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Title	Strong variability of Martian water ice clouds during dust storms revealed from ExoMars Trace Gas Orbiter/NOMAD
Authors	Giuliano Liuzzi; Geronimo L. Villanueva; Matteo M. J. Crismani; Michael D. Smith; Michael J. Mumma; et al.
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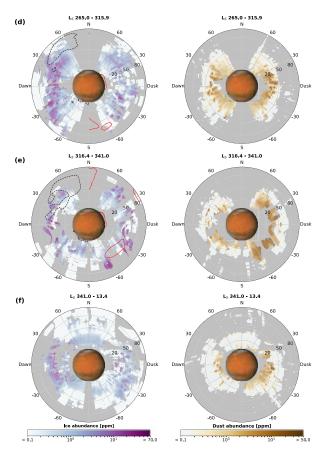


Figure 8. (continued)

atmospheric region, which is likely to occur as long as significant amounts of dust are in the middle atmosphere.

In the context of cloud formation processes, more interesting elements emerge from the comparison between the GDS and the January 2019 dust event. While there are numerous similarities between the spatial distributions of the mesospheric clouds that form during the peak of the two events, the average particle sizes are different (Figure 9). Limiting our comparison to the altitude range in which clouds form in both cases (40 to 80 km), we note that at all latitudes particle sizes are different, with values in the range 0.1-0.7 μm for the 2018 GDS and 0.1-2.0 μm in the January 2019 event. This significant difference is attributable to the differing water vapor abundance in the upper atmosphere during the two events. Indeed, because of the season and lower dust activity of the January 2019 with respect to the 2018 GDS, we effectively observe a lower water vapor abundance in the mesosphere during the January 2019 event, compared to the 2018 GDS, when water was observed at 80 km (Vandaele et al., 2019). The water profiles shown in Figure 9 are retrieved from NOMAD full resolution data, combining the retrievals from different diffraction orders (Aoki et al., 2019), and are the global averages of the profiles during the onset of the two storms. These phenomena are supported by Hartwick et al. (2019) that discuss the relation between the abundance of water vapor in the upper atmosphere and the availability of condensation nuclei. In this case, the 2019 dust event is characterized by less water vapor in the upper atmosphere than the GDS. Moreover, Figure 8 indicates that much less dust is available in the upper atmosphere in the 2019 event than during the GDS. The combination of these two leads to less competition in the condensation process, yielding to formation of larger ice particles (>1.0 µm), as observed during the 2019 dust event.

# 4. Discussion

# 4.1. Spatial and Temporal Distribution of Clouds

The results illustrated herein contribute to the discussion of atmospheric circulation and formation mechanisms for water ice clouds on Mars. Although a detailed comparison between our results and global circulation modeling is beyond the scope of this paper, we will make broad comparisons with the existing literature

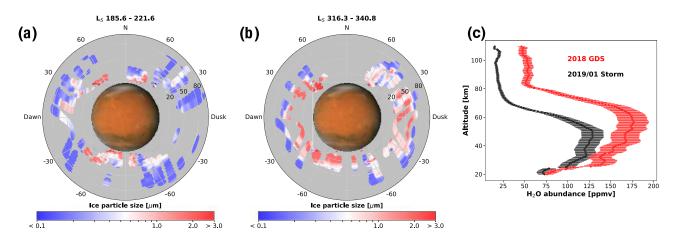


Figure 9. In the same format as Figure 8 we illustrate water ice particle sizes retrieved during the (left) GDS and (middle) January 2019 dust storm. On the right, the global averages of the water vapor vertical profiles retrieved from NOMAD data for the 2018 GDS (red) and the January 2019 storm (black).

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on these topics. The latitudinal and temporal distribution of water ice clouds during the 2018 GDS (Figure 8) suggests a correlation between the enhancement of the water vapor circulation toward southern high latitudes during the GDS (seen in Neary et al., 2019) and the formation of high water ice clouds at high latitudes. Despite this, our results indicate that there are no significant differences in the ways that clouds are distributed at high latitudes in a global dust storm and non-GDS situation. The ideal reference for comparison are MCS retrievals (McCleese et al., 2010) during MY29. In both cases (McCleese et al., 2010, Figures 16 and 17), there is a clear break in the formation of high-altitude clouds in the NH, which occurs between midlatitudes (45 to 60°N; Figures 8b and 8c) and in the polar region. Clouds are seen only below 40 km both in MCS non-GDS retrievals and in this analysis.

Such comparisons show several differences for high-altitude water ice clouds. MCS results in the SH summer report that the top and the bottom levels of most clouds are roughly separated by 1 order of magnitude in pressure, which corresponds to more than two scale heights (25 to 30 km). As seen here though, the altitude distribution is frequently much more complex (Figures 5 and 8c), with clouds at the tropics and middle southern latitudes that extend for more than 40 km. The vertical extension is also subjected to dawn/dusk variations, which in the presence of a GDS are likely to occur, as discussed previously and suggested in other works (e.g., Smith, 2009). This depicts a complex cloud formation mechanism, which in the perihelion season considers both nucleation around dust and vertical and diurnal temperature gradients from the boundary layer through the mesosphere.

Comparison with MCS non-GDS retrievals also confirms that the rapid increase of cloud formation altitudes, seen in both the dust storms observed with NOMAD, is a storm-induced effect. Such a dramatic increase is not seen in MCS retrievals, as there is no tangible sign of a sudden variation of the vertical distribution of the clouds in the different time slots covered by MCS ( $L_S$  180–225–270–315; McCleese et al., 2010, Figures 16 and 17). Low-altitude clouds presented in this work are in agreement with MCS retrievals between  $L_S$  180 and 360. Both retrievals (GDS and non-GDS) cannot identify significant water ice extinction below 25 km around  $L_S$  225, at nighttime for MCS and on the dawn terminator for NOMAD. The same agreement is found between daytime MCS retrievals and NOMAD at dusk, with significant differences only in the middle southern latitude toward  $L_S$  270, where MCS found no significant amounts of water ice below 50 km. Such differences can also be explained by the enhancement of the downwelling branch of the meridional circulation during the GDS. No discrepancies are found between MCS and NOMAD retrievals at the equinox ( $L_S$  0), where clouds form at decreasing altitudes as latitude increases. In this case, both retrievals show that at low latitudes water ice forms as low as 15 km at nighttime (dawn), while during the day the condensation altitude increases because of the solar heating, and clouds form only at 30 km.

### 4.2. Water Ice Particle Size: A Comparison With CRISM Retrievals

A great deal of information about the particle sizes of water ice in the atmosphere of Mars has been obtained by analyzing the limb data of the CRISM spectrometer. The present work increases the data coverage both in time and space with respect to CRISM, providing a more comprehensive assessment and validation of water ice particle sizes and their vertical and temporal variations.

The vertical structure found herein is consistent overall with the results presented in Clancy et al. (2019). Mesospheric water ice clouds exhibit a narrow range of particle sizes (0.1 to  $0.3~\mu m$ ); however, we have a larger number of cases where the retrieved average particle size is  $0.1~\mu m$  (lower boundary imposed in the retrieval), as NOMAD retrievals are indicative of a particle size <0.1  $\mu m$ . In the work of Clancy et al. (2019), there is a distinct decrease in detections for particles smaller than  $0.1~\mu m$ , because smaller aerosols are difficult to discriminate against bigger particles in CRISM data, and can only appear as a continuum scattering component. While our results agree with the general conclusion that particle sizes decrease with increasing altitude, we detect a significant number of water ice clouds in the mesosphere, where the vertical structure of particle sizes is more complex. In such cases, particle sizes exhibit local maxima at the center of the cloud layer, and then decline rapidly toward the cloud top (Figure 9a). While it is difficult to track the spatial distribution of such cases, they appear more frequent in optically thick clouds characterized by the presence of one main layer, which form above 60 km. In any case, the water ice clouds presented in the CRISM study are typically discrete layers, presumably formed in the cold phase of gravity or tidal waves (Clancy et al., 2019), while NOMAD is frequently observing extended cloud hazes during the dust storm, which are situated below the bulk of mesospheric clouds.

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Similar differences in complexity are seen in comparison with previous works (Guzewich et al., 2014; Guzewich & Smith, 2019) regarding water ice clouds in the lower atmosphere. In particular, Figure 6 (top) informs variations of water ice particle size in two different altitude ranges. Our lower atmospheric retrievals (10–40 km) show substantial disagreement with CRISM retrievals. Considering only those cases where retrieved water ice concentrations are significant, the average retrieved particle size (30°S–30°N; 20–40 km) is 2.7  $\mu$ m at perihelion ( $L_S$  240–260) and 2.2  $\mu$ m at the equinoxes ( $L_S$  170–190 and 350–10, MY35), in contrast to values, respectively, of 2.1 and 1.7  $\mu$ m from CRISM. While the perihelion difference can be attributed to the dynamics related to the GDS, the discrepancy observed during the equinoxes is more difficult to explain, although it seems consistent with the sharp vertical gradients we see in particle sizes. Vertical variation of particle size appears to be much steeper than CRISM retrievals (Guzewich et al., 2014). On one hand, limb observations show greater sensitivity to characteristics otherwise difficult to constrain (i.e., particle phase function, shape) so the difference between retrievals might not be really significant. On the other hand, there is the possibility that water saturation is sufficiently high or low that particle growth is more or less efficient than expected. This aspect deserves to be further investigated by dedicated modeling work.

In general, the frequent detection of mesospheric water ice clouds composed of small particles poses some interesting questions related to their impact on the radiative balance in the upper atmosphere, and the lower altitude layers. Previous works (e.g., Madeleine et al., 2011) have already shown the presence of a permanent cold bias around 0.1 mbar, which can be mainly attributed to the exclusion of the radiative effects of high-altitude water ice clouds composed by small particles, especially during the perihelion. In fact, this population of clouds cannot be captured by the unimodal size distribution assumed in dust transport models included in some GCMs. This modeling limitation has been questioned recently in Hartwick et al. (2019), where it is shown how a model including meteoric smoke can account for such observations. The retrievals we have presented here do not provide a final word on nucleation processes in the upper atmosphere, since the thermal information is not fully integrated into this analysis. However, the dawn/dusk NOMAD measurements can certainly inform on those processes in a complementary fashion to the MCS 3 am/3 pm retrievals, which have been used as a benchmark in the work by *Hartwick et al.* In addition, the capability of NOMAD observations to extensively constrain water ice and dust abundance and properties up to 100 km is of great importance in this context. The present work is only the most recent showing the persistent presence of such clouds at many latitudes.

#### 5. Conclusions

NOMAD measurements contain a breadth of information on Martian atmospheric aerosols. By using all the available data taken by NOMAD in Solar Occultation, we have retrieved vertical profiles of water ice, dust, and their particle sizes, with a resolution around 1 km, a maximum vertical sampling of 600 m, from the lower atmosphere to 110 km. To accomplish this, we have developed a robust retrieval methodology to treat NOMAD SO broadband data. Given the number of available points, this is an underconstrained problem, the resolution of which constitutes an important piece of work. We have generally obtained robust results, characterized the errors and the information content, highlighting the consistency of retrieved water ice properties, and the caveats associated to the dust retrievals. In general, these results indicate that when the observed transmittance is above 1%, retrievals are robust enough to separate water ice from dust, and quantify their microphysical properties.

We have analyzed NOMAD data from April 2018 to April 2019, for a total of 1,781 profiles. This period encompasses the 2018 GDS, which was observed to have tremendous effects not only on the vertical distribution of water vapor, as shown in previous studies, but also on water ice cloud formation. The rapid lifting of the water ice condensation altitude is a peculiarity of dust storms, since it has not been observed in a non-GDS situation previously. This effect has been seen to last for a long period (80°  $L_S$ ) after the onset of the storm. Clouds are observed as high as 90 km at the beginning of the GDS, while dust elevates up to 70 km.

Water ice clouds have been observed at dawn and dusk. There are remarkable differences between the two, with optically thicker (larger concentrations) mesospheric clouds at dawn than dusk due to nighttime condensation of water vapor. The combination of dust and ice observations between dawn and dusk reveals how

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dust grains are subjected to nighttime scavenging by water ice. Observations suggest that this process decreases in intensity as the GDS dissipates. Dawn versus dusk analysis also reveals the presence of dusk high-latitude mesospheric clouds in the SH during the most intense phase of the GDS, which is compatible with a strong enhancement of the downwelling branch of the meridional circulation.

We characterized the particle sizes of mesospheric water ice clouds with a precision around 0.1 µm. The majority of water ice cloud particle size vertical profiles exhibit sharp vertical gradients. Specifically, mesospheric water ice particles have sizes between 0.1 and 0.5 µm, which decrease with altitude. However, the comparison with literature shows previously undetected complexities in the vertical profiles of water ice and particle sizes, which constitute exceptions to this general trend. Retrievals have shown significant discrepancies between particle sizes of mesospheric clouds during the GDS and those during the January 2019 dust event. We have attributed this difference to the larger availability of H<sub>2</sub>O and dust in the mesosphere during the GDS than the 2019 event, which results in differing condensation efficiencies.

These elements, together with the observed large vertical and temporal variability of water ice particle size, pose questions about the description of water ice nucleation processes into models. In particular, the accuracy of NOMAD retrievals of water ice in the mesosphere up to 100 km constitute a precious source to validate current working hypotheses on the role of both planetary and interplanetary dust as condensation nuclei at various altitudes. Furthermore, the observations we have presented are important to fill in the existing temporal gaps in the literature, and can serve as a database to be assimilated into global circulation models, going beyond the simple elaboration of climatology for these quantities.

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