Lineshape parameters for water vapor in the 3.2–17.76 μm region for atmospheric applications

Robert R. Gamache*

Department of Environmental, Earth, and Atmospheric Sciences, University of Massachusetts Lowell, 265 Riverside Street, Lowell, MA 01854, USA

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Abstract

Several NASA EOS instruments, the atmospheric infrared sounder (AIRS) on Aqua, and the tropospheric emission spectrometer (TES) and the high-resolution dynamics limb sounder (HIRDLS) on AURA, will be measuring water vapor in the Earth’s atmosphere in the 3.2–17.76 μm spectral region. In order to do retrievals of temperature and concentration profiles, the spectral parameters for many thousands of water vapor transitions must be known. Currently the largest uncertainty in these data is associated with the pressure-broadened half-width. To help ameliorate this situation, complex Robert–Bonamy calculations were made to determine N₂-, O₂-, and air-broadened half-widths and line shifts for 5442 transitions of the principal isotopologue of water vapor for the 11 vibrational bands in this region. The intermolecular potential is a sum of electrostatic terms (dipole–quadrupole and quadrupole–quadrupole), isotropic induction and London dispersion terms, and the atom–atom potential expanded to eighth order. The parameters are adjusted as described in Gamache and Hartmann [J. Quant. Spectrosc. Radiat. Transfer 83 (2004) 119]. Calculations were made at 225 and 296 K in order to determine the temperature dependence of the half-widths. When possible the data are compared with measurements. The average percent difference between the measured and calculated half-widths is 1.97, 2.6, and 1.55 for N₂-, O₂-, and air-broadening of water vapor, respectively. The agreement for the line shifts is less satisfactory. It is clear from this work that the calculations will benefit from a comprehensive adjustment of the intermolecular potential.

Keywords: Half-widths; Line shifts; Water vapor; H₂O; Nitrogen-, oxygen-, air-broadening; Temperature dependence of the half-width

1. Introduction

The scientific evidence that human activities are increasing the rates of global changes is growing [1]. For example, carbon dioxide levels have risen 25% since the industrial revolution and continue to rise. The Earth system is complex and the cause-and-effect relationships among lands, oceans, and atmosphere are not known well enough to predict accurately the impacts these anthropogenic forcings will have on future climate. To help address these questions, measurements must be made over the entire globe for long periods of time. These data can then be used to construct accurate computer models that will enable a forecast of the causes and effects of climate change. The only feasible way to measure such data is from satellites with “remote” sensors. This approach is one of the principal components of NASA’s Earth Observing System. A number of satellites with a variety of instrumentation are either in orbit or will be launched into orbit to collect remote sensed data to use as input to climate models.

The translation of remotely measured data (including ground based, balloon-borne, rocket, and satellite observations) to an understanding of the Earth system is based on the use of physical models. The reduction of these remote sensed data into concentration or temperature profiles, wind velocities, pollutant concentra-

* Fax: +1 978 934 3069.
E-mail address: Robert_Gamache@uml.edu.

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tions, or atmospheric heating rates requires additional modeling. Key to these models is the absorption coefficients of atmospheric gases. The absorption coefficient can be described by the molecular spectroscopic parameters given, for example, in the HITRAN or GEISA databases [2,3], i.e., the vacuum wavenumber of a molecular quantum transition, the corresponding transition intensity, the half-width and line shift of the transition, and the lower state energy. These parameters and their temperature dependence, are at the root of our understanding of the atmosphere. All improvements in these data lead to a better understanding of the Earth system, to better tests of the models, and to developing improved models. Of the parameters needed for inverting remotely sensed data, the collision-broadened half-width is the least well known for atmospheric applications [4]. The effects of uncertainty in half-widths on the accuracy of retrieved parameters are well understood [5,6]. In addition, the importance of the line shift has recently come to light [7,8].

Several NASA EOS instruments will be measuring water vapor in the Earth’s atmosphere. In particular, the atmospheric infrared sounder (AIRS) on Aqua, and the tropospheric emission spectrometer (TES) and the high-resolution dynamics limb sounder (HIRDLS) on AURA will measure H2O in addition to a number of other molecules. AIRS consists of an array grating spectrometer that provides coverage in the infrared and will provide clear-column air temperature profiles and surface temperatures. TES is a high-resolution infrared imaging Fourier transform spectrometer with a spectral coverage from 3.2 to 15.4 μm at a spectral resolution of 0.1 cm⁻¹ in the low-resolution mode and 0.025 cm⁻¹ in the high-resolution mode. HIRDLS is an infrared limb-scanning radiometer designed to sound the upper troposphere, stratosphere, and mesosphere to determine temperature, the concentration of O3, H2O, CH4, N2O, NO2, HNO3, N2O5, CFC11, CFC12, and aerosols. The instrument measures infrared emission in 21 channels ranging from 6.12 to 17.76 μm. (More information on the Aqua and Aura missions can be obtained at http://eosweb.gsfc.nasa.gov/eos_homepage/mission_profiles/index.php).

Water is the principle absorber of longwave radiation in the terrestrial atmosphere, responsible for some 80% of “greenhouse” warming of the Earth’s surface [9]. In this process, water in its vapor phase plays an important and unique role, distinguishable from liquid or ice phases (clouds) in terms of spectral properties, geographic location, etc. A good understanding of the spectroscopy underlying the greenhouse warming, especially pressure broadening of water vapor, is important for two identifiable reasons. First, as peak absorptivity is redistributed to line wings, higher concentrations are required to saturate pressure-broadened lines. This redistribution causes an increase in the radiative forcing, a fact that could have important consequences in radiatively unsaturated conditions such as prevail in the polar winter sky. A second motivation, closely related to the first, derives from the fact that a good understanding of the spectroscopy is essential for a proper interpretation of remote sensing measurements of the atmosphere.

Theory can contribute to the spectroscopic picture in uniquely useful ways. Atmospheric radiative transfer models, which are used both to model natural radiative processes and to interpret remote sensing data, require high-precision parameters describing line positions, intensities, pressure-broadened half-widths and line shifts. Although laboratory measurements can and do supply such parameters, it is difficult to exhaustively cover the vast spectral range and diversity of environmental conditions encountered in the actual atmosphere. Theoretical calculations are in principle an attractive alternative, depending on the accuracy requirements of the radiative transfer application, and of course the credibility of the theory. Even when laboratory measurements are available, however, certain effects (such as line mixing) may still require a sophisticated theoretical model in order to unravel observed spectra.

In this work, calculations of the half-width and line shift for water vapor transitions in the 3.2–17.76 μm region are made for nitrogen and oxygen as perturbing gases at temperatures of 225 and 296 K. The results of these calculations are used to determine the air-broadened half-widths and line shifts for H2O. The temperature dependence of the half-width was determined for the transitions studied in this work.
Within the CRB formalism the half-width, \( \gamma \), and line shift, \( \delta \), of a ro-vibrational transition \( f \rightarrow i \) are given by

\[
\gamma = \frac{n_2}{2\pi c} \left( v \times |1 - e^{-i\delta} S_1(f,i;J_2,v;b) e^{-i\delta} S_1(f,i;J_2,v;b)| \right) e_{ij} f_{ij},
\]

where \( n_2 \) is the number density of perturbers and \( \langle \cdots \rangle_{e_{ij} f_{ij}} \) represents an average over all trajectories (impact parameter \( b \) and initial relative velocity \( v \)) and initial rotational state \( J_2 \) of the collision partner. \( S_1 \) (real) and \( S_2 = R S_1 + i S_1 \) are the first- and second-order terms in the expansion of the scattering matrix; they depend on the ro-vibrational states involved and associated collision-induced jumps from these levels, on the intermolecular potential and characteristics of the collision dynamics. The exact forms of the \( S_2 \) and \( S_1 \) terms are given in [12–14].

The \( S_1 \) term, which makes a purely imaginary contribution, is isotropic in the absence of any vibrational dependence of the anisotropic intermolecular forces. It then has the appellation of the vibrational dephasing term and arises only for transitions where there is a change in the vibrational state. The potential leading to \( S_1 \) is written in terms of the isotropic induction and London dispersion interactions:

\[
\gamma_{\text{induction}} = -\frac{\mu_1^2 \alpha_2}{R^6}, \quad \gamma_{\text{dispersion}} = \frac{3}{2} \frac{I_1 I_2}{I_1 + I_2} \frac{Z_1 Z_2}{R^6},
\]

where \( \mu_1 \) is the dipole moment of water vapor and \( \alpha_2 \) and \( I_k \) are the polarizability and ionization potential for water vapor (\( k = 1 \)) and collision partner (\( k = 2 \)).

For the ground state the Watson constants of Matsushima et al. [33] were used. For the \( v_1, v_2, v_3 \), and \( v_2 \) bands, Watson constants are from Fland and Camy-Peyret [34], and for the \( v_3 \) band the Watson constants from [35] are used so that the wavefunctions would have proper symmetry properties. The rotational constants for \( N_2 \) and \( O_2 \) are 2.0069 and 1.4377 cm\(^{-1} \), respectively [36].

Many of the molecular parameters for the \( H_2O-N_2 \) or \( H_2O-O_2 \) systems are well known and the present calculations use the best available values from the literature. The dipole and quadrupole moments of water vapor are taken from [24,37], respectively. The quadrupole moment of nitrogen is from Mulder et al. [38], that for oxygen is from Stogryn and Stogryn [39]. The numerical values are listed in Table 1. The ionization potential of water is taken to be a vibrationally independent value of 12.6 eV [40]. For nitrogen the polarizability, 17.4 \( \times 10^{-25} \) cm\(^3 \), is taken from [41] and the ionization potential, 15.576 eV, from [42]. For oxygen the polarizability, 15.80 \( \times 10^{-25} \) cm\(^3 \), is taken from [41] and the ionization potential, 12.063(\( \pm 0.001 \)) eV, from [40]. In the parabolic approximation, the isotropic
part of the interaction potential is taken into account in
determining distance, effective velocity, and force at
closest approach [11]. To simplify the trajectory calcula-
tions, the isotropic part of the atom–atom expansion is
fit to an isotropic Lennard-Jones 6-12 potential.

Generally the atom–atom parameters are determined
by taking the homonuclear atom–atom parameters, for
example, see Bouanich [43], and using combination rules
[28] to produce the heteronuclear atom–atom parameters.
Calculations made using these parameters for some
transitions in the \( v_2 \) band showed some structured dis-
crepancies when compared with measurements of both
the shifts and widths. In a recent study of the lesser iso-
topologues of water vapor [44] it was observed that a
relatively small change in the atom–atom parameters
for the N–D and O–D interactions resulted in much im-
proved agreement with measurement. The atom–atom
parameters for the interactions N–D and O–D were ob-
tained using \( D_2 \) atom–atom parameters determined via
ab initio calculation [45] and combination rules. Fur-
thermore, there are a number of different methods which
have been proposed to determine heteronuclear poten-
tial parameters from homonuclear parameters [46 and
references therein], 47]. Good and Hope [29] showed
that different combination rules used to determine \( \epsilon \)
cause variations of up to 15% in the final values. Thus
the resulting parameters have an increased uncertainty
(besides that resulting from the imprecision of the
homonuclear data) which depends on the method chosen
to go from the homonuclear to the heteronuclear
parameters. From these facts, we conclude that adjust-
ment of the atom–atom parameters within \( \pm 15\% \)
around the values given by the combination formula
of Hirschfelder et al. is not unreasonable provided there
are reliable experimental data on collisional parameters
for adjustment.

In order to improve the quality of calculations for
nitrogen broadening of \( \text{H}_2^{16}\text{O} \), six transitions of the \( v_2 \)
band were chosen for which there are multiple measure-
ments and for which the calculated widths using unad-
justed atom–atom parameters were too high for two
trees, too low for two lines, and in agreement for the last
two transitions. The potential parameters \( \sigma_{\text{HN}}, \sigma_{\text{HN}}, \)
\( \epsilon_{\text{ON}}, \) and \( \sigma_{\text{ON}} \) were adjusted to give a good fit of all these
lines. With respect to the starting values (from combina-
tion rules of [28] and the homonuclear data by Bouanich
[43]), the final parameters correspond to a 10% lowering
of \( \epsilon_{\text{ON}} \), a 10% increase in \( \epsilon_{\text{HN}} \), a 3.9% decrease in \( \sigma_{\text{HN}} \),
and no change in \( \sigma_{\text{ON}} \). These potential parameters were
then further refined for transitions in the \( v_2 \) band. The
final values are presented in Table 1. Note, a full least-
squares minimization was not thought worthwhile until
a detailed study of velocity averaging effects (see below)
are completed. The final adjustment of the potential
parameters was made by comparing calculated line
shifts to measurement. For transitions involving changes
in a number of quanta in \( v_1 \) and \( v_3 \) the calculations made
using the coefficients in Eq. (4) give sufficiently good
agreement with the measurements [13]. Comparing cal-
culations for the \( v_2 \) band with the measurements of Toth
[48] indicated that the ab initio value of 0.03 in Eq. (4)
should be changed to 0.07 [49]. Currently, there are
collisions and for half-widths of \( \text{H}_2\text{O} \) at \( \approx 300 \) K the mrvt
analysis and the velocity averaged calculation gave re-
results within a few percent of one another [50].

3. Calculations

In the spectral region covered by the instruments,
3.2–17.76 \( \mu \)m (3124.2–563.2 cm\(^{-1}\)), the 2001 HITRAN
database [2] contains 13622 water vapor transitions
for all isotopologues of water. Here we focus on the
range 563–3125 cm\(^{-1}\) for the principal isotopologue of
water vapor and transitions with \( J \leq 18 \) (5442 transi-
tions). Table 2 lists the vibrational bands of \( \text{H}_2^{16}\text{O} \)
in the wavenumber region and the number of lines consid-
ered in this work. These data were taken and sorted into
files according to the 11 vibrational bands shown in Ta-
ble 2. From these files the rotational quantum numbers
were extracted to create lists to use in the CRB
calculations.

For each vibrational band calculations were made at
225 and 296 K for the \( \text{H}_2\text{O}–\text{N}_2 \) and \( \text{H}_2\text{O}–\text{O}_2 \) systems.
The half-width and line shift for air as the perturbing
gas were obtained assuming binary collisions and Dal- 
aton’s law
\[ \gamma_{\text{air}} = 0.79 \gamma_{\text{N}_2} + 0.21 \gamma_{\text{O}_2} \]
and
\[ \delta_{\text{air}} = 0.79 \delta_{\text{N}_2} + 0.21 \delta_{\text{O}_2}. \]  
Eq. (6) applied to determine the air-broadened half-widths.

This was done at both temperatures of the study.

From the calculations made at the two temperatures,
the temperature dependence of the half-widths can be

determined assuming the usual power law form
\[ \gamma(T) = \gamma(T_0) \left( \frac{T}{T_0} \right)^n, \]  
where the reference temperature \( T_0 \) is usually taken as
296 K but is not restricted to that value.

The data files for each band for nitrogen or oxygen as
the perturbing gas were taken at each temperature and
Eq. (6) applied to determine the air-broadened half-width
and line shift at 225 and 296 K. These data were then used
to determine the temperature dependence of the air-
broadened half-widths. The results of these calculations
are available as Supplementary Material to this article

4. Discussion

The CRB half-width data were compared to the
water vapor databases of lineshape parameters [51].

Comparisons were made between measurement and cal-
culation for cases where multiple measurements and sin-
gle measurements exist for the same ro-vibrational
transition. The results of the comparisons are given in
Tables 3–5 for each vibrational band studied and for
\( \text{N}_2\), \( \text{O}_2\), and air broadening of water vapor. The tables
are divided into columns where the CRB values are
compared with a single measurement (\( N_{\text{Exp}} = 1 \)) and
with multiple measurements of the same transition
(\( N_{\text{Exp}} > 1 \)). Note that for many of the bands there are
no measured data. In general the agreement between
measurement and calculation is quite good especially
considering the results of an intercomparison of meas-
ured half-widths published recently [51]. The average
percent difference between the measured and calculated
half-widths is \( -1.97, 2.6, \) and \( -1.55 \) for \( \text{N}_2\), \( \text{O}_2\), and
air-broadening of water vapor, respectively. While these
numbers are good it should be realized that when there
are a large number of comparisons the standard devia-
tions are in general between 10 and 20%.

In addition, plots were made to compare the CRB cal-
culated value of the half-width with measurements for
each ro-vibrational transition where two or more mea-
surements were made. A note here to explain some fea-
tures of the databases. Often there are pairs of
measured data for a particular ro-vibrational transition;
however, one datum will be for the principal isotopo-
logue and the other datum for one of the other isotopo-
logues. Thus, there are a number of plots with only one
measured point and the CRB point. Fig. 1 shows the
air-broadened half-width data for the \( 7_3 \rightarrow 6_2 \) transi-
tion of the \( v_2 \) band, for which there are seven mea-
surements. The CRB calculated value is 2.45% from
the average of the measurements. Note: the number above
each measurement refers to a reference number in the
bibliography of the measurement database [51], the solid
line is the average of the measurements and the dashed
lines are 1 and 2 standard deviations of the comparison
of the measurements. Fig. 2 shows the air-broadened
half-width data for a high \( J \) line \( (13_1 \rightarrow 12_2,_{11}) \) of
the rotation band. Again there are seven measurements,
however, the CRB calculated value is \( \sim 20\% \) from the
average of the measurements. Note that the half-width

### Table 2

Number of \( H_2^{16}O \) transitions in the 3.2–17.76 \( \mu \)m region as a function of vibrational band

<table>
<thead>
<tr>
<th>( v_0' )</th>
<th>( v_1' )</th>
<th>( v_2' )</th>
<th>Number of lines</th>
</tr>
</thead>
<tbody>
<tr>
<td>000</td>
<td>000</td>
<td>000</td>
<td>506</td>
</tr>
<tr>
<td>010</td>
<td>010</td>
<td>010</td>
<td>144</td>
</tr>
<tr>
<td>010</td>
<td>000</td>
<td>000</td>
<td>1885</td>
</tr>
<tr>
<td>020</td>
<td>010</td>
<td>010</td>
<td>872</td>
</tr>
<tr>
<td>100</td>
<td>010</td>
<td>010</td>
<td>532</td>
</tr>
<tr>
<td>030</td>
<td>020</td>
<td>020</td>
<td>382</td>
</tr>
<tr>
<td>001</td>
<td>010</td>
<td>010</td>
<td>484</td>
</tr>
<tr>
<td>020</td>
<td>000</td>
<td>000</td>
<td>326</td>
</tr>
<tr>
<td>030</td>
<td>010</td>
<td>010</td>
<td>146</td>
</tr>
<tr>
<td>100</td>
<td>000</td>
<td>000</td>
<td>90</td>
</tr>
<tr>
<td>001</td>
<td>000</td>
<td>000</td>
<td>75</td>
</tr>
</tbody>
</table>

### Table 3

Comparison of the CRB calculations with measurement for \( \text{N}_2 \)-broadened half-widths of water vapor transitions in the 3.2–17.76 \( \mu \)m region at 296 K

<table>
<thead>
<tr>
<th>( v_0' )</th>
<th>( v_1' )</th>
<th>( v_2' )</th>
<th>( H_2O-N_2 )</th>
<th>( N_{\text{Exp}} = 1 )</th>
<th>( N_{\text{Exp}} &gt; 1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of lines</td>
<td>% error</td>
<td>Number of lines</td>
<td>% error</td>
<td></td>
<td></td>
</tr>
<tr>
<td>000</td>
<td>000</td>
<td>160</td>
<td>7.04</td>
<td>10</td>
<td>5.97</td>
</tr>
<tr>
<td>010</td>
<td>010</td>
<td>2</td>
<td>17.93</td>
<td>371</td>
<td>-5.31</td>
</tr>
<tr>
<td>010</td>
<td>010</td>
<td>570</td>
<td>-1.76</td>
<td>6</td>
<td>-15.82</td>
</tr>
<tr>
<td>020</td>
<td>010</td>
<td>90</td>
<td>-5.24</td>
<td></td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>010</td>
<td>15</td>
<td>-8.15</td>
<td></td>
<td></td>
</tr>
<tr>
<td>001</td>
<td>010</td>
<td>27</td>
<td>-0.70</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
is roughly a factor of 4 smaller than that of Fig. 1 so percentage difference does not tell the entire story. Fig. 3 shows air-broadened half-width data for a high J line (17\,2\,15 \leftarrow 16\,11\,6) of the v2 band where there is excellent agreement with measurement. Fig. 4 demonstrates a situation where scatter in the measurements can lead to what appears to be poor agreement between the CRB value and the average measurement. Here what is reported as approximately an 11% difference between measurement and the calculation is in reality good agreement.

Eliminating the stray point gives \(\frac{1}{C_0} \times 0.42\%\) difference.

Plots for all transitions of each perturbing gas and the measurement database with bibliography are available at [http://faculty.uml.edu/Robert_Gamache](http://faculty.uml.edu/Robert_Gamache).

Comparisons between measurement and CRB calculations for line shifts are fewer due to the number of
438 measurements reporting line shifts. Table 6 gives the
comparisons possible for the bands and wavenumber re-
439 gion of this study. Reported in the table are the number
440 of comparisons and the average absolute difference
441 (AAD) in cm$^{-1}$ between measurement and theory
442 (Exp – CRB). This is thought to be a better way to com-
443 pare the data since percent difference for comparison of
444 line shifts is generally large. In fact there are transitions
445 where the uncertainty of the line shift is more than 100%
446 of the shift value. Hence the absolute difference between
447 the measured and calculated line shifts was considered.
448 Of the 5442 transitions in this region shifts have been
449 studied experimentally for 2894 transitions: 1100, 342,
450 and 1452 for N$_2$-, O$_2$-, and air broadening, respectively.
451 Seven hundred and sixty-five transitions have been mea-
452 sured by more than one group, 132 and 633 for N$_2$- and
453 air-broadening, respectively. Of these transitions five
454 air-broadened transitions have been measured by three
455 groups; all other multiple measurements are pairs of
456 data.

![Fig. 2. Measured air-broadened half-widths (triangle symbol with error bars) and CRB value (asterisk) for the 13$_112$ $\leftrightarrow$ 12$_{211}$ transition of the $v_2$ band of water vapor.](image1)

![Fig. 3. Measured air-broadened half-widths (triangle symbol with error bars) and CRB value (asterisk) for the 17$_{215}$ $\leftrightarrow$ 16$_{116}$ transition of the rotation band water vapor.](image2)
For nitrogen and air broadening of H$_2$O$_{16}$O the multiplicity measurements are for transitions in the $v_2$ band.

Comparing the multiple measurement comparisons with the single measurement comparisons shows that the average absolute difference between measurement and calculation is lower for comparison with multiple points.

For the pairs of data the AAD between the measurements and the AAD between the CRB value and the average of the measurements can be compared. For nitrogen broadening there are 132 pair comparisons. The AAD between the CRB value and the average of the measured values is 0.00411 cm$^{-1}$ compared with the AAD between the pairs of measurements of 0.00050 cm$^{-1}$. For air broadening there are 628 pairs of measurements with an average absolute difference of 0.00046 cm$^{-1}$ compared with the AAD between measurement and theory of 0.00186 cm$^{-1}$ (note this contains the five transitions for which there are three measurements each). Thus in both cases the agreement between measurement is better than the agreement between theory and measurement.

From the results in Table 6, it is obvious that the overall agreement is not as good as that described in [49]. One reason is that the adjustment of the coefficient of the vibrational dependence of the polarizability for the $v_2$ bending mode made in [49] was done for the doublet transitions studied in that work. Here there are very few doublet transitions, hence the adjustment of the vibrational dephasing terms is not optimum for these transitions. This clearly indicates that the adjustment of the intermolecular potential must be revisited. This is underway and the results will be presented in a future publication.

5. Conclusion

Calculations of the pressure-broadened half-width and pressure-induced line shift have been made for nitrogen-, oxygen-, and air-broadening of H$_2$O$_{16}$O transitions for the 11 vibrational bands in the 563–3125 cm$^{-1}$ region of the spectrum. The calculations were made at...
two temperatures, 225 and 296 K, and for rotational states with $J \leq 18$. In total 5442 transitions were studied.

From the calculations the temperature dependence of the nitrogen-, oxygen-, and air-broadened half-widths was determined. The CRB results for the half-widths compared very well with measurement. The comparisons for the line shifts were not as good in part due to the use of parameters from [49]. The final air-broadened data file was given to Dr. Larry Rothman to be merged with the measured half-widths for addition to the HI-TRAN database.

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517 Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jms.2004.08.004.

521 References


