



On the detectability of trace chemical species in the martian atmosphere using gas correlation filter radiometry



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ABSTRACT

The martian atmosphere is host to many trace gases including water (H₂O) and its isotopologues, methane (CH₄) and potentially sulphur dioxide (SO₂), nitrous oxide (N₂O) and further organic compounds, which would serve as indirect tracers of geological, chemical and biological processes on Mars. With exception of the recent detection of CH₄ by Curiosity, previous detections of these species have been unsuccessful or considered tentative due to the low concentrations of these species in the atmosphere ($\sim 10^{-9}$ partial pressures), limited spectral resolving power and/or signal-to-noise and the challenge of discriminating between telluric and martian features when observing from the Earth. In this study, we present radiative transfer simulations of an alternative method for detection of trace gas species – the gas correlation radiometry method. Two potential observing scenarios were explored where a gas correlation filter radiometer (GCFR) instrument: (1) performs nadir and/or limb sounding of the martian atmosphere in the thermal infrared (200–2000 cm⁻¹ from an orbiting spacecraft or (2) performs solar occultation measurements in the near-infrared (2000–5000 cm⁻¹) from a lander on the martian surface. In both scenarios, simulations of a narrowband filter radiometer (without gas correlation) were also generated to serve as a comparison. From a spacecraft, we find that a gas correlation filter radiometer, in comparison to a filter radiometer (FR), offers a greater discrimination between temperature and dust, a greater discrimination between H₂O and HDO, and would allow detection of N₂O and CH₃OH at concentrations of ~ 10 ppbv and ~ 2 ppbv, respectively, which are lower than previously-derived upper limits. However, the lowest retrievable concentration of SO₂ (approximately 2 ppbv) is comparable with previous upper limits and CH₄ is only detectable at concentrations of approximately 10 ppbv, which is an order of magnitude higher than the concentration recently measured by Curiosity. From a lander in low dust conditions, both a filter radiometer and gas correlation filter radiometer would provide measurement of H₂O and HDO, which allows the D/H ratio in H₂O to be determined. Detection of N₂O, CH₄, SO₂, C₂H₂, C₂H₆ at concentrations lower than previously-derived upper limits would be possible using a gas correlation filter radiometer in low dust conditions. However, either radiometer would be unable to detect these trace gases in high dust conditions, with the exception of H₂O.

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1. Introduction

The atmosphere of Mars may contain a number of trace gases, which remain undetected due to their low concentrations. Outgassing from the surface, evaporation of ices and perhaps sub-surface microbial metabolism, together with photochemical reactions in the martian atmosphere, are thought to produce a range of organic, nitrogen-based and sulphur-based compounds

(Atreya et al., 2007; Wong et al., 2003, 2004). The detection and observed evolution of these species would therefore provide a strong insight into the atmospheric chemistry of Mars and additionally serve as indirect tracers of geological and perhaps biological activity.

Fluvial or river-like features strongly indicate that liquid water (H₂O) used to flow on the surface of Mars (e.g. Craddock and Maxwell, 1993). However, at present, water only exists in the form of ice at the polar caps (Titus et al., 2003), perhaps as a permafrost below the surface (Boynton et al., 2002; Wade and De Wys, 1968), and with a small amount (on the order of 100 ppmv) as vapour in the atmosphere (Steele et al., 2014; Barker et al., 1970). Water

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vapour in the martian atmosphere exhibits a higher D/H (deuterium-to-hydrogen) ratio than terrestrial water (Webster et al., 2013; Krasnopolsky et al., 1998). This is considered evidence that the dramatic loss of water from past-Mars to present-Mars is a result of ion and neutral escape to space: H₂O is lighter than HDO and thus the former is more vulnerable to escape processes (Greenwood et al., 2008). The D/H ratio in H₂O vapour was demonstrated to vary both with latitude and season on Mars with higher D/H values in the summer hemisphere (Mumma et al., 2003; Novak et al., 2005, 2007), which indicates a fractionation of the two water isotopologues within the martian water cycle (Novak et al., 2011). Fisher et al. (2008) also suggested that the latitudinally and seasonally variable D/H values could be the result of multilatitude subsurface reservoirs, which exhibit different seasonal response timescales to warming and thus differing contributions to water vapour in the atmosphere at any one time. The relative abundances of H₂O and HDO and their spatial and temporal variations are therefore highly insightful into the martian water cycle.

The search for methane (CH₄) is highly topical since the molecule could have a biogenic origin and therefore serve as a tracer of life on Mars. Colonies of methanogenic bacteria (microbes that produce CH₄) could exist in the aqueous subpermafrost environment on Mars though other sources such as volcanism, exogenous introduction (from meteorites, comets) and hydrogeochemical serpentinization are also considered plausible (Atreya et al., 2007). CH₄ was recently (at the time this paper was being written) detected *in situ* in the martian atmosphere at a mean concentration of 0.69 ± 0.25 ppbv by the SAM (Sample Analysis at Mars) Instrument Suite on the Curiosity Rover (Webster et al., 2015). This measured concentration is lower than earlier detections of CH₄, which measured concentrations on the order of 10 ppbv (Villanueva et al., 2013; Formisano et al., 2004; Krasnopolsky et al., 2004) and with a possible latitudinal and seasonal variation up to 50 ppbv (Mumma et al., 2009). Zahnle et al. (2011) suggested these earlier detections of methane should be considered tentative because of (1) the challenge of discriminating between telluric and martian features and (2) concentrations of this magnitude would have implausible consequences on the redox budget of the martian atmosphere if photochemical lifetime of CH₄ are on the order of 100 years (Lefèvre and Forget, 2009; Wong et al., 2003). Thus, there is significant scope to continue to detect CH₄, measure its variation with season and latitude and gain insight into its sources and sinks in the atmosphere.

If methane is indeed present on Mars, some clues of its source could be deduced by searching for other trace gases. For instance, nitrifying bacteria have been shown to produce N₂O in anoxic environments at levels four times higher than oxygenated environments (Goreau et al., 1980). Thus, colonies of subsurface microbes that produce CH₄ might be expected to accompany microbes that produce N₂O. Therefore, a detection of N₂O in the same region as a detection of CH₄ would be suggestive of a biological source of both species. At present, only an upper limit on the concentration of N₂O of 0.087 ppmv has been derived from ground-based spectroscopy (Villanueva et al., 2013). SO₂ would be expected to accompany CH₄ if the latter had a volcanic source. However, SO₂ has yet to be detected in the martian atmosphere, with an upper limit of 2 ppbv derived by Encrenaz et al. (2011). The detection and measurement of other organic compounds such as ethane (C₂H₆), acetylene (C₂H₂), methanol (CH₃OH) and formaldehyde (CH₂O) may also be insightful. The latter three species are largely produced by photodissociation and oxidation of CH₄ (Wong et al., 2004). Thus, the detection of these species would effectively provide a secondary detection of CH₄. In addition, their lifetimes in the martian atmosphere are much shorter (on the order of 100 days) compared to the lifetime of CH₄. Therefore, their detection would imply the source of methane is a current or very

recent source. Ethane can be produced from CH₄ photochemically but may also be produced from thermogenesis of deeply-buried organic material (Allen et al., 2006) or hydrogeochemically through serpentinization of silicates (Geminal and Formisano, 2009). Thus, a detection of C₂H₆ would possibly favour a hydrogeochemical source of CH₄.

Thus, future measurements of the D/H ratio in H₂O, CH₄ together with detections of previously-undetected trace gases such as N₂O and SO₂ would be highly insightful into the geological, chemical, and perhaps biological, processes occurring in the martian atmosphere. Several studies have searched for these species from ground-based instruments with a high resolving power such as CSHELL (Tokunaga et al., 1990) on NASA's IRTF (Infrared Telescope Facility), CRILES (Kaeuff et al., 2004) on the VLT and NIRSPEC on Keck II (McLean et al., 1998). However, only upper limits have been determined so far (Villanueva et al., 2013; Krasnopolsky, 2012; Encrenaz et al., 2011) since discrimination between martian and telluric spectral features, achieving a sufficient signal-to-noise ratio and their likely very low volume mixing ratios ($<10^{-8}$) make their detection a challenge. Observing from a spacecraft in orbit around Mars or from a lander on the surface removes the challenge of observing through the Earth's atmosphere. However, existing spectroscopic instruments on board spacecraft/landers, such as the PFS (Planetary Fourier Spectrometer) on board the Mars Express (Formisano et al., 2005) and the TES (Thermal Emission Spectrometer) on board the Mars Global Surveyor (Christensen et al., 1992) lack the spectral resolving power in order to resolve the small spectral features of trace gases. The ACS (Atmospheric Chemistry Suite, Trokhimovskiy et al., 2014) and NOMAD (Nadir and Occultation for Mars Discovery, Patel et al., 2014) instruments on the payload of the Trace Gas Orbiter in the ExoMars mission, to be launched in 2016, may prove successful. ACS features near-, mid- and thermal infrared spectrometers with an effective spectral range of 1.7–17 μm at a resolving power of $\lambda/\delta\lambda = 4000$ and will perform both nadir and solar occultation observations. NOMAD, similar to the SOIR instrument developed for the Venus Express (Mahieux et al., 2008), will focus on shorter wavelengths (0.2–4.3 μm), but with a similar resolving power. Simulation testing reveals NOMAD could detect methane at concentrations of 10 pptv in solar occultation mode, assuming a signal-to-noise ratio of 4000 can be achieved, and 1 ppbv in nadir viewing, assuming a signal-to-noise ratio of 700 is achievable (Drummond et al., 2011).

In this paper, we present and test an alternative method for detecting and measuring the concentrations of trace gas species in the martian atmosphere – the gas correlation radiometry method. Fig. 1 shows a schematic of a gas correlation filter radiometer. In this method, the planetary atmosphere is viewed through a narrow bandpass filter covering a spectral region containing absorption lines of the candidate gas. A beamsplitter ('BS' in Fig. 1) then divides the beam into two optical paths – the first into an evacuated gas cell and the second into a gas cell filled with the candidate gas. The right-hand schematic in Fig. 1 shows an instrument with several filled gas cells, each containing a different gas. The first optical path is sensitive to the total signal from the source within the spectral range of the narrow band filter. However, the second optical path is much more sensitive to the signal in the continuum since the gas cell strongly attenuates light within the absorption lines of the candidate gas. The signals of both optical paths are then amplified (using pre-amps) and measured by a detector. Hot and cold targets inside the instrument (not shown in Fig. 1) would be used to conduct a two-point radiometric calibration of these two optical paths. The wideband signal (the average signal between the two optical paths, Eq. (1)) and sideband signal (the difference between the two optical paths, Eq. (2)) are then computed.

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