model response of atmospheric CH_4 mole fraction sampled at northern and southern polar regions to a pulsed emission at (A) mid-latitude USA and (B) the tropics. Panel C shows the IPD response to these mid-latitude and tropical perturbations. In the interest of clarity, we omit error bars from the plots. Vertical red dashed lines denote the peak response time for each polar region.
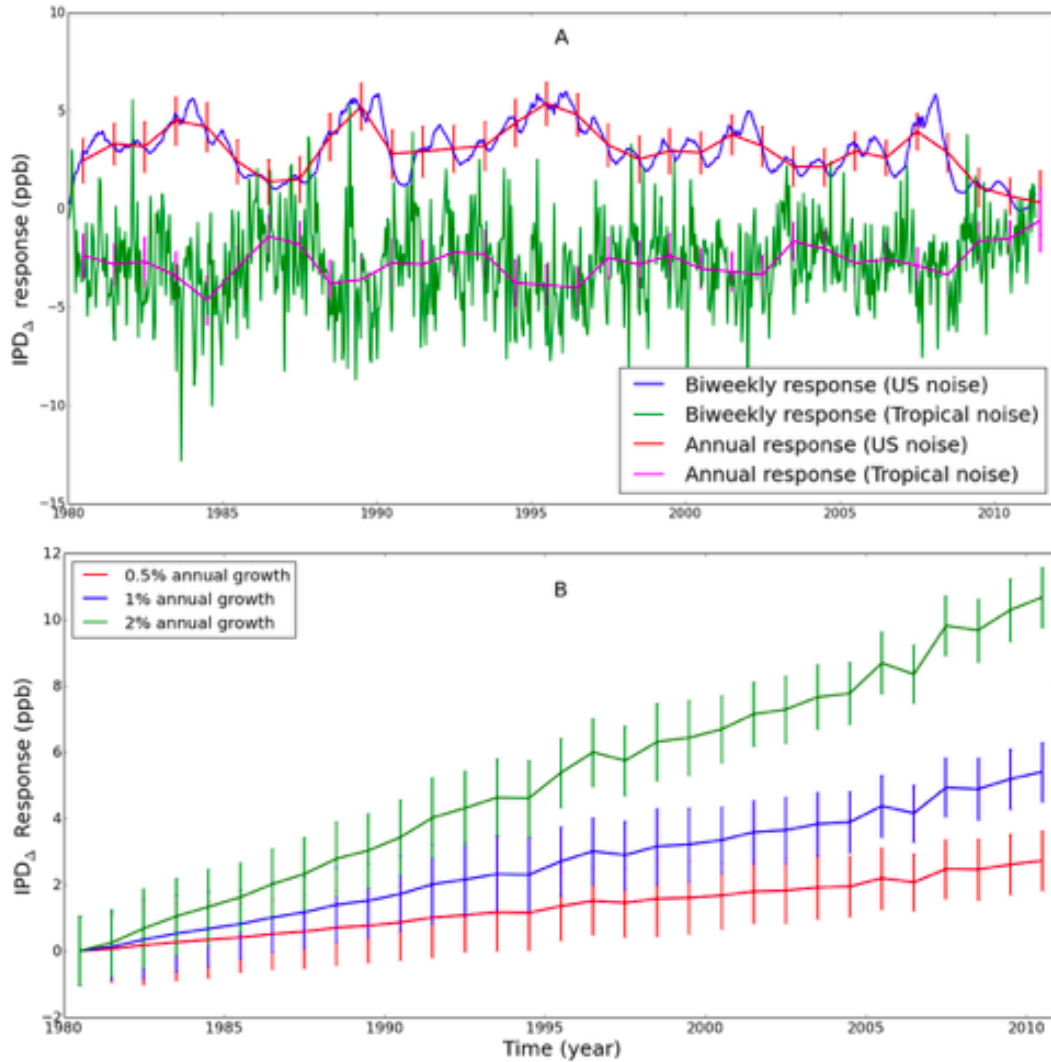
Figure 4. (A) Biweekly and annual model response of the IPD$_\Delta$ to changes in standard uniform distribution of random noise on prior mid-latitude USA and tropical emissions. (B) annual mean response of IPD$_\Delta$ to constant growth of Arctic emissions. Vertical lines denote uncertainties on responses.

Table 1. Details of the polar station used to calculated the IPD_Δ.

Station Name	Abbreviation	Latitude (°)	Longitude (°)	Altitude (m)
Barrow, Alaska	BRW	71.32	-156.61	11.0
Alert, Canada	ALT	82.45	-62.51	190.0
Cold Bay, Alaska	CBA	55.21	-162.72	21.3
Ocean Station M, Norway	STM	66.00	2.00	0.0
Shemya Island, Alaska	SHM	52.71	174.13	23.0
South Pole, Antarctica	SPO	-89.98	-24.80	2810.0
Palmer Station, Antarctica	PSA	-64.92	-64.00	10.0

Table 2. Number of years required to detect a statistically significant trend in Arctic emissions in the presence of inter-polar emission variations.

Arctic Emission Annual Growth rate	Inter-polar Variation	Years to detect trend in IPD$_\triangle$
0.5%	USA (10 Tg amplitude random noise)	16.3
	Tropics (20 Tg amplitude random noisee)	10.9
1.0%	USA (10 Tg amplitude random noise)	10.3
	Tropics (20 Tg amplitude random noise)	6.9
2.0%	USA (10 Tg amplitude random noise)	6.5
	Tropics (20 Tg amplitude random noise)	4.3

Appendix A: Development of Analytical Model for the IPD

Here, we include the additional steps required to derive equation 2 in the main text. At a time t the generalized IPD is given by:

$$IPD(t) = L(t) + \int_r^0 B(t - \tau_N(r'), r') dr' - \int_r^R B(t - \tau_S(r'), r') dr' \tag{A1}$$

In practice, the response function B that describes the northward and southward transport pathways will be different. First, the fraction of emitted mass going northward will be different to the fraction going southward. Second, in the absence of any additional sources that lie between r and each pole the atmospheric transport mechanisms acting on the southward and northward airmass will be different. We have chosen to neglect these effects, although to some extent differences in atmospheric transport differences are crudely described by τ_N and τ_S.

We can perform a time expansion of the two integral terms as follows:

$$B((t+dt), r') = B(t, r') + \frac{dB(t, r')}{dt}\Big|_{t+dt, r} dt. \tag{A2}$$

Assuming a slow-varying source $B(t)$ (comparable or slower than transport timescales) we can use that expansion:

$$IPD(t) \approxeq L(t) + \int_r^0 \left(B(t, r') - \frac{dB(t, r')}{dt}\Big|_{t-\tau_N(r')} \tau_N(r') \right) dr'$$

$$- \int_r^R \left(B(t, r') - \frac{dB(t, r')}{dt}\Big|_{t-\tau_S(r')} \tau_S(r') \right) dr'. \tag{A3}$$

We can then describe the IPD as:

$$IPD(t) \approxeq L(t) + \left(\int_r^R \frac{dB(t, r')}{dt}\Big|_{t-\tau_S(r')} \tau_S(r') \ dr' - \int_r^0 \frac{dB(t, r')}{dt}\Big|_{t-\tau_N(r')} \tau_N(r') \ dr' \right) +$$

$$\left(\int_r^0 B(t, r') \ dr' - \int_r^R B(t, r') \ dr' \right). \tag{A4}$$

The first grouped term describes the integrated response of the source term B as it is transported from the point of emission r. We assume the only source outside of the Arctic is at r so the second grouped term becomes zero. Note that with differences in the response functions, as described above, the second grouped-term would be non-zero and further invalidate the use of $IPD_\triangle$.

Appendix B: IPD plots

For completeness, here we include the plots that complement the analysis reported in the main text. Figure 5 shows the model CH_4 mole fraction corresponding to the weighted mean values at northern and southern polar

region used to calculate the IPD$_\Delta$ in the control and pulsed experiments using the TM5. Figure 6 shows values of the annual mean IPD$_\Delta$ corresponding to our numerical experiments.
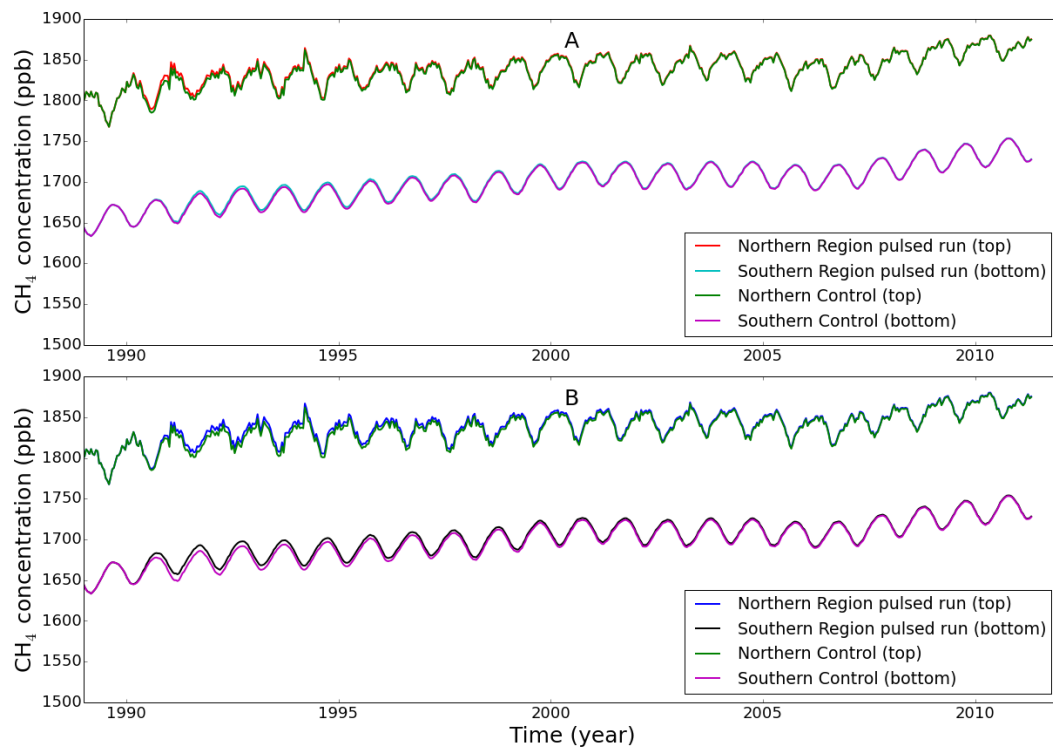
Figure 5. TM5 model CH$_4$ mole fractions (ppb) sampled at polar regions (Table 2) and weighted inversely by station latitude and standard deviation of the data at that site (see main text). Panel A shows the response of a 10 Tg pulse over mid-latitude USA in 1990 over the northern and southern pole. Panel B shows the response of a 20 Tg pulse over the tropics during 1990.
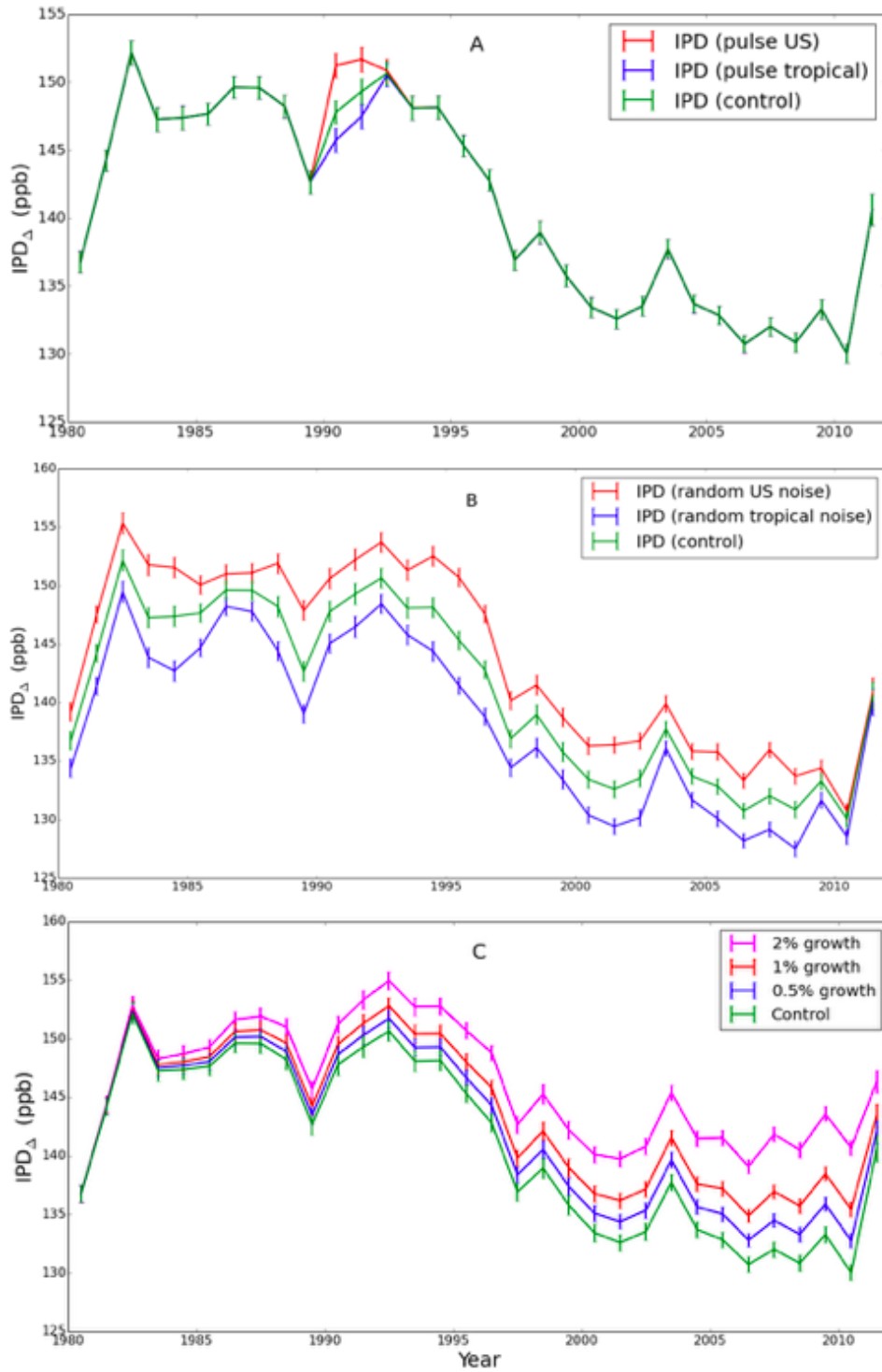
Figure 6. The model IPD$_\Delta$ corresponding to the control and all the sensitivity experiments described in the main text.