

Available online at www.sciencedirect.com





Procedia Chemistry 8 (2013) 248 - 257

Youth in Conservation of Cultural Heritage, YOCOCU 2012

The painted silk panels of Palazzo Barberini at Rome. The scientific investigation and preservation challenge

Claudia Pelosi^{a*}, Giulia Falletta^b, Barbara De Dominicis^c, Pietro Baraldi^d

^aDepartment of Cultural Heritage Sciences, University of Tuscia, Piazzale dell'Università, Viterbo 01100, Italy ^bOrganic material area, University of Palermo, Via Provinciale Petralia Sottana, Palermo, Italy ^cLaboratorio Tessili Antichi, Via Genova, Viterbo 01100, Italy ^dDepartment of Chemistry, University of Modena and Reggio Emilia, Via Giuseppe Campi, Modena, Italy

Abstract

This paper reports the study carried out on the 18th century painted silk panels that cover the walls of the so called *Salotto delle sete dipinte* in Palazzo Barberini at Rome on the occasion of the conservation work started in 2007. Microscope and spectroscopy investigations were performed in order to study the materials and the realization techniques of the painted silk panels and to evaluate the conservation conditions. Moreover ageing tests were performed in order to evaluate the stability of the consolidant/protective product used for the silk panels.

© 2013 The Authors. Published by Elsevier B.V. Open access under CC BY-NC-ND license. Selection and peer-review under responsibility of the IA-CS (Italian Association of Conservation Scientists) and University of Antwerp

Keywords: painted silk, pigments, fibers, infrared spectrometry, micro-Raman spectroscopy, consolidant/protective

1. Introduction

Object of this paper are the painted silk panels that adorn the walls of the so called 18th century *Salotto delle sete dipinte* in Palazzo Barberini at Rome (Fig. 1). The drawing room is part of the eighteenth-century apartment of Palazzo Barberini [1, 2]. The silk panels depict life scenes of the American Indians, based especially on the 16th century drawings of John White, an English naturalist traveller [3]. The drawings were spread in Europe by

1876-6196 $\[mathbb{O}\]$ 2013 The Authors. Published by Elsevier B.V. Open access under CC BY-NC-ND license.

Selection and peer-review under responsibility of the IA-CS (Italian Association of Conservation Scientists) and University of Antwerp doi:10.1016/j.proche.2013.03.031

^{*} Corresponding author. Tel.: +39-0761-357684; fax: +39-0761-357017.

E-mail address: pelosi@unitus.it

the Jesuits which promoted the knowledge of the countries in course of Christianisation till from the end of 16th century [4].



Fig. 1. A view of the Salotto delle sete dipinte with the plan and the numbers of the panels.

The decorative structure of the drawing room is made of 15 panels, 4 ovals above the doors and 2 other ovals above the mirrors of the side walls. Precious wooden frames enrich the panels and the ovals. The panels were marked with alpha numeric abbreviations where the letters specify the position of the panel in the room, for example SW indicates South-West, NE North-East, and so on, followed by a progressive number starting from south west side in counterclockwise (Fig. 1).

The conservation work, carried out under the Direction of Dr. Anna Lo Bianco of the Soprintendenza per il Patrimonio Storico, Artistico ed Etnoantropologico e del Polo Museale della Città di Roma, was the occasion to perform some investigations to evaluate the state of preservation of the silks and to know the constitutive materials to supply a valid aid to the restorers' work. The private restorers who carried out the intervention deemed it absolutely necessary to carry out a diagnostic campaign before and during their work in spite of the laboratory analysis could extend the time for the restoration. It is worth noting that this procedure is not usually adopted by the private restorers that often must respect the times imposed by the customer renouncing to the possibility of a diagnostic campaign.

2. The technique and state of preservation

The conservation work, started in 2007, was the occasion to carefully investigate the technique, the materials and the state of preservation of the silks. In order to document the state of preservation, conservative cards were created for the panels and ovals together with their graphical representation [1].

Concerning the technique of execution of the painted silks, the restorers carried out an in-depth investigation both of the textiles and of the paintings. The observations, especially after the disassembly of the panels to bring them in the restoration laboratory, allowed detecting that the silk textile pieces were sewn together by a variant of a felled seam ("English" technique) or with an "open edges" system, before the application of the paintings. The "English" seam was created by putting the selvage of a silk length on another and by sewing them with small stitches. The paintings were created directly on the silk without a setting layer by using an inch drawing. The silk textiles were glued to a support fabric visible in the surface lacunae. This procedure was usually performed to create a support for the silk textiles as they are too weak [5]. It was not possible to know the original system used to hang the silk panels to the walls due to the various interventions carried out during the centuries. The 1960's restoration examination reported that the panels were directly nailed to the walls [6].

The *in situ* and laboratory observation of the silks allowed detecting different kinds of damages caused by the natural ageing of the materials but also by the past restoration interventions. In particular, the last restoration, carried out in 1960, was performed with the same procedure used for the paintings without recognizing the peculiarity of the artefact. The archive documentation of this intervention is very poor; it reports only that a rubberized fabric was applied on the back of the panels and that a fixative was spread on the surface [6]. The rubberized fabric and the fixative appeared deeply yellowed at the beginning of the 2007's intervention. Moreover, the careful observation of the artefact revealed other damages like: depolimerization of the silk fibers that caused tears and holes in the silk; dust and dirt deposits; water stains; colour fading. The negligence and the lack of ordinary maintenance, as pointed out also in the restoration relation of 1960, made the situation worse. Probably the damages of the silk and of the painting materials were also caused by the light coming from a large window in the south-east wall [7]. Some light measurements, carried out during the *in situ* inspection put in evidence high values of illuminance and irradiance due both to the window and to the unsuited lighting sources. According to the CIE 157: 2004 (Control of damage to museum objects by optical radiation), silk and colorants are included in the category 4 of light sensitivity, i.e. high responsive objects [8].

3. Scientific investigation

The thorough *in situ* investigation, also with the aid of macrophotography, allowed choosing the most appropriate number of samples to perform the laboratory analysis by microscopy and spectroscopy techniques. The samples' list and description is shown in Table 1.

Sample nr.	Description
1	Panel SW5, powder from the gold frame
2	Panel NE12, powder from a red flower in the frame
3	Panel NE12, powder from the blue area of the frame
4	Panel SW1, micro fragment of the cloth
5	Panel SW1, powder from the green in the landscape
6	Panel SE6, micro fragment from the green of a tree
7	Panel SE6, micro fragment from the gold frame
8	Panel SE6, micro fragment from the blue area of the frame
9	Panel SE6, micro fragment from a red flower in the frame
10	Panel NW12, dark blue powder from a flower
11	Panel NW12, light blue powder from a leaf in the frame
12	Panel SE6, micro fragment from the red plumage of the parrot
13	Panel NW12, powder from a light blue-green leaf in the frame
14	Panel SW1, powder from the very light blue-green of a leaf in the frame
15	Panel NE10, powder from the highlighting on the blue of the frame

Table 1. Samples' description and their location on the panels.

Observation and photography of the sample cross-sections and pigment powders embedded in Canada balsam were performed by a Zeiss Axioskop polarising microscope equipped with a Zeiss AxioCam digital camera. Cross-sections were studied also under UV lighting using a Mercury Vapour lamp directly connected to the

microscope in order to observe fluorescence of the materials. A filter was interposed between the mercury lamp and the sample with the following characteristics: excitation BP 365/12, beam splitter FT 395, emission LP 397.

Scanning electron microscopy (SEM) on the fibers was carried out using a scanning electron microscope Jeol modello JSM-5200. The samples were sputter-coated with gold in a Balzers MED 010 unit.

Infrared spectra were obtained using a Nicolet iS10 Fourier transform spectrometer. For each sample 200 scans were recorded in the 4000 to 400 cm⁻¹ spectral range in diffuse reflection modality (DRIFT) with a resolution of 4 cm⁻¹. Spectral data were collected with OMNIC 8.0 (Thermo Electron Corporation) software. Samples have been ground with spectrophotometric grade KBr (1% sample in KBr) in an agate mortar. As background the spectrum of the KBr powder was used.

The micro-Raman spectrometer was a Labram Model of the Horiba JobinYvon with a spatial resolution of 1 μ m and the possibility of fast detecting owing to the use of a CCD detector with 1026x256 pixels cooled to -70°C by the Peltier effect. The spectral resolution was 1 cm⁻¹. The exciting wavelength was the 632.8 nm red line of a He-Ne laser. Integration times varied between 10 and 20 s with 5 accumulations.

A relatively new polymer in conservation science, commercially named Aquazol200[®], was tested for the silks. To perform the accelerated ageing tests, an 8% (w/v) water solution of Aguazol200[®] was applied on glass slides and on an 18th century silk fragment available to the Laboratory Tessili Antichi. Only one half of the fragment was treated with Aquazol200® in order to make a comparison between the colour changes of the treated and untreated silk. The accelerated ageing of the samples was performed in a Model 1500E Solar Box (Erichsen Instruments). The system is equipped with a 2.5 kW xenon-arc lamp and an UV filter that cuts off the spectrum at 280 nm. The samples were exposed in the Solar Box chamber up to 1000 h at 550 W/m^2 , 55°C and the UV filter at 280 nm. The experimental conditions were chosen following the specifications supplied by Erichsen, in order to simulate the sunlight exposition. After the exposure time the sample was removed from the Solar Box chamber and the colour was measured using an X-Rite CA22 reflectance spectrophotometer. The characteristics of the colour measuring instrument are the following: colour scale CIEL*a*b*; illuminant D65; standard observer 10°; geometry of measurement $45^{\circ}/0^{\circ}$; spectral range 400-700 nm; spectral resolution 10 nm; measurement diameter 4 mm; white reference supplied with the instrument. The CIELAB colour system was used where L* describes the lightness while a* and b* describe the chromatic coordinates on the green-red and blue-yellow axes, respectively. The differences in lightness (ΔL^*), chromatic coordinates (Δa^* and Δb^*), and total colour (ΔE^*) were then calculated using these parameters according to Normal 14/93 (1993) and EN 15886 (2010). Colour measurements of the Aquazol200[®] samples were performed by putting a standard white paper (Kodak Color Control Patches - white) on the back of the glass plates.

4. Results and discussion

4.1. Painting materials

The main pigments were identified by optical microscopy and micro-Raman spectroscopy by comparing the obtained images and spectra to literature data and spectral databases [9-15], but also to samples prepared in our laboratory.

Red colour of the flowers was obtained with an organic dye (Fig. 2). The optical characteristic of this dye suggest the presence of Brazil wood dye. The bright red of the birds was obtained with the addition of mercury red. The Raman spectrum of the sample 12 exhibits the characteristic bands of vermilion: 259(vs) cm⁻¹, 294(w) cm⁻¹, 348(m) cm⁻¹ and 356(m) cm⁻¹.

Blue colour in the frame was created with indigo and smalt (samples 3 and 8), whereas in the flowers and in the leaves only indigo was found (samples 10 and 11). The Raman spectrum of indigo detected in the examined samples is shown in fig. 3. Smalt is characterized by transparent isotropic light blue particles that appear dark between crossed polars under the microscope [16].



Fig. 2. Microphotographs in transmitted light of the sample nr. 2 (A) and of Brazil wood lake (B) prepared in our laboratory from Brazil wood.

The green colour in the landscape was obtained with malachite found both in the trees and in the hills. Malachite was identified both through optical microscopy and micro Raman spectroscopy (Fig. 4).



Fig. 3. Raman spectrum of indigo, found in the blue samples.



Fig. 4. Raman spectrum of malachite, found in the green samples from the trees and from the hills.

The green colour of the leaves in the frames of the panels appear faded, and it was hypothesized the presence of an organic dye. Two samples were analysed under optical microscopy (13 and 14) and by micro Raman spectroscopy and only indigo was found. According to these results, we can say that the leaves in the frame were obtained with indigo and that the differences observed in the colour hue are due both to the amount of the dye and to its fading caused by the exposition to the light. In fact, the leaves that were protected by the wood frames appeared dark blue in the colour during the disassembly of the panels.

The internal micro stratigraphic analysis confirmed that the pigments were applied directly on the silk without any setting layer (Fig. 5A,B). The gilding in the frame was applied by a priming (the Italian *missione*) that exhibits an intense orange fluorescence under ultraviolet radiation characteristic of shellac (Fig. 5C,D). Under the priming the silk fibers and those of the linen can be observed.



Fig. 5. Microphotographs of samples 8 (A, B) and 7 (C, D), under reflected light (A, C) and UV fluorescence (B, D).

FTIR analysis was performed in order to investigate the binder typology and the composition of the materials used in the 1960's restoration to supply an aid to the conservation work.

Study and interpretation of FTIR spectra were obtained by comparing the experimental results with literature data [17-18], and spectral databases [13].

FTIR spectrum of the materials of the rubberized fabric (sample 4) was obtained on the powder scraped from the surface of the fabric (Fig. 6).



Fig. 6. FTIR spectrum in DRIFT modality of sample 4 (powder scraped from the rubberized fabric).

The main bands visible in the spectrum (1713 cm⁻¹, 1453 cm⁻¹, 1379 cm⁻¹ and 1245 cm⁻¹) can be associated to a natural isoprene resin, normally used as main component of the rubberized fabric [19]. The two narrow bands at 2918-2850 cm⁻¹ together with the peak at 1168 cm⁻¹ suggest the presence of a wax, another common ingredient of

the rubberized fabric. The bands at 3676 cm⁻¹, 1016 cm⁻¹, 830 cm⁻¹, 768 cm⁻¹, 537 cm⁻¹, 467 cm⁻¹ and 456 cm⁻¹ can be associated to the presence of clay minerals [20]. In the rubberized fabric, clay was usually contained in a percentage similar to that of the isoprene resin [19].

The characterization of the materials of the rubberized fabric supplied a valid aid to the restorers in order to choice the most appropriate solvent system to remove it from the back of the linen.

The interpretation of FTIR spectra, performed on some chosen samples from the painted surfaces, was far from easy due to the presence of several different materials both original and added during the past restorations.

For example, in fig. 7 the spectrum of sample 2 is shown. Proteinaceous compounds were detected due to silk and to the glue used to fasten the silk to the linen, bands at 3420 cm⁻¹, 3290 cm⁻¹, 3085 cm⁻¹, 1650 cm⁻¹, 1546 cm⁻¹, 1444 cm⁻¹, 1408 cm⁻¹, 1163 cm⁻¹. Some bands can be associated to polysaccharide compounds (3468 cm⁻¹, 1630 cm⁻¹, 1039 cm⁻¹). The bands at 2932 cm⁻¹ and 2862 cm⁻¹, due to the C-H stretching, are common to proteins and polysaccharides. According to these results, a polysaccharide binder was hypothesized for the paintings (watercolour technique).



Fig. 7. FTIR spectrum in DRIFT modality of sample 2.

In the sample taken from the gilding in the frame (sample 1), FTIR analysis revealed the presence of lead white (bands at 1409 cm⁻¹, 1045 cm⁻¹, 839 cm⁻¹, 683 cm⁻¹), of a resin and a siccative oil (Fig. 8).



Fig. 8. FTIR spectrum in DRIFT modality of sample 1 (gilding).

The bands of the resin and of the siccative oil are partially overlapped, in particular the bands at 2924 cm⁻¹ and 2854 cm⁻¹ due to C-H stretching. These bands are usually well defined and very strong both for siccative oils and shellac. The other bands associated to siccative oil are: 1715 cm^{-1} (C=O stretching of free carboxylic acids due to oil ageing), 1170 cm^{-1} , 1093 cm^{-1} and 722 cm^{-1} (C-H torsion band, common to shellac). The bands at 1623 cm⁻¹ and 1540 cm⁻¹ can be attributed to the formation of carboxylate anions coordinated to lead [21]. The bands at 1245 cm⁻¹ and 947 cm⁻¹ are due to shellac.

The presence of shellac was gathered combining the results of the FTIR analysis and UV fluorescence observation of the cross section (see fig. 5D).

4.2. Fibers

The optical microscope observation of the fibers from the cloth glued to the silk allowed detecting the presence of flax. The flax fibers have a cylindrical regular shape with a fine central canal (lumen) [22]. The cross markings, known as nodes, visible on the fibers give them the characteristic microscopic appearance of flax. At last, the polygonal cross-section of the fibers is another typical feature of flax.

Silk have very fine, regular fibers with a triangular cross-section (see fig. 6). The silk fibers, observed under the scanning electron microscope, appear broken and disjoined suggesting the necessity to perform a consolidation (Fig. 9).



Fig. 9. SEM image of silk fibers

4.3. Consolidant testing

To perform both the adhesion of the silks and the consolidation of the painted layer, the restoration laboratory chose to use Aquazol200® a relatively new polymer in conservation science and, in particular, for textiles [23-24]. Aquazol is a polymeric material based on the monomer 2-ethyl-2-oxazoline with some interesting properties: it is soluble in a wide range of solvents, including water, exhibits both adhesive and consolidant capabilities and maintains its solubility after ageing tests [25-28].

Aquazol® 200 was tested on a silk fragment, available at the laboratory Tessili Antichi, and on glass slides by measuring the colour before and after the ageing time of 1000 hours in a Solar Box chamber. During the restoration work there was no time to perform further laboratory tests, but only colour measurements were used to evaluate the Aquazol® 200 stability. Nevertheless, it must be stressed that, as demonstrated in literature papers, colour change method is highly sensitive to determine the extent of photo degradation of various kinds of

materials exposed to ultraviolet and visible radiation [29]. Moreover, non-destructive methods must be applied to avoid the paradox of damaging a work of art while monitoring its preservation state [30].

As the colour changes can be correlated to the chemical modifications of the surfaces, colour measurements could become a simple method to evaluate the photo degradation of the materials.

The results of the colour measurements, expressed as ΔL^* , Δa^* , Δb^* and ΔE^* , showed that in general Aquazol® doesn't affect colour changes. In fact, colour changes depend mainly on silk and pigment colour changes. The maximum ΔE^* value was obtained for the red areas both in the treated and untreated part of the silk fragment ($\Delta E^*=15$, due especially to a decrease of b* coordinate, $\Delta b^*=-10$). The ΔE^* values measured for Aquazol® samples on glass plates were less than 3, demonstrating the colour stability of this product to the artificial ageing.

Further experimental tests will be necessary to verify the behaviour of Aquazol \mathbb{R} on silk textiles also with the aid of FTIR and chromatography – mass spectrometry techniques and mechanical tests. In this regards, the study is still in progress.

5. Conclusions

The study on the painted silks of Palazzo Barberini in Rome allowed to characterize the pigments and partially the binders. The difficulty to characterize the binders by FTIR spectrometry was due to the presence of several organic materials both original and added during past restoration interventions.

Some restoration materials were examined especially with the aim at supplying a valid aid to the restores during the cleaning, consolidation and adhesion processes. In particular, the materials of the rubberized fabric were characterized allowing the restores to choose the most appropriate solvent system to remove it. Some tests were performed on the consolidant/adhesive chosen for the silks. As far as we know, Aquazol® 200 was applied for the first time on painted silk textiles both as adhesive and consolidant.

Some light measurements, carried out during the *in situ* inspections put in evidence high values of illuminance and irradiance due both to the presence of a large windows in front of the silk panels and to the unsuited lighting sources. We pointed out this problem to the Superintendence suggesting the removal of the standard lamps and the advisability of keeping close the curtains of the large windows. During our last inspection, carried out in June 2012, we were pleased to verify that the lamps were removed and that anti-UV glasses were applied to the windows. Moreover, an extraordinary maintenance of the rooms was performed. We hope that our work contributed to a better conservation and preservation of the painted silks in the *Salotto delle Sete Dipinte* in Palazzo Barberini at Rome.

References

- [1] Falletta G. Le tappezzerie in seta dipinta di Palazzo Barberini, Thesis, Viterbo: Università degli Studi della Tuscia; 2007.
- [2] Magnanimi G. The eighteenth century apartments in the Palazzo barberini. Apollo 1984; 272:252-61.
- [3] Guadalupi G. Wingandacoa. La colonia perduta con acquerelli di John White. Milano: F. M. Ricci; 1992.
- [4] Cocchiara G. L'eterno selvaggio. Palermo, Sellerio Editore; 2000.
- [5] Skelton M., Lee-Whitman L., A systematic method for Differentiating Between 18th century Painted-Printed Chinese and Western Silks. In: Needles HL, Zeronian SH, editors, *Historic textile and paper materials: conservation and characterization*, Advances in Chemistry, 212, Washington DC: American Chemical Society; 1986, p. 131-151.
- [6] Brugnoli M.V. Relazione tecnico-artistica. In: Perizia del restauro del 1959/60, Archivio dei restauri storici della Galleria Nazionale d'Arte Antica di Palazzo barberini, Roma, 1960.
- [7] Willis K., Flaherty S., Paulocik C., The Conservation of 18th-Century Painted Silk Dress. New York: The Costume Institute, The Metropolitan Museum of Art; 1995.
- [8] Bacci M., Cucci C. The role of light. In: Camuffo D, Fassina V, Havermans J, editors. Basic Environmental Mechanisms Affecting the Cultural Heritage, Firenze: Nardini Editore; 2010, p. 59-68.
- [9] Eastaugh N., Walsh V., Chaplin T., Siddall R. Pigment Compendium. Optical Microscopy of Historical Pigments. Amsterdam, Elsevier Butterworth-Heinemann; 2004.

- [10] http://www.fis.unipr.it/phevix/ramandb.php
- [11] http://minerals.gps.caltech.edu/FILES/raman
- [12] http://rruff.info
- [13] Infrared and Raman User Group (IRUG), Edition 2000, Spectral Data Base, http://www.irug.org
- [14] Burgio L., Clark R.J.H. Library of FT-Raman spectra of pigments, minerals, pigment media and varnishes, and supplement to existing library of Raman spectra of pigments with visible excitation. Spectrochimica Acta Part A 2001;57:1491-1521.
- [15] Bouchard M., Smith D.C., Catalogue of 45 reference Raman spectra of minerals concerning research in art history or archaeology, especially on corroded metals and coloured glass. *Spectrochimica Acta Part A* 2003;59:2247-2266.
- [16] Mühlethaler B., Thissen J. Smalt. In: Roy A, editor. Artists' Pigments. A Handbook of Their History and Characteristics, Volume 2, Washington: National Gallery of Art; 1993, p. 113-130.
- [17] Derrick MR, Stulik D, Landry JM. Infrared Spectroscopy in Conservation Science. Los Angeles: The Getty Conservation Institute; 1999.
- [18] Adrover Gracia I, Applicazioni della spettrofotometria IR allo studio dei beni culturali. Collana i Talenti. Padova: Il Prato; 2001.
- [19] Turco A. Nuovissimo ricettario chimico. 3rd ed. Milano: Hoepli, 1990, p. 1511-1512.
- [20] De Benedetto GE, Laviano R, Sabbatini L, Zambonin PG. Infrared spectroscopy in the mineralogical characterization of ancient pottery. J Cult Her 2002; 3:177-186.
- [21] Meilunas RJ, Bentsen JG, Steinberg A. Analysis of aged paint binders by FTIR spectroscopy. Studies in Conservation 1990; 35:33-51.
- [22] Identification of Textile Materials. Manchester: The Textile Institute; 1985, p.
- [23] Borgioli L, Cremonesi P. Le resine sintetiche usate nel trattamento di opere policrome. Collana i Talenti. Padova: Il Prato; 2005, p. 171-172.
- [24] Lechuga K. Aquazol As a Heat Set Adhesive for Textile Conservation Treatments. American Institute for Conservation of Historic and Artistic Textile Specialty Group Postprints 2009; 9:187-193.
- [25] Wolbers RC, McGynn M, Duerbeck D. Poly(2-Ethyl-2-Oxazoline): a new conservation consolidant. In Dorge V, Howlett FC, editors, Painted wood: Hisrory and Conservation. Los Angeles: Getty Conservation Institute; 1994, p. 514-527.
- [26] Friend S. Aquazol: one conservator's empirical evaluations. WAAC Newsletter 1996; 18:n° 2.
- [27] Arslanoglu J. Evaluation of the use of Aquazol as an adhesive in painting conservation, WAAC Newsletter 2003; 25:nº 2
- [28] Arslanoglu J. Aquazol as used in conservation practice, WAAC Newsletter 2004; 26:nº 1.
- [29] Lo Monaco A, Marabelli M, Pelosi C, Picchio R. Colour measurements of surfaces to evaluate the restoration materials. In Pezzati L, Salimbeni R, edirors, O3A: Optics for Arts, Architecture, and Archeology III, Proceedings of SPIE, 25-26 May 2011, Munich, Germany, SPIE, Washington, 2011, Volume 8084, art. Nr. 80840P-1-14.
- [30] Marengo E, Robotti E, Liparota MC, Gennaro MC. Monitoring of pigmented and wooden surfaces in accelerated ageing processes by FT-Raman spectroscopy and multivariate control charts. *Talanta* 2004; 63: 987-1002.