

We would like to thank J. Laube for this comment that inspired some further thoughts on the missing CH₃Cl source

Increased anthropogenic emissions (Li et al., 2017) may to some extent help to close the CH₃Cl budget. From a purely isotopic perspective, they would fit into the picture and in conjunction with the Central East Asian outflow (Oram et al., 2017), they might appear as tropical emissions. But, this further requires a careful evaluation of increased anthropogenic emissions with respect to the interhemispheric distribution of the CH₃Cl sources. The lack of a substantial interhemispheric gradient in annual mean mixing ratios (Rassmussen, 1980; Simmonds et al. 2004) along with a small interhemispheric gradient in the annual mean OH mixing ratio implies a quite even interhemispheric distribution of the CH₃Cl sources. In line with previous model studies (Yoshida et al, 2006; Xiao et al., 2010 we assumed NH:SH ratio of 1.25 in our model. A substantial increase of the anthropogenic emissions, mainly showing up in the northern hemisphere, would require an adjustment of the interhemispheric ratio of the other sources and or the sinks.

There are other sources and formation processes, being worth to be (re)-evaluated with respect to the missing CH₃Cl emissions.

In the 2014 WMO Ozone Assessment (Carpenter et al. 2014) the CH₃Cl emissions from biomass burning have been revised downward. On the other hand two recent studies (Santee et al., 2013; Umezawa et al., 2014) have shown that enhanced levels of CH₃Cl in the upper troposphere can often be attributed to biomass burning and highlighted the importance of the CH₃Cl biomass burning source. . Hence a careful revision of the biomass burning source may help to reduce the gap between CH₃Cl sinks and sources.

Keppler et al. (2000) reported on the iron catalyzed formation of CH₃Cl from methoxy phenols of soil organic matter. While organic rich peat lands have long been recognized as a source for CH₃Cl (Dimmer et al., 2001), soils in general have been considered as a net sink for CH₃Cl but with highly variable gross emission and deposition fluxes. A recent study has shown remarkable net emissions of $77 \pm 2.8 \mu\text{g m}^{-2}\text{d}^{-1}$ from organic rich temperate forest soils (Redeker et al. 2012). In addition substantial production of CH₃Cl has also been demonstrated for hypersaline soils (Kotte et al. 2012). Manley et al. (2006) reported average fluxes of $3960 \text{ nmol m}^{-2}\text{d}^{-1}$ from bare saltmarsh soils. Thus on a global scale, soils may probably turn from a net sink to a net source.

Moore (2008) proposed a photochemical production pathway for CH₃Cl in saline waters from methoxy phenols. The reaction comprises the photochemical cleavage of the aromatic ring system followed by a nucleophilic substitution with chloride. This mechanism might explain strongly enhanced CH₃Cl concentrations of up to 2 nmol L⁻¹ reported from European estuaries (Christoph et al. 2002). With the vast amounts of DOC exported by the large tropical rivers this source may become significant on a global scale.

Methoxyphenols as well as chloride are widespread in the environment. The photochemical mechanism proposed by Moore (2008) may also be relevant for saline surface soils (see above). Further, phenols and methoxy phenols can contribute to 20–40 % of particulate mass from burning hardwood and softwood (Hawthorne et al., 1989). Given this, the photochemical production pathway proposed by Moore (2008) may also be relevant with respect to biomass burning aerosols. Notably, such a mechanism could also take place for quite some time after the burning event in aged biomass burning plumes and in particular after mixing with sea salt aerosols.

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