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ARTICLES

Phyllosilicates on Mars and implications for early martian climate

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The recent identification of large deposits of sulphates by remote sensing and *in situ* observations has been considered evidence of the past presence of liquid water on Mars. Here we report the unambiguous detection of diverse phyllosilicates, a family of aqueous alteration products, on the basis of observations by the OMEGA imaging spectrometer on board the Mars Express spacecraft. These minerals are mainly associated with Noachian outcrops, which is consistent with an early active hydrological system, sustaining the long-term contact of igneous minerals with liquid water. We infer that the two main families of hydrated alteration products detected—phyllosilicates and sulphates—result from different formation processes. These occurred during two distinct climatic episodes: an early Noachian Mars, resulting in the formation of hydrated silicates, followed by a more acidic environment, in which sulphates formed.

The presence of hydrated minerals on Mars provides a record of water-related processes. Hydrated sulphates have been observed with the OMEGA (Observatoire pour la Mineralogie, l'Eau, les Glaces et l'Activité) instrument on board the European Space Agency (ESA) Mars Express mission, in numerous light-toned layered deposits in Valles Marineris, Aram Chaos, and Terra Meridiani[1] and in deposits adjacent to the north polar cap[2]. Observations in Terra Meridiani by the Opportunity rover of a variety of sulphates in layered rocks also require an active hydrologic system to account for these deposits[3]. The presence of phyllosilicates on Mars has been previously suggested on the basis of *in situ* elemental analyses by the Viking Landers[4], the identification of smectites in some SNC (Shergottite–Nakhlite–Chassigny) meteorites[5], and remote sensing infrared observations[6–9]. An unambiguous detection of water-bearing phyllosilicates has been reported over large areas[10]. Here we present an overview of the detection of phyllosilicates made by OMEGA, and we discuss their geological context inferred from analyses of imaging data. Phyllosilicates represent a very specific family of highly altered

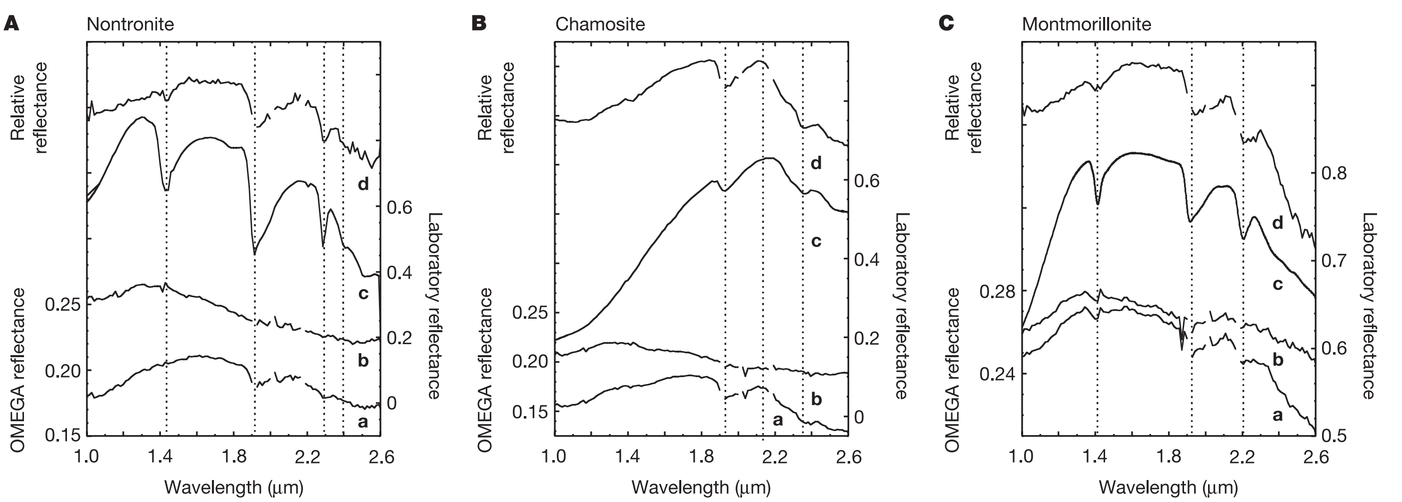

Figure 1 | Phyllosilicate spectra as identified in the OMEGA observations. **A**, Nontronite from the Nili Syrtis Mensae (73.32° E, 29.30° N); **B**, Chamosite from a crater floor in northern Syrtis Major (71.73° E, 17.09° N); **C**, Montmorillonite from light-toned deposits in the vicinity of Mawrth Vallis (20.60° W, 25.53° E). Within each panel, the spectra are labelled as follows: a, I/F atmospherically corrected phyllosilicate spectrum; b, I/F atmospherically corrected reference spectrum; c, laboratory spectra; d, spectral ratio (a divided by b). The OMEGA reflectance scale corresponds to the two lower OMEGA spectra (a and b), the laboratory reflectance to spectrum c, and the relative reflectance to the ratio of OMEGA phyllosilicate/reference spectra (d). The spectra are averages of 5–9 pixels. The reference spectra are selected from regions near to the phyllosilicate spectra, which were acquired during the same OMEGA observation to best-match atmospheric conditions at the time of measurement.

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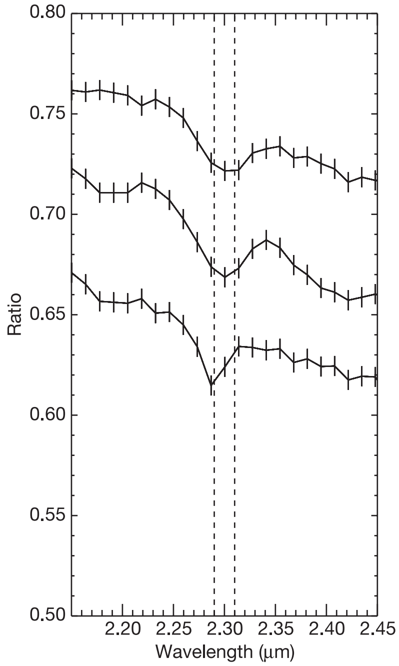

Figure 2 | Variation of the position and shape of the absorption feature in the 2.30-μm region, attributed to varying Fe/Mg abundance. The spectra have been extracted from a terrain in northern Syrtis Major. Three ratioed OMEGA spectra are shown; each spectrum has been divided by a reference spectrum. The ratio process removed instrumental and atmospheric effects. The error bars represent one standard error.

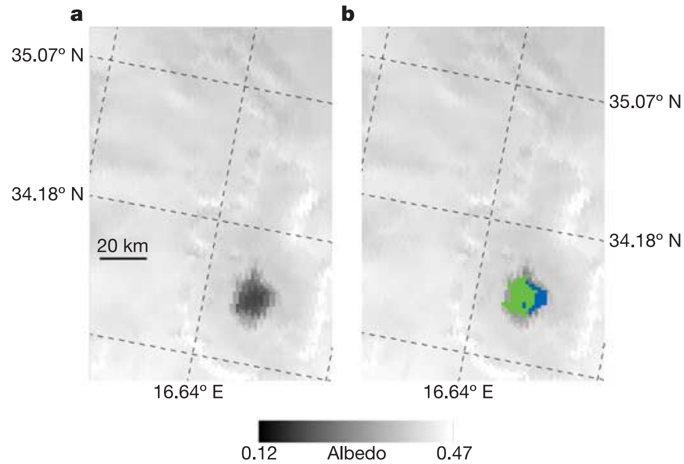

Figure 3 | Dark deposit in a depression in Ismenius Lacus. a, Lambertian albedo map at 1.085 μm; **b**, spatial distributions of pyroxene in green and clays in blue are superimposed for areas exhibiting an absorption feature at 2.15 and 1.93 μm respectively. Fe/Mg clays are present in this dark terrain.

products involving water, so their identification puts constraints on the evolution of Mars. We discuss the implications of our detections to the understanding of the early history of Mars.

Data reduction and mineral identification

The OMEGA instrument acquires three-dimensional (x,y,λ) image cubes, in a spectral domain dominated by solar reflected light (0.3–3.0 μm, with a spectral sampling of ≤14 nm), where most minerals exhibit diagnostic absorption bands. The presence of water induces specific vibrational absorptions for the different classes and subclasses of hydrated minerals; it is therefore possible to discriminate between major minerals resulting from alteration and aqueous processes, such as carbonates, sulphates, phyllosilicates and zeolites[1].

We applied standard processing and reduction procedures to the OMEGA data in the 1.0–2.6 m wavelength range[10,11] to identify absorption features due to water of hydration near ~1.9 μm and metal–OH vibrations in the 2.2–2.4-μm range. The identification of hydrated silicate is first based on the detection of the 1.9-μm absorption band, calculated using spectral channels at 1.93 μm for the band centre and at 1.86 and 2.14 μm for the continuum. These two wavelengths are selected to lie outside the main atmospheric bands. The 1.9-μm band is due to water molecules, either physically or chemically adsorbed. The latter occurs in phyllosilicates, in which water is bound to interlayer cations[12] or cations in tetrahedral sites[13]. Once hydrated regions are identified by this 1.9-μm band, we focus our analysis on the 2.2–2.4-μm interval. Mineralogical assignment is then based on comparison with laboratory spectra. To enhance the spectral signatures of the materials, we perform spectral ratios. Spectra

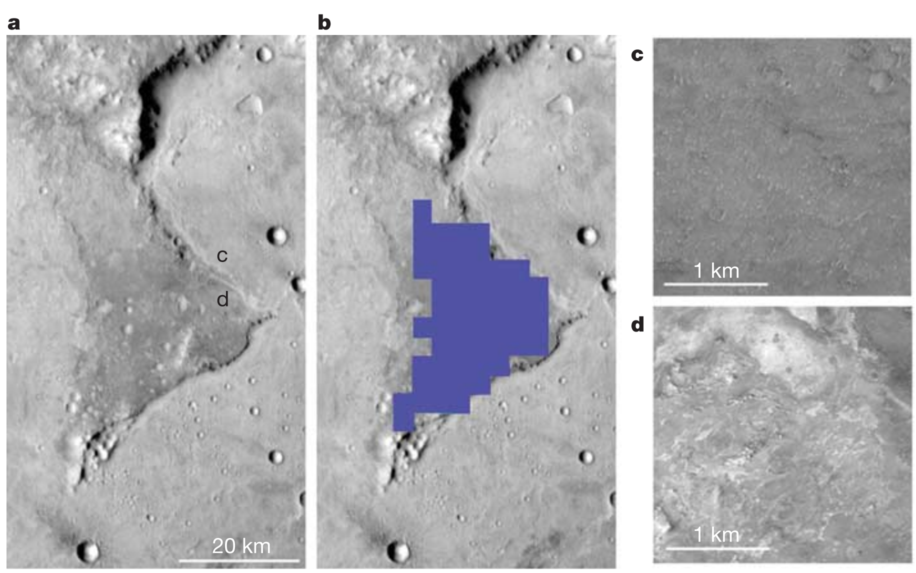

Figure 4 | Detection of Fe-rich clays over Noachian outcrop in Syrtis Major. a, THEMIS daytime infrared I02469002 image. The lavas appear brighter than the peninsula outcrop because of the higher temperature resulting from the dark albedo. **b**, Superimposed on the black and white THEMIS image is the mapping in blue of the Fe-rich clays. The difference in morphology between the early Hesperian pyroxene-rich lava and the unburied Noachian hydrated outcrop can be seen on small portions **c** (dark lava) and **d** (lighter outcrop) of the MOC M0701150 image.

with a 1.9-μm band, exhibiting no features in the 2.2–2.4-μm interval, have been identified, but they are not considered in the present study, because many hydrated minerals exhibit a 1.9-μm band. It is generally considered that absorptions in the 2.2–2.4-μm wavelength region correspond to metal–OH vibrations, where the precise position of this band is a function of the cation species: Al–OH, Fe–OH and Mg–OH vibrational features are centred at 2.2, 2.29 and >2.30 μm respectively[14–16]. Features in the 2.3–2.35-μm region can also be found in laboratory spectra due to an Al–OH vibration, but they are always associated with a 2.2-μm feature and their strength is much weaker than the 2.2-μm absorption[15].

Figure 1 illustrates the diversity of 1–2.6-μm OMEGA spectra, attributed to phyllosilicates on the basis of their narrow absorption features at ~1.9 μm and in the range 2.2–2.4 μm. In Fig. 1A, the spectral features at 1.41, 2.29 and 2.40 μm and the global shape are typical of Fe-rich smectites such as nontronite[17]. In Fig. 1B, the spectral features at 1.41 and 2.35 μm and the global shape are those of chamosite, a (Fe/Mg)-phyllosilicate. In Fig. 1C, a third spectral type is identified, with band centres at 1.41, 2.21 and 2.35 μm. Al-rich phyllosilicates such as montmorillonite provide a very good match. Figure 2 shows expanded OMEGA spectra in the 2.3-μm region, to illustrate the degree of sensitivity observed for the position and shape of this feature. Laboratory spectra of smectites have shown a shift from Fe–OH near 2.29 μm to Mg–OH near 2.31 μm (refs 14, 16). The variety of spectra observed in OMEGA data indicates a range of clay composition, from Fe-rich to Mg-rich smectites. We note that no serpentine clay, usually characterized by a strong 2.33–2.34 μm feature, has been detected so far.

Mapping

Maps of phyllosilicates have been built using spectra in which the 1.9-μm band depth exceeds 2%. The variation of the band depth depends on the degree of hydration, the grain size and the relative abundance of phyllosilicates. We do not discuss the quantitative evaluation of the abundances here, because it requires a complex and nonlinear spectral deconvolution taking into account the potential presence of spectrally neutral species.

After 18 months of operation, OMEGA has covered over 75% of the surface of Mars at a 1.5–4.8 km pixel^{-1} sampling. A major outcome of the present work is that phyllosilicates are detected in only a very restricted number of areas, commonly in association with two types of terrains: dark deposits and eroded outcrops. The key regions of each class are discussed separately below.

The dark deposits are mainly located in and around Arabia Terra, northern Syrtis Major, northern Terra Meridiani, and a few small spots are also found in the Xanthe Terra and Lunae Planum regions. The absorption features at 1.9 μm and 2.30 ± 0.01 μm indicate the presence in these deposits of Fe/Mg-smectites. No montmorillonite-like phyllosilicates have been detected in these dark terrains. A typical example of clay-rich dark soil is shown in Fig. 3. The clays are inside a depression but constrained to a part of the dark deposit only; spectral signatures of pyroxenes dominate the rest of the dark surface. The Mars Orbiter Camera (MOC) narrow-angle images show that the hydrated dark material is probably constituted by a thin surface layer of dark material. This region is representative of most of the observed dark clay-rich occurrences. The low albedo seems to indicate that clays, usually bright in terrestrial analogues, are mixed with an opaque material, which does not present diagnostic spectral features in this wavelength range (a 'spectrally neutral' component). Two scenarios could be proposed to account for the existence of these clay-rich deposits. The first one is the recent surface alteration of mafic material. However, this should have led to a planet-wide distribution of altered surface material, which is not the case. In the other scenario, the alteration could have taken place much earlier. The altered material would then be buried, and eventually exposed by erosion in very specific locations. The dark deposits would then originate from the local erosion of ancient clay-rich sub-surface terrains. In some cases, MOC narrow-angle images show the presence of some layered terrains underlying the dark dust, which would favour this second scenario.

The second major type of clay-rich terrains consists of outcrops. Such terrains were first detected in the Syrtis Major region[10]. More recently, large occurrences were observed in Nili Fossae and Mawrth Vallis. A few spots have been also identified south of the Isidis basin, northern Hellas, and around Terra Meridiani. The composition of these areas is more diverse than that of the dark deposits. An example of clays in Syrtis Major is given on Fig. 4. The presence of Fe-rich clays perfectly matches the contours of a peninsula of ancient basement; the surrounding younger Syrtis pyroxene-bearing lava flows do not contain hydrated silicates. This confirms our initial interpretation[10]

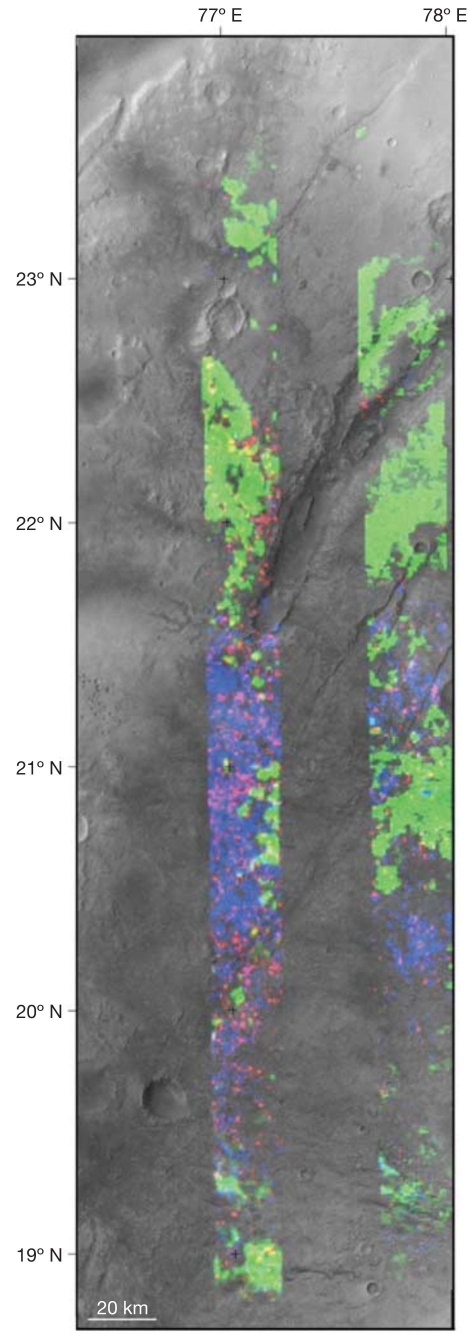

Figure 5 | Spatial distributions of minerals in the Nili Fossae region. Superimposed on a HRSC/Mars Express image, olivine-rich mineral is mapped in green, hydrated minerals are identified by the 1.93-μm band (see text for the definition of the spectral parameter) in purple, and (Fe/Mg)-rich clays are identified by a 2.30-μm feature in red.

that the alteration that produced these clays occurred before the lava outflows, which are dated to the Early Hesperian period[18]. Additional evidence for the presence of clays in the crust of this region comes from the detection of clays in the ejecta of several craters. Although some clays could be formed by the impact, provided water was abundant in the impacted material, it is more likely that most, if not all, of the clays have been excavated from altered crust, as shown by clay-rich outcrops outside the ejecta.

Nili Fossae is an interesting area of Mars in which large abundances of olivine have been detected by the Thermal Emission Spectrometer (TES)[19], the Thermal Emission Imaging System (THEMIS)[20,21] and OMEGA[11]. The identification of olivine in this region as well as in several areas of Mars has strengthened the cold and dry Mars scenario[19,20]. In Nili Fossae, OMEGA has also mapped phyllosilicate minerals (Fig. 5). The areas of high olivine abundance are spatially distinct from those containing high concentrations of hydrated minerals. Nili Fossae cratered terrains are Noachian; they experienced strong erosional processes indicated by dissected terrains with rough chaotic texture, isolated mesas and partially eroded craters. Close examination of the high-resolution High Resolution Stereo Camera (HRSC), MOC and THEMIS imaging data shows that the olivine outcrops rest above the clay-bearing substrate. Thus, the olivine-rich rocks appear to have deposited on top of an older, aqueously altered crust. In addition to the various hypotheses accounting for the origin of olivine-rich terrains[21], they could have originated from the impact that formed the Isidis basin, placing the aqueous-clay-formation episode in the very early Noachian epoch. Alternatively, one may consider that the initial olivine bedrocks were altered to clays, and brought to the surface in a few patches by erosional processes.

OMEGA has discovered a sizeable accumulation of clays in the Mawrth Vallis region between 20° and 28° N and 17° and 22° W (Fig. 6). Phyllosilicates are identified in several light-toned outcrop terrains in the flanks of Mawrth Vallis between −3,200 and −2,700 m as well as on the plateau at an altitude of about −2,300 m. No hydrated mineral has been detected in the valley itself, except for a deposit in a small eroded basin. A remarkable montmorillonite-rich deposit is detected in the more western part (Fig. 6b). THEMIS night-time observations of these terrains are very homogeneous and among the warmest in the region; MOC images indicate geomorphic features typical of eroded terrains. A series of layered deposits occur in these clay-rich areas giving the attributes of material emplaced as sediment[22]. On the west part of the valley (18° W, 23° N), clays are identified in the ejecta of a crater, which reinforces the fact that the clays are in the bulk component of the outcrops and were formed before the occurrence of the water discharge that sculpted Mawrth Vallis. The clay-rich areas are surrounded by low-albedo cratered terrains extending to the south towards Terra Meridiani and showing strong signatures of pyroxene. This region is considered to be ancient, with a late Noachian age[23,24]. Therefore, the presence of clays in eroded outcrops distributed vertically over 500 m provides additional strong evidence for major alteration processes involving liquid water occurring during the early history of Mars. Furthermore, the detection of phyllosilicates in small areas of Arabia Terra and northern Terra Meridiani suggests that the alteration processes could have been intense over this entire region.

Implications for early Mars

From these observations, the presence of clays in outcrops and ejecta strongly supports the following conclusions: (1) the deposits in the crust (Syrtis Major, Nili Fossae) predate the volcanism of Syrtis Major, and possibly the formation of Isidis basin itself; in Mawrth Vallis, clay deposits predate the Noachian/early Hesperian cratering; (2) the clays are a bulk component of the deposits rather than a surface coating or dust layer; (3) the diversity of the composition indicates that the alteration processes affected the variety of igneous rocks (mafic and Al-rich silicates) constituting the martian crust. Note that these conclusions on clay-rich outcrops are consistent with the idea that the dark clay-rich deposits (discussed above) are formed through the erosion of ancient subsurface clay-rich layers.

The formation of clays is controlled by bedrock composition and topography, climate-derived parameters (temperature and long-term availability of liquid water, at or below the surface), availability of water, time and kinetics of mineral reactions[25]. In terms of bedrock origin, Fe-rich smectites such as nontronites are typical of the alteration of mafic material such as gabbros or basalts[26], which are common on Mars. The Al-rich phyllosilicate could either indicate a higher alteration[27] or originate from the alteration of more acidic crustal rocks containing Al-rich minerals such as orthoclase[26].

In terms of surface temperature and long-term availability of water, the formation of clays, and specifically of smectites, requires conditions very different from those currently observed for Mars[28]. For clay formation on the Earth, smectites dominate in the areas of moderate alteration of the temperate regions[29], rocks of tropical zones are mainly weathered to kaolinite and hydroxides, and rocks of arid regions (<200 mm yr^{-1} precipitation) or polar regions alter to poorly

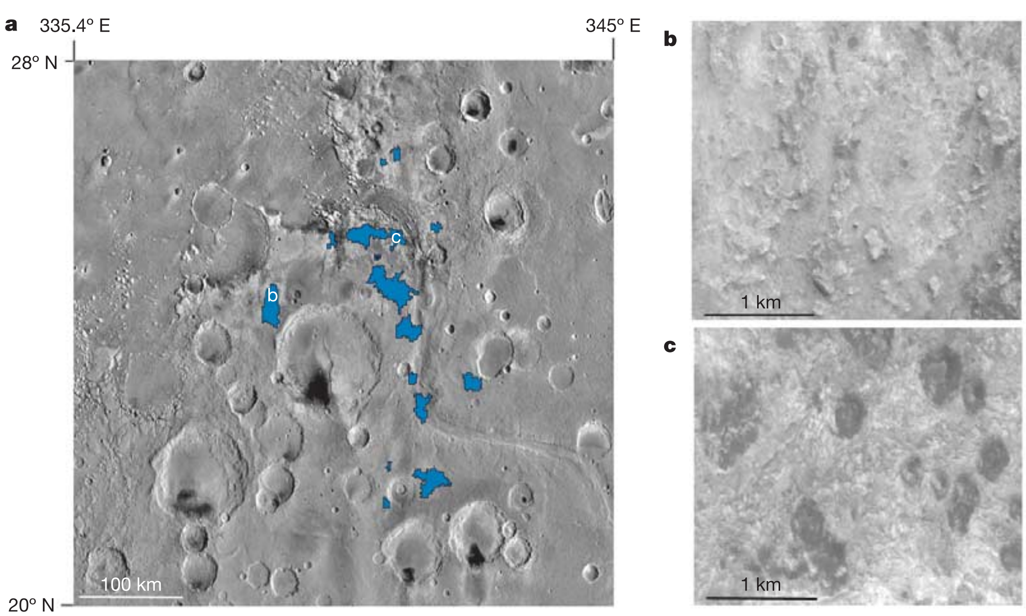

Figure 6 | Identification of clays in Mawrth Vallis. **a**, Map of hydrated minerals in blue over Viking image. Al/Fe/Mg-clays are present. **b**, MOC illustration (E1101550) of the morphology of a montmorillonite-rich outcrop. **c**, MOC illustration (MOC R0801755) of the morphology of a Fe-smectite-rich outcrop. The presence of small mesas indicates strong erosion.

crystalline clays or immature weathering products. In the Dry Valleys of Antarctica, clays are indeed found, as a result of seasonal processes over million of years of activity[30]. However, a similar mechanism on Mars would result in small amounts of clays as thin coatings and does not account for the clay-rich units detected by OMEGA.

The presence of widespread smectites in the martian Noachian rocks suggests an early active hydrologic system that could have sustained a long-term contact of igneous minerals with liquid water, altering such igneous minerals into clays[25]. This process could either have been maintained at the surface, if the climate was warm enough, or have occurred through the actions of fluids in a warm, shallow crust. In both cases, martian clays are likely to record an alteration having taken place over geologic timescales with liquid water present at thermodynamic equilibrium. However, the formation of clays on Mars by impact and volcanic hydrothermal activity has been discussed by several authors[31,32]. Such processes, which do not require liquid water to be stable at the surface, could account for the hydrated silicates identified at least in some specific areas.

Finally, it is important to note that most phyllosilicate deposits are distinct from sulphate deposits as mapped by OMEGA: in general, they do not occur together (in a few cases, such as a few deposits within Aram Chaos, Terra Meridiani and the Becquerel crater, sulphates and clays might be mixed). In contrast to clays, sulphate formation is favoured under acidic water conditions, and does not necessarily imply the long-term presence of liquid water[10]. The two major families of alteration products detected by OMEGA—phyllosilicates and sulphates—could thus trace two different processes separated in time, referring to two major climatic episodes in the history of Mars: an early Noachian Mars, resulting in the formation of hydrated silicates, followed by a more acidic environment in which sulphates formed, rather than clays.

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