Geophysical Research Abstracts
Vol. 12, EGU2010-12188-1, 2010
EGU General Assembly 2010

Long term trends and stratospheric profiles of three fluorosulfur gases: SO_2F_2, SF_5CF_3 and SF_6

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Measurements have been made of three industrially-produced fluorosulfur gases in 'old air' collected from deep firn on the Greenland ice cap (collected as part of the North Eemian ice drilling project), from a balloon-borne cryosampler, and from aircraft measurements. The oldest firn air dates back to the middle of the last century, and contained no measurable amounts of the gases, consistent with them having no significant natural sources. Whereas we have previously shown that SF_5CF_3 and SF_6 were growing at the same rate up until at least 1999, it is evident that SF_5CF_3 is now rising at a significantly lower rate than SF_6; calling in to question the earlier assertion that they share a common origin. All three gases show only slight declines in concentration with altitude in the stratosphere, confirming that they have long stratospheric lifetimes. Interhemispheric gradients in the upper troposphere were significant for SF_6 and SO_2F_2, but not SF_5CF_3, consistent with a reduction in the growth rate of the latter.