InGOS 2015 talk notes:

1. Methane is arguably the most interesting of the LLGHGs because of its diverse range of sources, baffling trends over the past 3 decades, and alarming claims about its role in climate change, especially in the Arctic. Unfortunately, there is a good deal of confusion about it too. Here I will look at what we’ve learned for large spatial scales from more than 30 years of measurements from our global cooperative air sampling network, and I will also update its growth rate, which includes a large annual increase in 2014.

2. As way of an outline, I will talk about our sampling and measurement program, including QC. Then, I’ll look at long-term global trends, which, with some assumptions, allow us to calculate total global emissions. Then we’ll look at what we can learn from IAV – I will use an old study of the eruption of Mt. Pinatubo to illustrate that. We’ll look at the renewed increase in CH4 since 2007 and possible causes. And, finally, I will summarize.

3. Our network consists of about 60 globally-distributed air sampling sites shown as the red symbols on this map. Samples are collected weekly with a portable sampler and returned to Boulder for analysis by GC/FID. All data are available with a short delay from our FTP server.

4. We employ various forms of QC on the data. In addition to monthly measurements of target tanks near ambient and quarterly measurements of tanks that cover a broad range of CH4 mole fraction, we compare regularly with our continuous measurements are MLO and BRW. We also measure 2 “test flasks” each day we do analysis; these are filled with air at ambient CH4 mole fraction. Recent results are shown in the RH plot. Then, we compare with other GAW colleagues, this, for example, a comparison of our measurement from a discrete air sample with continuous measurements by EC at ALT. Agree is good, but varies with time, sometimes falling outside GAW recommended compatibility targets.

5. An example of CH4 data from one site is shown here for BRW. In blue and green are data flagged because of sampling or analysis problems, or not believed representative of large well-mixed volumes of the atmosphere. In red ae retained values that are used in the analysis. Data from all sites in our network are combined with data from other labs for global inversions of emissions, or they are used as boundary conditions in continental scale studies. There are 2 curve fits, a deseasonalized trend calculated by filtering the data and eliminating periods less than one year, and a smooth curve, where variability less than one month is filtered. These curves are used to calculate zonal averages by extracting values from the smooth curves of about 40 BG sites at equally spaced intervals and then fitting these values as a function of latitude.

6. The result is plotted here, where you see CH4 mole fraction vs latitude and time. Spacing in latitude is at intervals of sine latitude = 0.05 to ensure proper spatial weighting, and in time about 1 week. All the main features of the global CH4 budget are shown here qualitatively, the strong latitude gradient; the seasonal cycles at N and S latitudes, nearly out of phase with each other; the minimal seasonal cycle in the tropics, and the trend. But to get quantitative information, we use zonal averages calculated form this matrix.

7. Here, for example, are globally-averaged CH4 mole fractions shown here in the top panel in blue where CH4 mole fraction is plotted vs time. In red is the deseasonalized trend, and its time derivative is plotted in red as ppb yr-1 in the bottom panel. What we observe is a decreasing growth rate from the start of our measurements until 2006. Superimposed on top of that is interannual variability in the GR that has information about processes. But, what does the decreasing GR mean? If we assume that CH4’s lifetime has been constant, it means we’ve been approaching steady state. To illustrate that, I’ve fitted the global averages with a function that describes the approach to steady state and solved it for the lifetime. I get 9.3 yr, very close to recent estimates of 9.1±0.9 yr. The t-derivative of this function is plotted in the bottom panel. It is very sensitive, yet it agrees well with the observed GR. What all this means is, assuming the lifetime has been constant, there is not trend in emissions. Can we explore that further?

8. Yes, we can calculate emissions from observed atmospheric burden and annual increase and estimate of the CH4 lifetime (using literature value of 9.1±0.9 yr) with a 1-box mass balance of the global atmosphere as shown at the top. This slide is a bit busy, so let me explain what is plotted. First, in dark blue, are global annual emissions calculated with the mass balance equation. Through 2006, there is no trend in emissions, they remained constant at ~544 Tg CH4 yr‑1, consistent with the analysis I just described, then increasing afterwards. This does not mean that emissions from individual source sectors are not changing, only that if some are increasing others are decreasing. Starting in 2007, there is a step change in emissions; over the entire measurement period, the average emissions are 551 Tg. In red are emissions based on EDGAR v4.2 ft and fixed, annually repeating natural emissions. While the magnitude of natural emissions may be incorrect, this demonstrates that trends in emissions in EDGAR are not consistent with observations. Now, when we look at the period since 2006, we see a change has occurred in CH4’s global budget, especially prominent in 2014, where I’ve calculated emissions from 2 estimates of the annual increase in 2014. I do not yet have a reasonable explanation for this change.

9. Skip.

10. Now look at IAV of CH4. One strong signal happening after a period where CH4 variability was minimal was in 1991. I think every one has heard this story before, but it is such a nice example of how atmospheric measurements can be used to test understanding of processes that I will repeat it. When Mt. Pinatubo erupted in the Philippines, it injected SO2 and ash into the upper troposphere and lower stratosphere, but probably little CH4. It was the SO2 and ash that affected CH4 (and CO) growth rate as follows.

11. Oxidation of CH4 initiated by OH is the main sink for CH4 and also the largest term in the global CH4 budget. So, even small changes in OH can have large impacts on the CH4 GR. Just to remind everyone, OH is produced photochemically through the following process: O3 is photolyzed by UV light of sufficient E to produce electronically-excited O(1D). Most of the time, O(1D) collides with N2 or O2 to produce the electronic ground state, O(3P), but about 1% or so react with H2O to produce 2 OH radicals. OH is also produced through some recycling reactions involving HO2 (hydroperoxy radical). A key step in 1° production is O(1D) production, the rate of which is given by the following: *j* is the photolysis rate constant; the terms that determine it are the UV flux, absorption cross section, and quantum yield for the process. All are a fn of wavelength and F is also a fn of zenith angle. The only parameter that could be affected by Pinatubo is F. We performed radiative transfer calculations by distributing the SO2 throughout a narrow band in the tropics, then allowing it to decay with a lifetime of 31 days to form sulfate aerosols with a much lifetime of about 1 year. The impacts of direct absorption of UV by SO2 and scattering by sulfate on j were calculated and are plotted in the next figure.

12. Here, the relative change in the rate coefficient for production of O(1D) is plotted as a fn of time relative to a 10-year average climatological background. The initial change in O(1D) production, about 12% with rapid initial decay reflecting the fast oxidation of SO2 to sulfate, and then slower decay corresponding to the 6-mo to 1-yr lifetime of sulfate results in an initial 8 to 10% decrease in OH. Changes in stratospheric O3 from heterogeneous chemistry involving the aerosols would have cancelled some of the decrease by direct absorption alone. Our calculation neglected the role of the ash; it was likely short-lived, and we also did not know its optical properties. This is one example of many where we could investigate the impacts of changing GR. Cooling after the eruption, about 0.6°C, would have affected the rate coefficient for OH + CH4 (~2%/degree) as would changes in H2O as water responded following the Clausius Clapyron equation (~7%/degree). Those effects are also not accounted for.

13. The impact of the change in OH can be seen in residuals from a function fitted to both CH4 and CO, as I described before. The magnitude of the signal is similar for CH4 and CO in 1991 as expected.

14. I would like to consider the approach to SS and the renewed increase in CH4 together. Here, in the top panel, you see the global means (blue), the deseasonalized trend (red), and the fn that describes the approach to SS (green) as before. The bottom panel shows the difference between the trend curve in red and the SS curve in green. For most of the record, the residuals are near zero, with strong positive excursions superimposed on top of them in 1991, 1998, and 2003. From 2004-2006, the residuals are below zero trending downwards until 2007 when everything changes. This is not a temporary anomaly, but rather wholescale change in CH4’s global budget, the causes of which are unknown. Still, I suspect there are a number of factors that contribute.

15. Coming back to the residuals for CO and CH4, we have good evidence that increased BB is not the major source of increased CH4. In the tropics, there may be a small signal in CO in MOPPIT in late-2006, but it is not very large. In the surface measurements, through the period when CH4 began to increase, we see only noise in CO, with no signal that can be related to BB. Given the large emission ratios of CO/CH4, BB is likely not a significant influence, although a small contribution can not be ruled out and may have affected the isotopic composition of CH4. There were some clear signals for BB in CO at high N latitudes, for example 2002 and 2003, but they did not have a significant impact on CH4.

16. We get some hint as to the causes by looking at stable carbon isotope data for CH4 from our colleagues at INSTAAR. Plotted here are globally averaged CH4 in the top panel and δ13C in the bottom panel. The big picture is like this: as CH4 increases, 13C gets more negative, suggesting a shift to global mass-weighted emissions that are depleted in 13C from microbial sources. At finer scale, we see that atmospheric CH4 began increasing well before the decline in δ13C started. Also, the δ13C decrease stalled from ~2012 to 2013, but was renewed in 2014 when rate of increase in atmospheric CH4 accelerated.

17. We may be able to get a qualitative idea of where emissions have changed by looking at zonally-averaged GR as a fn of latitude plotted as contours shown here. Warm colors are areas of large growth and cool colors low growth. The transition from blue to green is a GR of zero ppb/yr. One thing you have to keep in mind is that, for equal changes in emissions, signals are going to be much larger at high latitudes (i.e., the Arctic) than in the tropics, because emissions in the tropics are diluted into a deeper, larger area than at high latitudes. As I’ve described before, at the start of the CH4 increase in 2007, the strongest signals were in the Arctic and tropics. In the Arctic, there was also a clear signal to CH4 that was depleted in 13C at Alert, our northern-most site. Since then, it looks like on-again/off-again tropical signals until 2014, which looks a lot like 2007/2008.

18. For the increased emissions to be depleted in 13C, they are likely from a biological source like wetlands. Wetland emissions in the Arctic are strongly dependent on T, but, in the tropics, precipitation is the dominant driver of variability with wetland inundation strongly correlated with precipitation. TL: The top panel shows the CH4 anomaly from the SS fn (LH y-axis) as before and monthly precipitation anomalies for the tropics (RH y-axis). In the bottom panel, monthly SO index is plotted in red, with a 5-month running mean in blue. Prior to 2007, there were on-again, off-again El Ninos with strong negative precip anomalies in the tropics. Since 2007, we’ve experienced a string of La Ninas which have resulted in positive precip anomalies in the tropics. I suspect that has resulted in increased wetland area and length of the season they are inundated resulting in increased CH4 emissions. We have some evidence of this for the eastern Amazon based on estimates of fluxes for the relatively dry and wet periods. Fluxes estimated from individual profiles are quite variable, but they were about 40% greater in each season in the wet years than the dry years.

19. There has also been a lot of talk about the role of the Arctic in contributing to the change in CH4 GR. I don’t think Arctic processes are contributing significantly yet. The top R panel shows changes in the CH4 latitude gradient referenced to SPO for the late-1990s minus the mid-1980s as a result of decreased emissions in EDGAR after economic changes in the fSU calculated by Sander Houweling using TM3. The bottom panel shows the interpolar difference, that is differences in annual means between polar N and polar S zonal averages. The change from 1991 to 1992 is quite dramatic, but more importantly, it has not recovered. This, I think provides strong observational evidence that the Arctic processes are not contributing significantly to this new state of the global CH4 budget.

20. Summary: With the assumption that there has been no trend in CH4 lifetime, it suggests no trend in emissions. I think there is still a good deal to learn about CH4 budget processes from IAV. We’ve seen increased GR and increased E since 2007 that, based on multiple tracers, is related to increased E from tropical wetlands. ENSO plays an important role in this, so it will be interesting to see what happens with the CH4 GR as we head into a very strong El Nino. Finally, the CH4 GR surged in 2014 at 12-14 ppb. Although it was a warm yet and wet in portions of the Amazon, the detailed causes are not yet clear.