National Institute of Standards and Technology transportable tunable ultraviolet laser irradiance facility for water pathogen inactivation

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ABSTRACT

A method of ultraviolet germicidal irradiation (UVGI) for water pathogen inactivation effectiveness using tunable, narrowband laser light is described. A transportable tunable UV (TTUV) laser system for providing a known irradiance (μW/cm^2) or dose (mJ/cm^2) suitable for irradiating water samples in Petri dishes over the wavelength range of 210 nm–300 nm was developed by the National Institute of Standards and Technology. The TTUV facility, consisting of a 1 kHz pulsed UV laser and light-tight enclosure containing the optics necessary to uniformly irradiate a water sample, was used in a microbiology laboratory to dose drinking water pathogens and surrogates as part of a Water Research Foundation study in the summer and fall of 2012. The approach demonstrated improved accuracy and simplified spectral analysis over conventional pathogen inactivation sources consisting of broadband UV sources and bandpass filters. In this work, the TTUV facility design and key components are described, including modifications in the field to provide the required irradiance levels. The irradiance and dose levels produced by the tunable UV laser during the project are also presented. The transportability of the TTUV system enabled it to be brought to a microbiology facility allowing the water samples (microbial suspensions) to be irradiated in a location with experienced staff and facilities for preparing, handling, analyzing, storing, and shipping the many samples studied. These results, published elsewhere, established that the tunable UV laser system provides unique UVGI capabilities for use with water pathogens and has applications for other pathogen experiments, for example, air-purification studies.

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I. INTRODUCTION

Ultraviolet (UV) radiation effectively inactivates common pathogens found in ground and surface waters, such as *Cryptosporidium*, *Giardia*, and most bacterial pathogens (e.g., *E. coli*).[1–6] Water treatment facilities are now using UV radiation for disinfection of drinking water, supplementing standard chemical treatment. These systems typically use either low-pressure (LP) or medium-pressure (MP) mercury vapor lamps. In the 200 nm–300 nm region, LP lamps emit primarily at 253.7 nm, and MP lamps emit across this spectral region. Research has been reported showing polychromatic light from MP lamps is more effective than that from LP lamps for inactivating certain pathogens, and the wavelengths below 240 nm are responsible for this enhancement.[3,7,8] At the time of this work in 2012, action spectral data for waterborne pathogens and surrogate

micro-organisms often did not exist below 240 nm and particularly not below 220 nm.

In 2011, the National Institute of Standards and Technology (NIST) was asked if they could build on their efforts in UV sensor calibration[9,10] and tunable laser systems[11–14] to develop a system to irradiate water samples as part of a Water Research Foundation Project.[15] The transportable tunable UV (TTUV) facility was developed to provide a known, spatially uniform irradiance (μW/cm^2) or dose (mJ/cm^2) to water samples (microbial suspensions) in Petri dishes.[16] The UV source covered the wavelength range of 210 nm–300 nm and irradiated a 5 cm diameter area with a uniformity of 10% or better. By tuning the laser wavelength and controlling the dose exposure, an "action spectrum" or germicidal effectiveness curve is developed to quantify the efficiency of micro-organism inactivation. The action spectrum is defined as "a plot of a relative biological

or chemical photoresponse (= Δy) per number of incident (prior to absorption) photons, vs wavelength."[17] Finally, the system was to be installed at a microbiology facility in St. Albans, VT. This permitted the staff and facility experienced with irradiating and analyzing water samples with broadband UV sources and bandpass filters to work with the TTUV facility.

II. TTUV LASER IRRADIANCE FACILITY

The TTUV laser irradiance facility is composed of four major parts: the laser, the water sample irradiance system, the sample irradiance test position, and a NIST calibrated UV irradiance transfer standard detector ("UV detector"). Each part will be described in more detail below along with the system's performance in the "field."
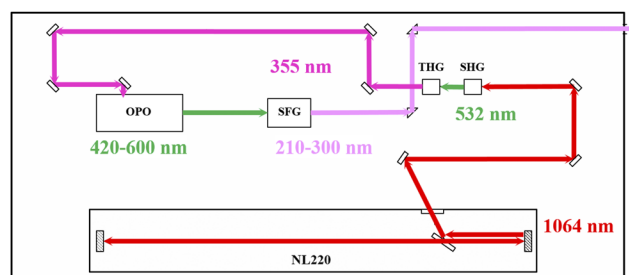
A. Laser

A tunable laser provides advantages over the traditional approaches using a broadband UV source with bandpass filters or coupled to a monochromator. The most apparent advantage of the laser is that high levels of quasi-monochromatic radiant flux are provided at a user-selectable wavelength, allowing any wavelength to be selected and not relying on the wavelength availability of bandpass filters. The tunable laser also has the advantage of higher flux levels than a monochromator system due to the trade-off between the monochromator bandpass and output flux. That is, there is a practical limit to the amount of radiant flux with a desired bandpass (1 nm). The use of quasi-monochromatic sources also reduces the complexity in the determination of pathogen disinfection action spectra because the irradiance no longer requires weighting the spectral source distribution with a bandpass function. In practice, a tunable UV laser is more complex than traditional UV sources and requires certain design considerations to provide the desired spectral purity and spatial uniformity.

The fundamental component of the NIST TTUV laser system is an Ekspla[18] NT242-SH/SFG 1 kHz pulsed laser,[29] tunable over the spectral range from 210 nm to 2600 nm. The manufacturer specifies the pulse width to be <7 ns with 5% pulse-to-pulse power stability. A simplified optical layout of the Ekspla NT242 laser is shown in Fig. 1. The optical parametric oscillator (OPO) is pumped by a solid state, diode-pumped laser at 1064 nm that has been doubled and tripled by the second and third harmonic generators (SHG and THG), to produce 355 nm radiation. The OPO output is tunable from 400 nm to 2600 nm and results in a signal and idler beam, with the signal covering the visible spectral region. The signal is doubled inside the sum frequency generator (SFG) to produce tunable flux over the region of study from 210 nm to 290 nm, but the output contains a small amount of the signal beam. The laser bandwidth varied from 0.04 nm at 300 nm to 0.07 nm at 210 nm. The average laser power ranged from 17 mW to 29 mW over the wavelengths 210 nm–300 nm as measured by a Coherent FieldMate laser power meter with an expanded uncertainty ($k = 2$) of 4%.

B. Sample irradiance system

In the initial TTUV system configuration deployed to the field, the sample irradiance system was configured with a computer-controlled shutter, prism, aluminum-coated mirror, lens, fiber optic,


FIG. 1. Simplified schematic diagram showing the optical layout of the Ekspla NT242 tunable laser (1 kHz pulse rate, <7 ns pulse width). See the manufacturer's manual[19] for detailed information about the optical components.

engineered diffuser, and second lens. The prism removed the signal beam, but the angle of the mirror had to be adjusted at each wavelength, so that the UV flux was coupled into the fiber. This design was not practical in the field because of the time required to manually change the aluminum mirror angle with wavelength and increasing losses in the fiber optic cable due to "solarization" (UV damage).

The configuration was modified to simplify the optics alignment and reduce the number of optical components contributing to the power losses in the UV. The final layout of the TTUV facility is shown in Fig. 2. Once the laser beam leaves the Ekspla laser, it enters a light-tight enclosure through a computer-controlled shutter that determines the dose to the water samples. Spectral filtering of the laser beam was required because of the visible light co-aligned with the laser beam but at double the selected UV laser wavelength. The spectral filtering was accomplished with dielectric mirrors or a prism and slit depending on the selected wavelength. For most of the wavelengths shorter than 300 nm, the beam is reflected off two dielectric mirrors, which filter out visible light.

The dielectric mirrors, labeled M1 and M2 in Fig. 2, were highly reflective from 240 nm to 300 nm. A second set of dielectric mirrors were procured from Alpine Research Optics (ARO)[20] partway through the project that worked well for 220 nm and 230 nm, but the prism and slit were still required at 210 nm due to the low reflectance
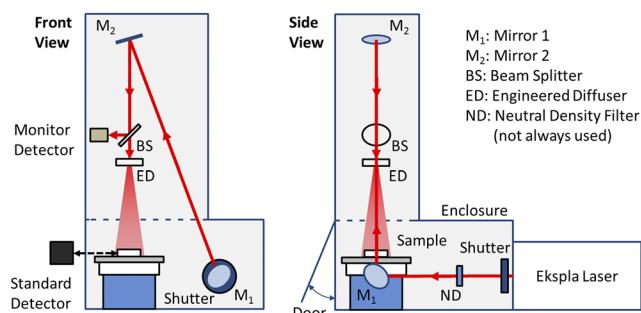
FIG. 2. The NIST tunable UV irradiance laser system 240 nm–290 nm optical configuration. The UV detector is substituted for the water sample to measure the irradiance at each wavelength of interest.
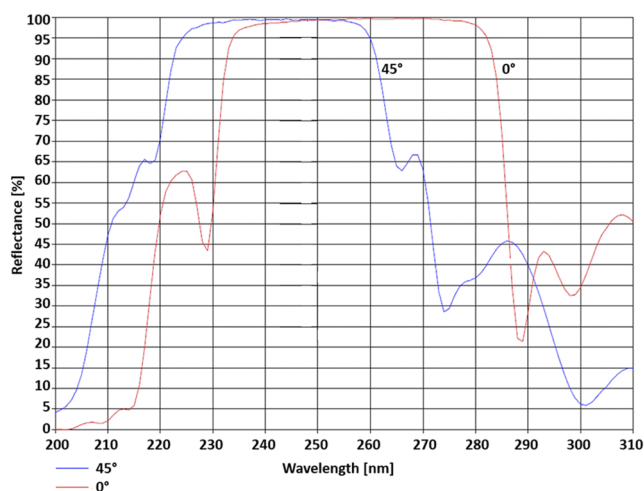
FIG. 3. The reflectance of ARO model MR1520 dielectric mirrors at both 0° and 45° incidence angles with random polarization.

from the dielectric mirrors. The reflectance of these ARO model MR1520 dielectric mirrors is shown in Fig. 3. Neutral density filters can be added to the optical path between the shutter and the first mirror, M1, to reduce the irradiance level at the water sample.

Due to low reflectance, the dielectric mirrors were replaced by aluminum front surface reflecting mirrors, a fused silica prism, and slit to filter out the visible light for 210 nm–230 nm. This configuration is shown in Fig. 4.

The beam then travels through a beam splitter and an engineered etched fused silica diffuser.[21] The beam splitter sends a small portion of the UV light to a silicon photodiode, which monitored the irradiance level during sample exposure. The diffuser was a critical component of the TTUV laser system. Unlike typical optical diffusers, this diffuser is specifically engineered to modify the laser beam from a collimated oval shape (1.5 mm by 10 mm) to a uniform diverging beam (10° half-angle) to irradiate the water samples.
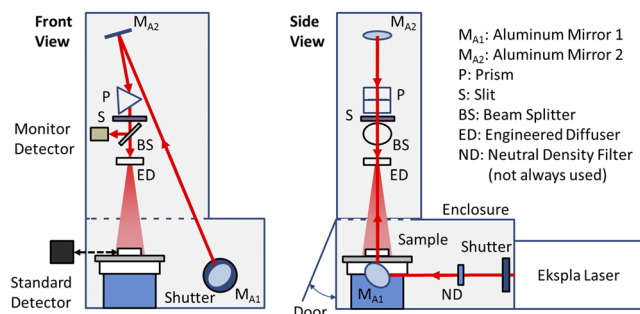
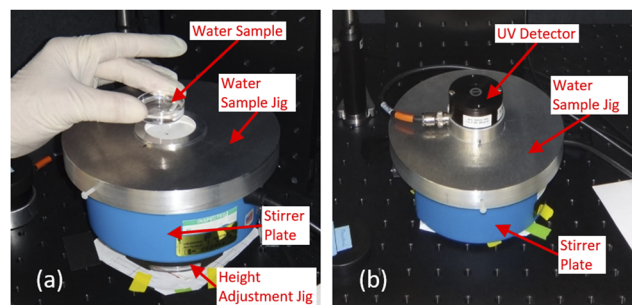## C. Test (Petri dish) position setup

Each water sample was a microbial suspension in a continuously stirred Petri dish of 36 mm diameter.[16,22] Key to these measurements was the ability to repeatably place the Petri dish containing the water sample in the same position in the optical beam and to measure the irradiance in the same position. This was done with an aluminum alignment jig placed on top of the stirring plate. The height of the stirring plate was also adjusted to measure the irradiance at the same plane as the surface of the water in the Petri dish. Figures 5 (a) and (b), respectively, show the water sample being placed into the jig and the UV detector in place for irradiance measurement.

D. Detector

The NIST calibrated UV detector was placed at the water sample location to measure the irradiance at each wavelength of interest. The UV detector consisted of an International Radiation Detectors (now Opto Diode Corp.[23]) SXUV100 silicon photodiode, known to be stable with UV exposure,[24] and a precision 8 mm diameter electroformed aperture in a cylindrical aluminum housing. The photodiode output was measured with a Keithley 6517 series electrometer. The irradiance responsivity $[A/(\mu W/cm^2)]$ of the UV detector was calibrated from 200 nm to 400 nm at the NIST in the UV Spectral Responsivity Facility.[25] The expanded uncertainty ($k = 2$) for the UV detector irradiance was 5%.

E. Performance

The uniformity of the UV irradiance at the water sample (measured separately by the project collaborators in St. Albans, VT) was calculated as the ratio of the average of the incident irradiance over the area of the Petri dish to the irradiance at the center of the dish. This has been referred to as the Petri Factor[16] and was measured at each wavelength before exposing any water samples. To determine the Petri factor, a broadband UV-C radiometer with a 1 mm aperture was scanned over the area of the Petri dish along orthogonal axes in 5 mm steps. The Petri factor was reported to range from 0.94 to 1.05,[22] which met the design goal of 10%. To reduce the time required to determine the Petri factor, a test using digital images and the fluorescence from typical card stock paper was explored. The test validated the approach, but time constraints kept it from being

FIG. 4. The NIST tunable UV irradiance laser system optical configuration for 210 nm–230 nm. The irradiance system with the dielectric mirrors replaced by aluminum mirrors and adding a prism and slit. The UV detector is substituted for the water sample to measure the irradiance at each wavelength of interest.


FIG. 5. Photographs of the stirring plate with (a) a water sample being placed into the Petri dish positioning jig and (b) the UV irradiance detector set for measurement at the same plane as the surface of the water in the Petri dish.
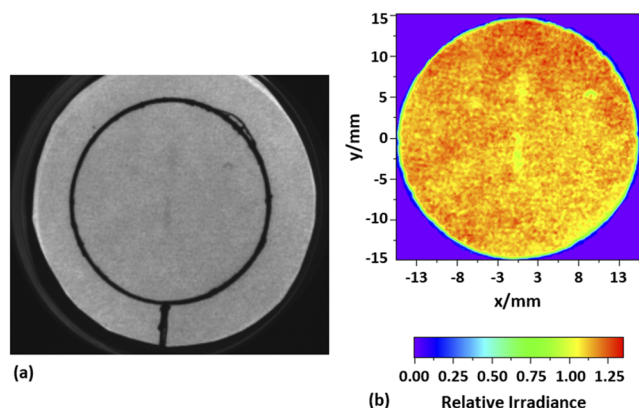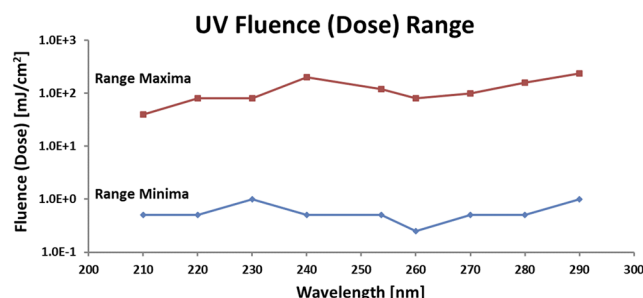
FIG. 6. Example of the irradiance uniformity at 253.7 nm. A photograph (a) from the camera mounted above the water sample imaging the fluorescence from typical card stock paper. The dark circle marks the area of the water sample Petri dish. A plot (b) of the relative irradiance uniformity normalized to the beam center.
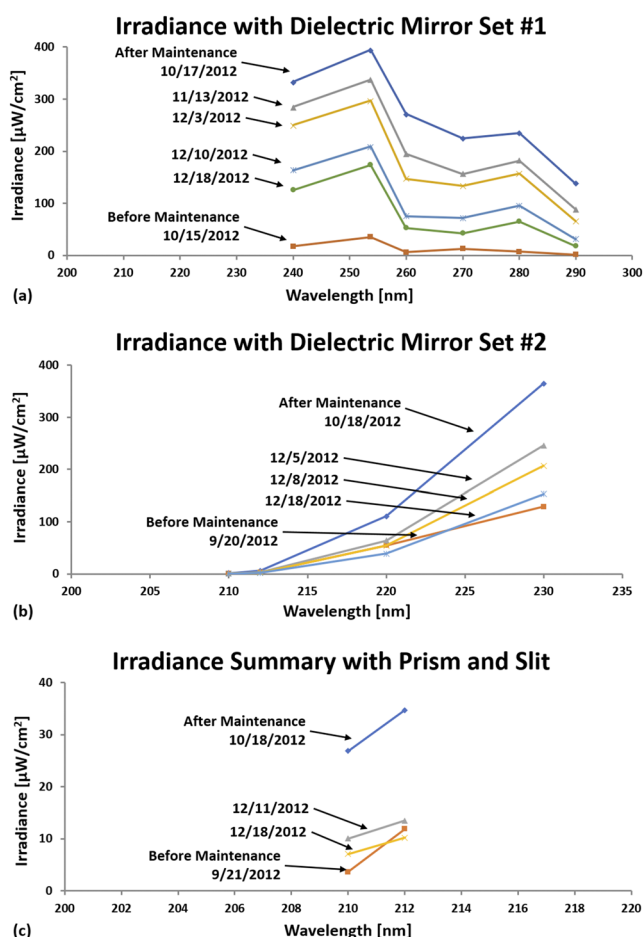
FIG. 7. Graphs (a)–(c) showing the decrease in irradiance (μW/cm^2) over time and the improvement when some of the damaged Ekspla laser optical components were replaced. Graph (a) is the irradiance with dielectric mirror set 1, (b) mirror set 2, and (c) the prism and slit.


FIG. 8. Graph of the range of fluence or dose (mJ/cm^2) by the wavelength used during this project.

studied further during this project. An example of a digital image from a camera mounted above the water sample and irradiance uniformity normalized to the beam center at 253.7 nm is shown in Fig. 6.

The irradiance levels consistently decreased over time due to unavoidable UV damage to some of the optical components in the Ekspla laser. Midway through the project, some of the damaged Ekspla laser optical components were replaced, which increased the irradiance levels at the UV wavelengths of interest. However, the UV damage continued to decrease the irradiance levels that could be provided. This did affect the scheduling of which microbes were exposed and the sequential order of wavelengths. Figure 7 shows the decrease in irradiance (μW/cm^2) over time and the improvements when some of the damaged Ekspla laser optical components were replaced. The range of fluence or dose (mJ/cm^2) by wavelength used during this project is shown in Fig. 8.

An Instrument Systems[26] CAS 140CT array spectrometer was used to measure the relative spectral irradiance at the water sample position over the spectral range from 200 nm to 600 nm both with and without spectral filtering (dielectric mirrors or a prism and slit). The visible light co-aligned with the laser beam without spectral filtering was lower by one order to one and a half orders of
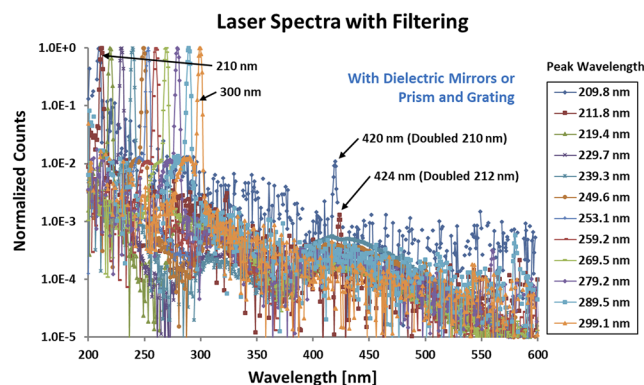
FIG. 9. Plots of the TTUV facility relative irradiance from 200 nm to 600 nm with spectral filtering (dielectric mirrors or a prism and slit) at each of the UV wavelengths of interest confirming the visible light in the beam was reduced to an acceptable level. Each spectral scan is normalized to the peak wavelength.

magnitude from the selected UV wavelength. Figure 9 shows the relative spectral irradiance with the spectral filtering at each of the UV wavelengths of interest confirming the visible light in the beam was reduced to an acceptable level. For example, the 420 nm peak in Fig. 9 is 1% of the UV flux at 210 nm. With the excitation source at 212 nm and the prism filtering shown in Fig. 4, the flux at 424 nm is reduced by an order of magnitude, to the 0.1% level.

III. DISCUSSION

The types of waterborne pathogens and surrogate micro-organisms studied were: Adenovirus—RG 2, also Type 40 and 41, *Cryptosporidium parvum* (Iowa strain)—RG 2, *Giardia*—RG 2, and Coliphages MS2, T1UV, T7m, Q beta.

Results and impact of the TTUV laser irradiance facility in the water treatment microbiology are shown in several papers.[8,22,27,28] A fundamental question is the equivalence of UV dose response using LP UV light and the NIST 1 kHz pulsed laser. The inactivation of adenovirus using a LP UV light source and the NIST laser at 253.7 nm did not show a statistically significant difference in response.[27] Figure 10 shows the relative spectral sensitivity, or action spectra, of several micro-organisms, MS2, T1UV, Q Beta, T7m, and T7 Coliphages and *C. parvum* to UV light, showing differences in the action spectra below 240 nm. Extending the action spectra to 210 nm for selected drinking water pathogens and surrogates used to validate water systems[22] is necessary for calculating the action spectra correction factors for MP UV water system validation. Better understanding of the action spectra improves the comparison of pathogens and various surrogates used in testing UV water treatment systems, the selection of wavelengths for dose monitoring, and the wavelengths for future technology exploration.[8]

There are several improvements under way for the TTUV facility. The simplest improvement to the tunable UV irradiance laser system is to establish computer control of the laser wavelength. Another improvement is to develop dielectric mirrors that can work at 210 nm. This would likely require some collaboration

with the manufacturer to verify the mirror performance. To reduce the uncertainty in the irradiance during a test, a calibrated detector will be installed to monitor the irradiance during the water sample exposure. Refinements to the detector signal measurement method will be explored. Finally, as mentioned above, a camera to analyze the irradiance uniformity and calculate the Petri factor in real time will be installed. The camera will reduce the time needed to measure the Petri factor and provide irradiance uniformity information for the entire beam and not just along two orthogonal axes through the beam center.

A static (non-transportable) UV dose irradiance system is under development for NIST's Remote Sensing Laboratory. The changes detailed above will be affected for the new static system, and the performance of the enhanced system will be evaluated. Changes deemed worthwhile will be introduced into the TTUV system.

IV. CONCLUSIONS

Current UV systems rely on lamp sources that are broadband and spectrally non-uniform, characteristics that complicate the measurement analysis process and ultimately degrade the accuracy of the pathogen disinfectant action spectra. The NIST TTUV facility has demonstrated its ability to be deployed to a field site and to provide irradiance at narrowband, and well defined, UV wavelengths at irradiance levels of 10 μW/cm^2 to >100 μW/cm^2 and fluences (or doses) from <1 mJ/cm^2 to >100 mJ/cm^2 from 210 nm to 300 nm.

Development of action spectra of waterborne micro-organisms to specific UV wavelengths as part of WaterRF Project 4376 demonstrates the unique capabilities of the NIST tunable UV irradiance laser system and its potential application for other biological experiments, for example, development of action spectra for airborne pathogens. Since the spectral range of the TTUV facility extends from 210 nm to 2500 nm, it can be used as a spectrally tunable source of radiant flux for a wide variety of applications ranging from UV dose studies to photometric sensor calibration, and more generally, filter radiometer and spectrograph calibrations. The ability to transport the system to a facility where subject micro-organisms and viruses are located allows experienced staff, often with specialized facilities, to prepare, irradiate, handle, and analyze them.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.
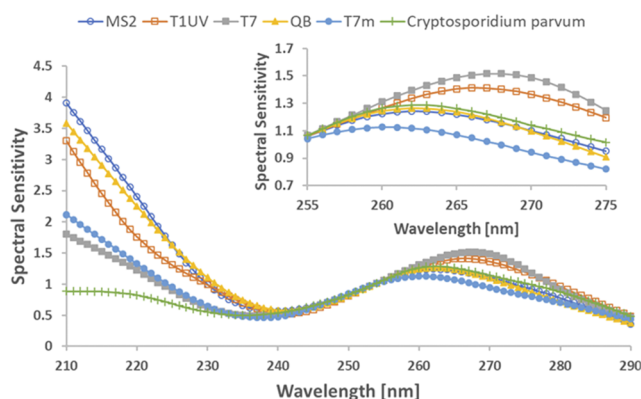
FIG. 10. Relative spectral sensitivity, or action spectra, of the studied micro-organisms MS2, T1UV, Q Beta, T7m, and T7 Coliphages and *Cryptosporidium parvum* to UV light from the tunable laser. Reproduced with permission from Water Res. **70**, 27–37 (2015).[8] Copyright 2015 Elsevier.

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[29] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.