Author Comment #1

The analysis of the attribution experiments in the ACPD paper contained some erroneous calculations associated with equilibrium methane concentrations for experiment #2 (CH4), which we have corrected. We have also updated the analysis in several other ways. These updates change our results and interpretations somewhat, but the general conclusions of the paper remain the same. The erroneous values were the result of an over-simplification which we have now addressed in much more thorough detail with the analysis described here. We also have added a more detailed comparison with the previous studies of Shindell et al (2005, 2009). This comment summarises this updated analysis, which has been incorporated into the revised paper.

Calculating equilibrium methane concentrations in the attribution experiments

All of the 1850s-2000s attribution experiments were carried out with fixed methane concentrations. However, in the NOx, CO and NMVOC attribution experiments, we want to diagnose how methane (and ozone) concentrations would have changed if methane *emissions* were fixed, and methane concentrations were free to adjust. Similarly, for the CH_4 experiment, we want to diagnose how methane and ozone concentrations would adjust to a change in CH_4 emissions. Experiments were carried out with fixed, rather than varying methane concentrations, as these experiments spin-up quickly (i.e. within about a year), and are thus relatively easily performed by the models. Experiments with free-running methane concentrations would take several methane lifetimes to adjust (i.e. decades), and are thus currently computationally unfeasible for a multi-model intercomparison project like the one performed here. This means that the methane and ozone concentrations directly output from the models from the attribution experiments do not reflect equilibrium conditions, and need to be adjusted.

For example, in the attribution experiment where anthropogenic NOx emissions are reduced to 1850s levels, the methane concentration is held fixed at 1751 ppb. Because NOx concentrations are significantly lower in this experiment, OH concentrations are also lower, and methane destruction is reduced. If methane concentration was a free variable, and methane emissions were kept fixed, then clearly methane concentrations would rise in response to the lower OH. It is this level that methane would rise to – the equilibrium methane concentration – that we wish to estimate for each experiment. Because methane needs to be adjusted, and it is an ozone precursor, we also need to estimate the ozone adjustment that would occur as a consequence of the methane adjustment.

The equilibrium methane concentration can be estimated by using the methane lifetime diagnosed from each attribution experiment, as although the methane concentration is fixed, the methane lifetime (τ) does respond, as OH concentrations, and hence the flux through CH_4+OH, changes. We can calculate equilibrium methane concentrations ($[CH_4]_{eq}$) using the formula:

$$[CH_4]_{eq} = [CH_4]_{base} (\tau_{att} / \tau_{base})^f$$

where the subscript 'base' refers to the base year 2000s experiment, and the subscript 'att' refers to the attribution experiment, and f is the model's CH_4-OH feedback factor (Prather, 1996). The above formula is taken from Fiore et al (2009), and is also used in West et al (2007) (NB it appears in the Auxiliary Material of this latter paper in an incorrect form, with the ratio of lifetimes inverted); its

scientific basis originates in Fuglestvedt et al. (1999). Application of this formula yields equilibrium methane concentrations for the NOx, CO and NMVOC attribution experiments.

The methane attribution experiment (#2: 1850CH4) has to be treated somewhat differently. For this experiment, the base is no longer the 2000s. Instead, we can consider its base to be the 'All 1850s' experiment (still year 2000s climate) (#0). It can then be treated as an attribution type experiment where the NOx, CO and NMVOC emissions are set to 2000s levels, but methane concentrations are held fixed at 1850s levels (791 ppb). This is equivalent to a 2000s experiment with only methane changed. Thus to calculate an equilibrium methane concentration for these experiments, the base values come from the All 1850s experiment (#0). This method was not followed in the calculations for this experiment in the ACPD paper, and consequently the results presented were erroneous.

We have added a further refinement to the analysis. In the ACPD paper calculations, we assumed a CH_4-OH feedback factor (f) equal to 1.35 for all models. But we can calculate this factor for each model, using the base 2000s experiment (#0) and 1850CH4 experiment (#2), using:

$$f = 1/(1-s), \qquad \text{where } s = \delta \ln \tau / \delta \ln [CH_4]$$

This yields the values of f in the following Table (these values have been incorporated into Table 7):

Model	f
oslo2	1.281
ncarc	1.345
stoch	1.283
umcam	1.234
tm5__	1.321
hadec	1.281

These values are similar to/slightly lower than literature values (e.g., Prather et al, 2001: range 1.33-1.45). It should be noted that the delta methane (and delta lifetime) is the change from 1850s to 2000s. These are quite large changes – ideally the calculation of the feedback factor would be for a small perturbation (e.g., 10% changes or thereabouts). This may explain some of the differences in f compared to earlier calculations, and it should be noted that f probably varies slightly with time (due to changes in atmospheric composition), so use of a constant value for both the 1850s and 2000s is another approximation.

In the ACPD version of the paper, the calculated equilibrium methane concentrations for the methane attribution experiment (#2: 1850CH4) are erroneous (ACPD Table 7). These erroneous values propagate into ACPD Table 8, and also into ACPD Table 9. Revised versions of Tables 7-9 are presented below.

The calculations in the new Tables 8 and 9 incorporate another refinement. In the ACPD version of the paper, the ozone RF component associated with the adjustment of CH_4 to equilibrium was calculated assuming a simple linear relationship between change in CH_4 and change in O_3 from attribution experiment #2. However, Wild et al. (2012) have shown that there is a small non-linearity in this relation, and quantified it using model experiments performed as part of the Hemispheric

Transport of Air Pollution (HTAP) project. In our revised analysis, we estimate the change in O_3 associated with the adjustment of CH_4 to equilibrium using the Wild et al. (2012) relationship.

The difference between the CH_4 RF calculated for a change in concentration from 791 to 1751 ppb (i.e. the observed CH_4 change between the 1850s and the 2000s) of 427 mW m^{-2}, and the sum of the individual components we have calculated (300 mW m^{-2}) is still quite large – we have now included an extra row in Table 9 'Other factors'. These 'other factors' are due to non-linear interactions in both the chemistry models and in the radiative forcing calculations that are not accounted for by the simple linear sum of the results from these attribution experiments. They may also be partly due to using a fixed value of f in our calculations of equilibrium methane concentrations. We will refer to these 'other factors' as 'non-linear interactions between emissions' in the rest of the text.

For O_3 RF, the non-linear interactions between emissions appear to be minor, as the linear sum of the O_3 RFs from the CH_4, NOx, CO and NMVOC experiments is equal to the O_3 RF for 1850-2000 (Table 9). Table 9 reports this for the mean of the six models, but the same linearity is also approximately found for individual models. Non-linear interactions between emissions appear to be much more important for the CH_4 RF (Table 9).

These changes somewhat alter the calculated attribution percentages for the 1850s-2000s tropospheric ozone RF from those presented in the ACPD paper. Based on the six models that performed the attribution experiments, the mean percentages are now methane (44%), nitrogen oxides (31%), carbon monoxide (15%), non-methane volatile organic compounds (10%). The contributions of these categories to the 1850s-2000s methane RF are also presented in Table 9 (it is not sensible to quote percentages as the NOx contribution is negative). These values are compared with the earlier work of Shindell et al (2005, 2009) in (new) Tables 10 and 11, included below.

As can be seen in Tables 10 and 11, there are some differences between this work and the two Shindell et al. studies. For the tropospheric O_3 RF, we find a smaller (but still dominant) contribution from methane emissions for the mean (the Shindell et al 2005 result is within the range found in the ACCMIP analysis), and a larger contribution from NOx emissions. The contributions from CO and NMVOC emissions are less important, and more similar to the two Shindell et al. studies. For CH_4 RF, all the studies find a rather similar contribution from CH_4 emissions, and also from the CO and NMVOC emissions. However, the ACCMIP models have a more strongly negative contribution from NOx emissions, and they show a significant non-linearity in that the net effect of all emissions does not sum to give the same RF as all emissions together.

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		#0 1850s		#1 2000s		#2 1850CH4		#3 1850NOx		#4 1850CO		#5 1850NMVOC	
Model	f	τ	$[CH_4]$	τ	$[CH_4]$	τ	$[CH_4]_{eq}$	τ	$[CH_4]_{eq}$	τ	$[CH_4]_{eq}$	τ	$[CH_4]_{eq}$
B	1.28	8.06	791	8.70	1751	7.31	698	11.60	2531	8.14	1606	8.61	1727
J	1.28	9.02	791	9.29	1751	7.80	657	12.02	2435	8.70	1610	9.29	1752
N	1.35	9.26	791	8.11	1751	6.62	504	12.06	2983	7.49	1572	7.82	1665
O	1.28	8.47	791	8.06	1751	6.76	592	10.83	2561	7.68	1646	7.99	1734
P	1.23	12.29	791	11.61	1751	9.99	612	16.38	2678	10.74	1591	11.09	1655
Q	1.32	8.55	791	8.65	1751	7.13	622	13.15	3045	8.01	1580	8.16	1621

Table 7: Methane adjustment factors (f, dimensionless), lifetimes (yr, for the whole atmosphere), from attribution experiments, and corresponding equilibrium methane concentrations (ppb), calculated using equation (1), for experiments #2-5. For experiments #0-1, we show observed imposed methane values.

	#2. 1850CH_4		#3. 1850NOx		#4. 1850CO		#5. 1850NMVOC	
Model	O_3 RF	CH_4 RF	O_3 RF	CH_4 RF	O_3 RF	CH_4 RF	O_3 RF	CH_4 RF
B	153		193		38		37	
	17		-96		21		3	
	170	480	97	-261	59	55	40	9
J	103		178		29		29	
	16		-58		13		0	
	119	505	120	-231	42	53	29	0
N	168		253		48		15	
	58		-154		28		13	
	226	606	99	-393	76	68	28	32
O	153		205		36		42	
	36		-99		15		2	
	189	546	106	-270	51	39	44	6
P	85		246		35		38	
	18		-62		13		8	
	103	533	184	-305	48	61	46	36
Q	155		252		45		6	
	31		-147		25		19	
	186	526	105	-410	70	65	25	49
Mean BJNOPQ	136		221		39		28	
	30		-102		19		8	
	166	533	119	-312	58	57	35	22

Table 8: Tropospheric ozone and methane radiative forcings (mW m^{-2}) for each model and attribution experiments #2-5 relative to experiment #1 (year 2000s). For methane radiative forcings in experiments #2-5, an equilibrium [CH_4] is calculated based on the diagnosed perturbation to the methane lifetime (Table 7); the RF is then calculated from the difference between the prescribed and equilibrium methane concentrations. For ozone radiative forcings, three numbers are given: the uppermost is the RF from the calculated ozone field (e.g., Figure S7); the middle value is the inferred ozone RF associated with the methane adjustment to equilibrium; the lower number is the net ozone RF.

	Radiative forcing (mW m^{-2}) via:			
Emission	CO_2	CH_4	O_3	CO_2+CH_4+O_3
CH_4	18	533	166	717
NOx		-312	119	-193
CO	87	57	58	202
NMVOC	33	22	35	90
Other factors[c]		127	0	127
Total:	138	427[a]	378[b]	943

Table 9: Emission-based RFs (for 1850s-2000s) (via changes in CO_2, CH_4 and tropospheric ozone) for emitted CH_4, NOx, CO, NMVOC, based on the mean response of the 6 models that conducted the attribution experiments. (cf. IPCC-AR4 Table 2.13).

[a]The total methane RF is constrained to be 427 mW m^{-2} by the observed increase in CH_4 concentrations from the 1850s (791 ppb) to 2000s (1751 ppb), as prescribed in the models. This is then used with the other components to infer the RF due to 'other factors[c]' (127=427-533+312-57-22 mW m^{-2}).

[b]The mean value for these models for the total O_3 RF for 1850s-2000s is 378 mW m^{-2}, which indicates that no other factors are required to explain the O_3 RF.

[c]The 'other factors' not estimated in our attribution experiments include: (i) non-linear interactions between emissions (i.e. simple linear addition of the effects of individual species misses interactions that occur when species change together); and (ii) changes in the value of f between 1850s and 2000s.

Model range of emission contributions to tropospheric O_3 RF			
Emission	ACCMIP	Shindell et al (2005)	Shindell et al (2009)
CH_4	166 ± 46 mW m^{-2} (44 ± 12 %)	200 ± 40 mW m^{-2} (51 ± 10 %)	275 mW m^{-2} (74 %)
NOx	119 ± 33 mW m^{-2} (31 ± 9 %)	60 ± 30 mW m^{-2} (15 ± 8 %)	41 mW m^{-2} (11 %)
CO	58 ± 13 mW m^{-2} (15 ± 3 %)	--	48 mW m^{-2} (13 %)
NMVOC	35 ± 9 mW m^{-2} (9 ± 2 %)	--	7 mW m^{-2} (2 %)
CO+NMVOC	93 ± 10 mW m^{-2} (25 ± 3 %)	130 ± 65 mW m^{-2} (33 ± 17 %)	55 mW m^{-2} (15%)

Table 10: Contributions of emissions of CH_4, NOx, CO and NMVOC to the 1850-2000 O_3 RF, in both absolute terms (mW m^{-2}) and as percentages, for this study, and also from Shindell et al (2005 and 2009). The Shindell et al (2005, 2009) values are all for 1750-2000, and are instantaneous RFs, and were calculated with a different methodology. The ACCMIP values are the means and standard deviations of the six models in Table 8. The Shindell et al (2005) values have estimated errors of ±20% for CH_4 and ±50% for other emissions.

Model range of emission contributions to CH_4 RF (mW m^{-2})			
Emission	ACCMIP	Shindell et al (2005)	Shindell et al (2009)
CH_4	533 ± 39	590 ± 120	530
NOx	-312 ± 67	-170 ± 85	-130
CO	57 ± 9	--	--
NMVOC	22 ± 18	--	--
CO+NMVOC	79 ± 26	80 ± 40	80

Table 11: Contributions of emissions of CH_4, NOx, CO and NMVOC to the 1850-2000 CH_4 RF (mW m^{-2}), for this study, and also from Shindell et al (2005 and 2009). The Shindell et al (2005, 2009) values are all for 1750-2000, and were calculated with a different methodology. The ACCMIP values are the means and standard deviations of the six models in Table 8. The Shindell et al (2005) values have estimated errors of ±20% for CH_4 and ±50% for other emissions.