




Electrohydrodynamic spray applicator for homogenous application and reduced overspray of sunscreen

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Abstract

Background: The recommended amount of sunscreen by hand application (2 mg/cm²) is in reality not achieved, which decreases the homogeneity and thereby the effective sun protection factor (SPF).

Materials and Methods: The homogeneity of sunscreen applied by a newly developed spray applicator using an electrostatically charged aerosol, for which a hand rubbing of the formulation is not necessary, is evaluated. In vivo experiments were performed on the volar forearms of human volunteers using the spray applicator compared to the standardized hand application according to ISO 24444.

Results: The distribution homogeneity was assessed qualitatively using in vivo laser scanning microscopy and quantitatively by absorption spectroscopy after tape stripping and by the standard deviation of multiple spatially displaced reflectance measurements for non-invasive SPF determination below the minimal erythemal dose, which showed a significantly higher homogeneity by 20.9% after spray application compared to hand application.

Conclusion: Non-invasive SPF determination of multiple spatially displaced reflectance measurements was proven to be a suitable method for the non-invasive determination of the sunscreen distribution homogeneity. Electrostatically charged spray application increased the sunscreen distribution homogeneity on the skin and can reduce the amount of overspray.

KEYWORDS

confocal laser scanning microscopy, skin, spectroscopy, sun protection factor, sunscreens

1 | INTRODUCTION

The occurrence of skin cancers is rising, which is linked to the exposition of solar radiation. The World Health Organization (WHO) estimates annually 132 000 cases of malignant melanoma and 2-3 mio cases of non-melanoma skin cancer worldwide.¹ Although the risk

awareness and thereby the use of sunscreens are rising, the skin cancer occurrence does not decrease.

This could be explained by the formation of free radicals not only due to the exposition in the UV, but also in the visible and infrared spectral range.² Further, it is expected that the recommended amount of 2 mg/cm² of sunscreen on the skin³ is in reality not achieved in most cases,⁴ entailing an effectively lower SPF.⁵ It has also been shown that the applied

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amount differs depending on the application method.⁶ Lower film thicknesses were found after firm rubbing, compared to light application.⁷ Novic et al found that the usual application amount was 1.1 mg/cm² for a sunscreen lotion and 1.6 mg/cm² for a pump spray.⁸ In this case, the uneven skin topography can limit the homogeneity of the sunscreen distribution.^{9,10} Especially when applying a lower amount of sunscreen on the skin by hand or rubbing in a sprayed sunscreen, the sunscreen will accumulate in the furrows and wrinkles, but will vanish from the elevated surfaces of the skin, entailing an effectively lower sun protection factor (SPF)¹¹⁻¹³ than determined according to current guidelines.¹⁴ If a sunscreen spray is applied without subsequent rubbing, this could increase homogeneity.¹⁵

Several spray applicators have been tested in order to increase the homogeneity of sunscreen distribution on the skin. However, a subsequent rubbing in by hand is mostly required. Essential advantages were not achieved by the current spray applicators, and a considerable amount of sunscreen is actually lost, because it never reaches the skin but might pollute the clothes or the environment.

In this study, the distribution homogeneity of sunscreen applied by a newly developed spray applicator using an electrostatically charged aerosol is evaluated and compared to the actually used standardized hand application according to ISO 24444.^{14,16} The ISO 24444 using hand application is the only accepted reference method to determine the SPF in vivo, also for sunscreens sold in conventional pump spray applicators. Electrostatic spraying is widely used in industrial settings to apply coatings, such as furniture, and automotive parts, with the purpose of achieving an increased homogeneity.^{17,18} The sunscreen spray is applied on the arm of a volunteer passing a charged electrode within the sprayer. The arm acts as the second neutral pole, when the volunteer holds on to a ground electrode. The statically charged sunscreen thereby gravitates to the skin, and only a low amount of sunscreen is being lost.

Laser scanning microscopy (LSM) in vivo and tape stripping have proved to be suitable methods to determine the homogeneity of applied sunscreen formulations.¹⁵ However, these methods have several shortcomings. LSM requires to add a fluorescent marker to the sunscreen, if the sunscreen itself does not fluoresce under the applied excitation wavelength. The penetration and distribution on skin of the added fluorescent marker might be different than that of the UV filters itself, or the penetration characteristics might be changed, if fluorescent markers like sodium fluorescein are fixed to the substances.¹⁹ The determination of the inhomogeneity factor by tape stripping is strongly dependent on the amount of the topically applied formulation.^{10,20}

In this study, the SPF is additionally determined non-invasively by fiber-coupled diffuse reflectance spectroscopy in vivo and the inhomogeneity is determined by the standard deviation of multiple transmittance measurements.

2 | MATERIALS AND METHODS

2.1 | Study protocol

Experiments were performed on 23 volunteers, who were instructed not to use any skin care products 12 hours prior to the

measurements. A positive vote from the ethics committee of the Charité – Universitätsmedizin Berlin (EA1/247/18 and EA1/160/19) had been obtained for the study, which was performed in accordance with the declaration of Helsinki. All volunteers had no known skin diseases, were of skin type I-III according to Fitzpatrick,²¹ were at least 18 years old, and gave their written informed consent to participate in this explorative proof of principle study.

A sunscreen was applied to the right volar forearm of the volunteers using a spray applicator. On the left volar forearm, the same sunscreen was applied by hand at 2 mg/cm² in an 8 × 10 cm² area. The hand application was performed by a trained technician using a weighed syringe and rubbing in with a glove saturated with the sunscreen according to the ISO 24444 standard.^{14,16} The standardized hand application served as a reference method allowing comparable amount of applied sunscreen. Comparison with conventional pump sprayers was not successful because the amount of sunscreen applied onto the skin varied too much. For proper comparison of the distribution homogeneity, the amount must be comparable and ideally around 2 mg/cm².³

The measurements were performed 30-60 minutes after the application of the sunscreens. In vivo LSM measurements were performed on 7 volunteers (5 female, 2 male, aged 40 ± 13 years), and tape stripping for the determination of the inhomogeneity factor and in vivo SPF measurements were performed on the remaining 16 volunteers (13 female, 3 male, aged 33 ± 11 years). The majority of volunteers were female, which allowed measurements on sufficient dimensions of hairless skin sites on the volar forearm without shaving.

2.2 | Sunscreen formulations

All measurements were performed on sunscreen formulations containing Butyl Methoxydibenzoylmethane, Octocrylene, Diethylhexyl Butamido Triazone, Ethylhexyl Salicylate, and Bis-Ethylhexyloxyphenol Methoxyphenyl Triazine as UV Filters (Institute Dr Schrader).

Only the three sunscreen formulations used for the LSM measurements (SPF 15) additionally contained 0.2% sodium fluorescein as a fluorescent marker excited at ≈490 nm and either 2% Polyamide-3 (PolyamFI), Acrylates/Octylacrylamide copolymer (AcryFI) or Polyester-10 and Propylene glycol dibenzoate (PolyesterFI) as film formers.

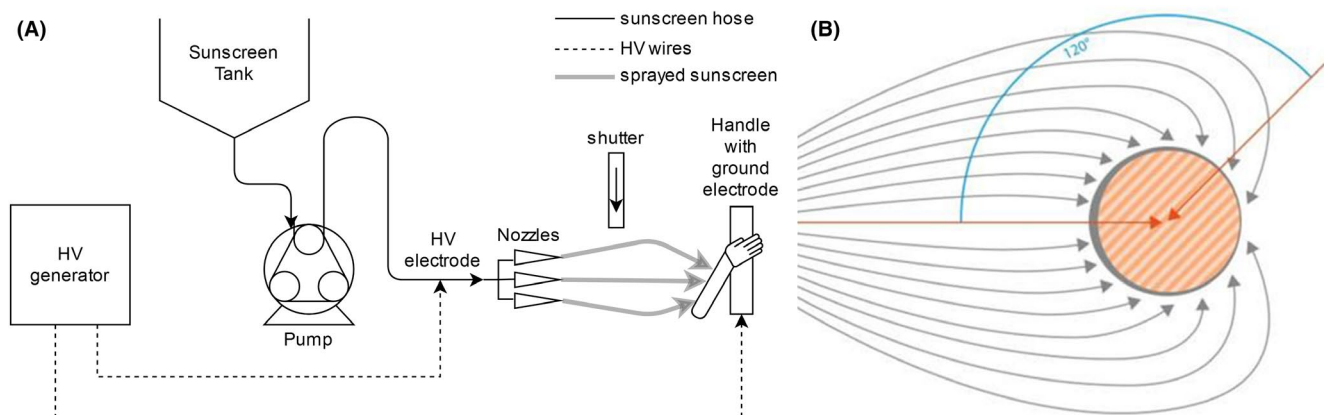
The measurements of the inhomogeneity by tape stripping and in vivo SPF determination were performed with sunscreen formulations (SPF 30) without sodium fluorescein and a combination of the Polyamide-3 and Acrylates/Octylacrylamide copolymer film formers (PolyamAcryl). All sunscreen formulations used in this study are listed in Table 1.

2.3 | Spray applicator

The spray applicator, which is schematically shown in Figure 1A, contained a tank filled with the sunscreen that was pumped by three

TABLE 1 Sunscreen formulations used in the study with corresponding contents, reference SPF value, and applied measurement methods

Formulation	Film former	Sodium fluorescein	Ref. SPF	LSM	Inhomogeneity factor	SD% transmittance
PolyamFI	Polyamide-3	0.2%	15	x		
AcryFI	Acrylates/Octylacrylamide copolymer	0.2%	15	x		
PolyesterFI	Polyester-10 (30%) and Propylene glycol dibenzoate	0.2%	15	x		
PolyamAcryl	Polyamide-3 and Acrylates/Octylacrylamide copolymer	–	30		x	x

**FIGURE 1** Schematic diagram of the spray applicator with high-voltage generator, sunscreen tank and pumps, tube adapter with HV electrode connected to the spray nozzles, shutter to block the sunscreen before and after activation, and the neutral ground electrode incorporated into a handle to be grabbed by the volunteer's hand (A). Upon activation, the sunscreen will be delivered from the nozzles to the arm along the field lines, schematically shown in (B) [Colour figure can be viewed at wileyonlinelibrary.com]

peristaltic pumps (400FD, Watson-Marlow, Limited, Wilmington, MA, USA) through nozzles with a 0.2 mm diameter at a flow rate of ≈ 8 mL/min, depending on the used sunscreen. The nozzles were arranged in triangular shape with 4.5 cm edge length. The volunteer's arms were placed at a distance of 13 cm from the nozzles and were in contact with a ground electrode by holding onto with the right hand. Initially, the area between the nozzles and the arm was blocked by a pneumatic shutter. Upon activation, an electrode located inside a tube adapter in front of the nozzles delivered a high voltage of 35 kV and the shutter opened for a set time interval of 2–4 seconds. This allowed the sunscreen to be delivered from the nozzles to the arms along the field lines describing the built-up electric field between the nozzles and the body. This is schematically shown in Figure 1B. The spray applicator covered a lateral distance of ≈ 16 cm on the volar forearm, which was assumed to be nearly circular and would correspond to an area of ≈ 200 cm². Depending on the sunscreen, an applied stable amount of ≈ 2 mg/cm² was achieved by varying the time period and the flow rate. The spray applicator was in conformity with the EN 60204-1:2006, EN 61000-3-2:2014, EN 61000-3-3:2013, EN 610006-2:2005, and EN 61000-6-4:2007 + A1:2011 standards.

For comparison, the same sunscreen was applied by hand at 2 mg/cm² on the left volar forearm of the same volunteers, as explained in section 2.1.

2.4 | Laser scanning microscopy (LSM)

In order to evaluate the lateral distribution of the sunscreens, *in vivo* laser scanning microscopy using a VivaScope 1500 (Lucid Inc) excited at 488 nm was performed to visualize the fluorescein contained in the sunscreens PolyamFI, AcryFI, and PolyesterFI. In order to evaluate an autofluorescence effect of skin or the sunscreen without fluorescein, measurements were also performed on untreated skin. The applied confocal laser scanning microscope is described in detail elsewhere.²²

Overview images of 4×4 mm² size were recorded at the skin surface and at 20 μ m depth. Further, depth scans of 500×500 μ m² image size were recorded from the skin surface down to 80 μ m depth at 1.5 μ m increments.

2.5 | Inhomogeneity factor by tape stripping

The inhomogeneity factor by tape stripping^{10,20} was determined for volunteers treated with sunscreen PolyamAcryl without fluorescein.

The calculation of the inhomogeneity factor is based on the finding that the initial distribution of the sunscreen on a tape strip is an approximate copy of the distribution of the sunscreen on the skin.^{10,20}

However, the distribution on the tapes is not stable, but will homogenize over time due to diffusion processes in the glue of the adhesive tape. Therefore, an absorption measurement has to be performed directly after the tape stripping procedure and after solving the tape in ethanol, entailing a completely homogenized solution of the sunscreen.

This was achieved by removing the superficial corneocytes of the skin applied with the sunscreen by a single $50 \times 19 \text{ mm}^2$ tape strip (tesa film No. 5529, Beiersdorf) 30 minutes after the application.²³ The tape was pressed to the skin with 14.2 bar for three seconds and removed in a constant motion. The absorption of the removed corneocytes with applied sunscreen was measured directly within 15 seconds after the tape removal from the skin, using UV/

VIS spectroscopy with an integrating sphere (Lambda 650s, Perkin Elmer) at the absorption peak in the 308–312 nm range. After solving in 6.46 mL ethanol (Uvasol, Merck), the solution was placed for 10 minutes in ultrasonic bath (Super RK102 H, Sonorex, Bandelin electronic GmbH & Co. KG) and subsequently centrifuged for 10 minutes at 1920 g (Universal 320R, Andreas Hettich GmbH & Co. KG). The solution was pipetted into quartz cuvettes with $d = 1 \text{ mm}$ thickness (Hellma) and the measured absorption was multiplied by 10 to match standard 10 mm cuvette thickness. The solution was then measured again using UV/VIS spectroscopy, and the inhomogeneity factor was calculated as the ratio between the direct tape extinction and the extinction of the solution.^{10,20} The baselines of

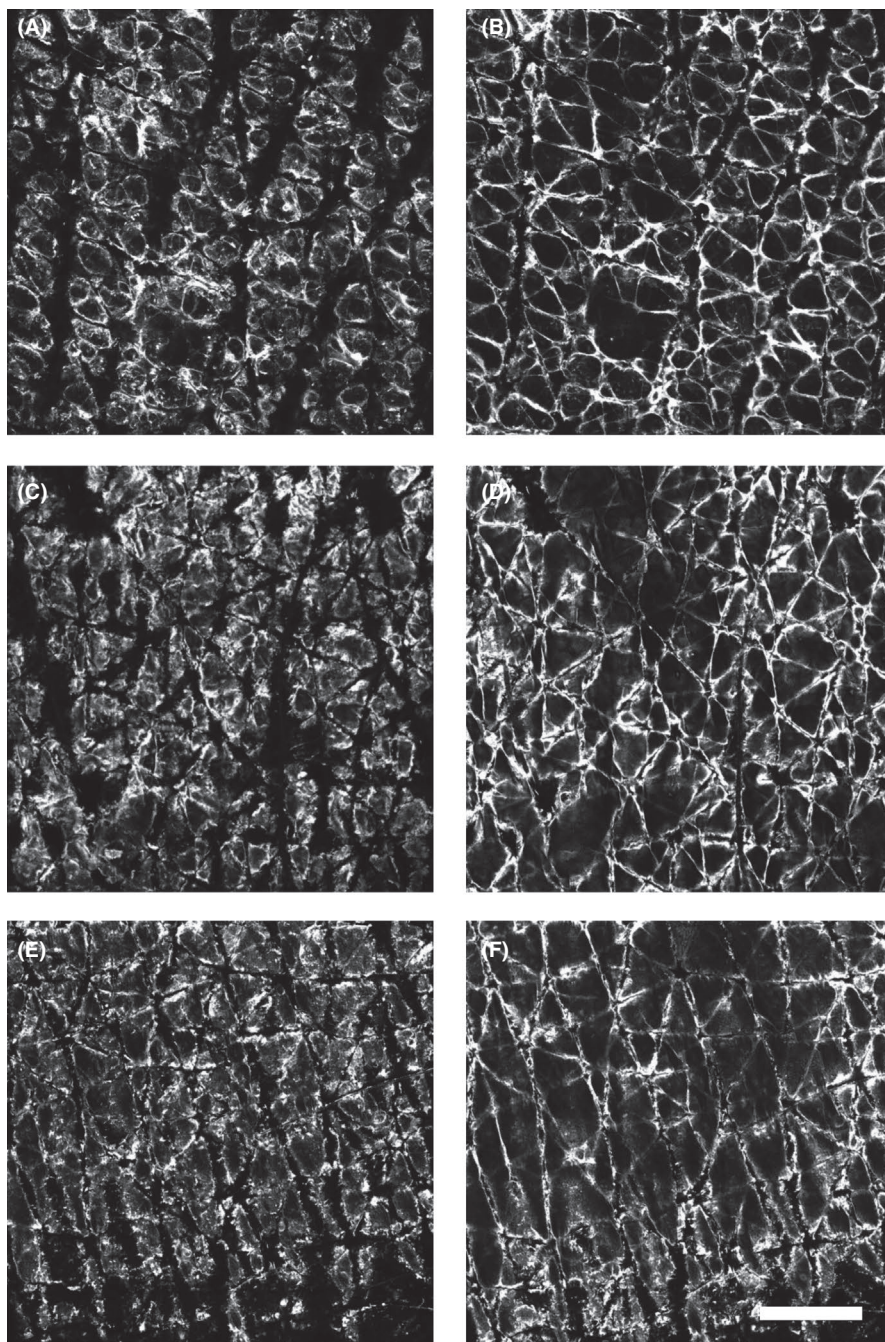


FIGURE 2 $4 \times 4 \text{ mm}^2$ mosaic images from the volar forearm in vivo, 30 min after topical treatment with sunscreen PolyamFI, (A, B), AcrylFI (C, D), and PolyesterFI (E, F) on the skin surface (A, C, E) and at $20 \mu\text{m}$ depth (B, D, F). Scale bar: 1 mm. The bright signal represents the presence of sodium fluorescein

both measurements were corrected by subtracting spectra of blank tape and pure ethanol in a glass cuvette, respectively. An inhomogeneity factor of 1 would imply a totally homogenous distribution, higher values indicate a decreased homogeneity.

2.6 | Inhomogeneity determined by in vivo SPF measurements

The SPF is commonly determined in vivo by irradiating the skin at erythral doses, which is an invasive measurement. Recently, a method to non-invasively determine the SPF in vivo has been established by measuring the diffuse reflectance signal from the skin in the 290-400 nm wavenumber range and calculating a transmittance signal²⁴ after illuminating with a xenon arc lamp. Then, the SPF is calculated from the wavelength-dependent transmittance, as well as the weighting functions of the sun spectrum and the erythema action spectrum. In this study, the calculation could be similarly achieved by illuminating with a single LED at 310 nm center wavelength²⁵ a dose below the maximal erythema dose of 30 J/m², which is the limit according to the European guideline 2006/25/EG, and matches 20% of the minimal erythema dose for skin type I according to Fitzpatrick.²¹ Although the device is not calibrated,

leading to decreased SPF values from those claimed by the manufacturer of the sunscreens, the method is well suited to determine the distribution homogeneity. If accurate SPF values are requested, the whole UV range must be considered as shown by Rohr et al.²⁶ or Reble et al.²⁴

The measurements were performed with a fiber bundle consisting of seven 100 μm excitation and multiple detection fibers at 4 seconds acquisition time before and 30 minutes after the sunscreen application. The diffuse reflectance signal was recorded, and the skin transmittance was calculated. Subtraction of the diffuse reflectance signal before the application from the signal after the application allowed to measure the extinction of the sunscreen twice, on the light path into, and out of the skin. The inhomogeneity of the distribution of the sunscreens was determined by the standard deviation of 20-30 subsequent measurements of the SPF by diffuse reflectance measurements in an area of 16 cm².²⁵ For each measurement, the SPF sensor was displaced by several millimeters within the applied skin area.

2.7 | Data analysis

All data analysis was performed in Matlab R2016a (The MathWorks Inc). As the data were not normally distributed, which was tested

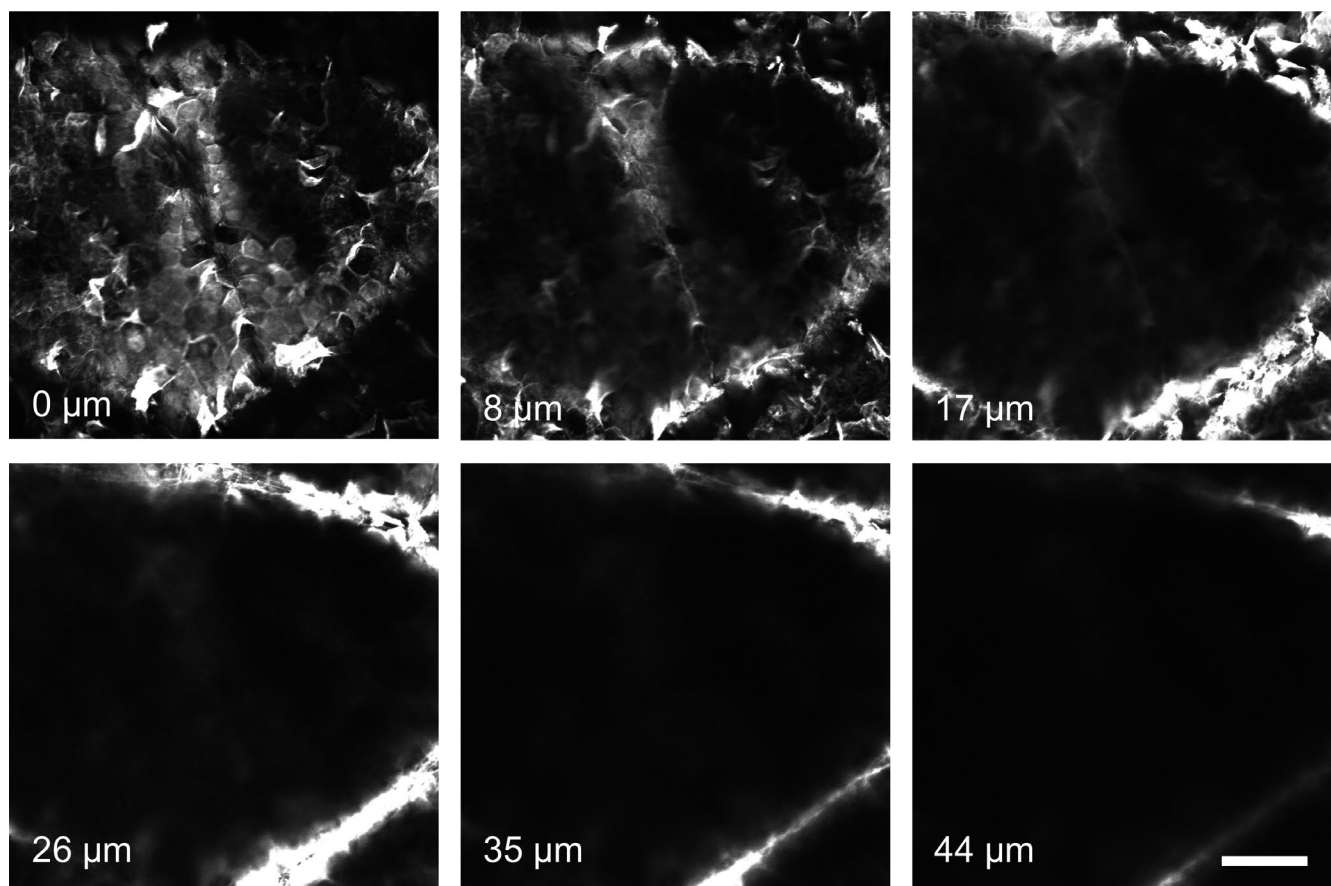


FIGURE 3 Depth scans of the volar forearm in vivo at depths 0 (1.7 mW), 8 (2 mW), 17 (3.2 mW), 26 (4.8 mW), 35 (5.1 mW), and 44 μm (5.1 mW) from the skin surface, 30 min after treatment with sunscreen PolyamFI containing polyamide as film former, excited at 488 nm, image size 500 \times 500 μm^2 , scale bar 100 μm . The bright signal represents the presence of the added fluorescence dye sodium fluorescein

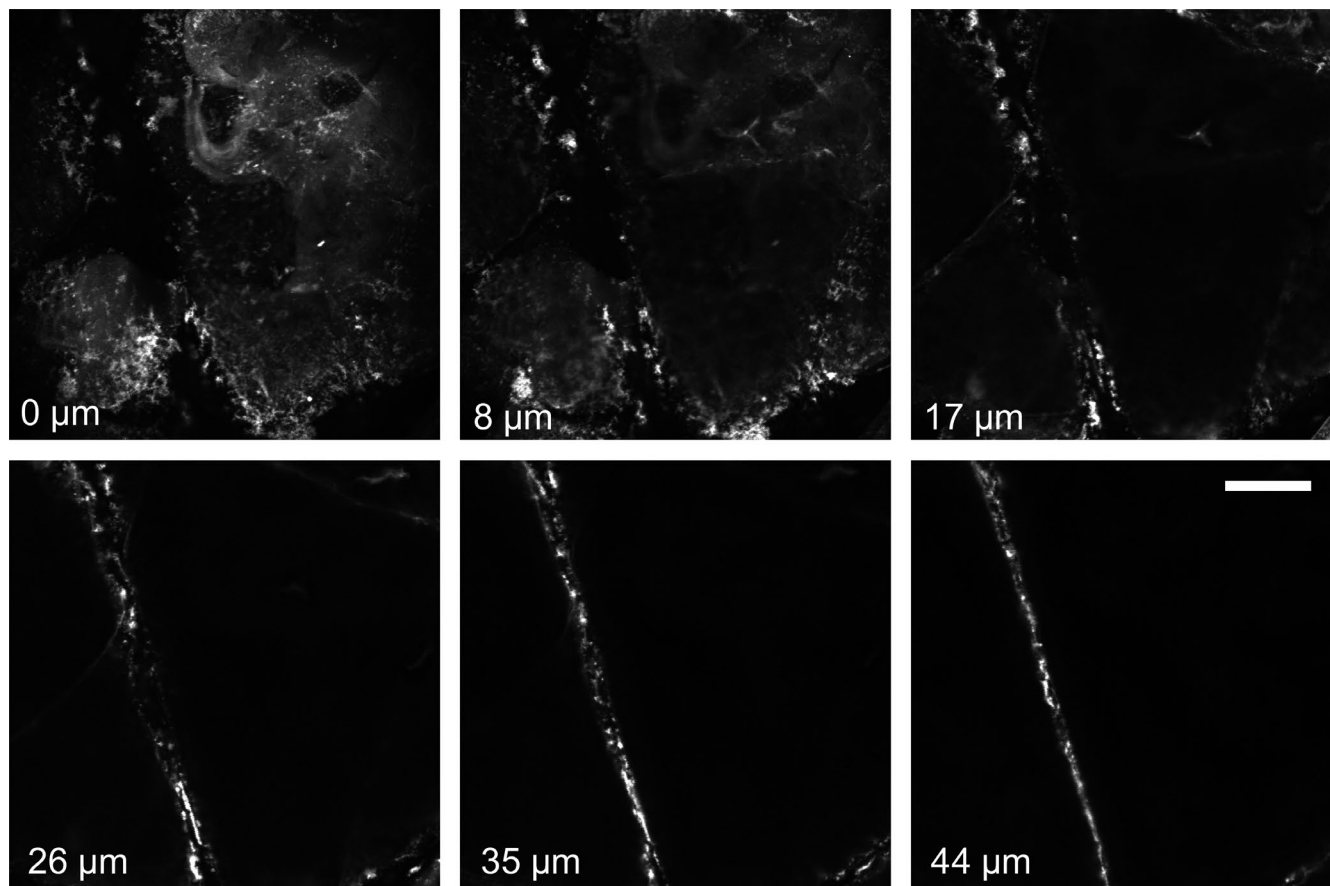


FIGURE 4 Depth scans of the volar forearm in vivo at depths 0, 8, 17, 26, 35, and 44 μm from the skin surface (5.11 mW), 30 min after treatment with sunscreen AcrylFI containing acrylate as film former, excited at 488 nm, image size $500 \times 500 \mu\text{m}^2$, scale bar 100 μm . The bright signal represents the presence of the added fluorescence dye sodium fluorescein

using a Jarque-Bera test, non-parametric Wilcoxon signed-rank tests on related data were performed in order to determine significance of mean differences. A p -value $< .05$ was considered to indicate significant difference.

3 | RESULTS AND DISCUSSION

3.1 | Distribution of sunscreens by Laser scanning microscopy

The purpose of the LSM measurements was to qualitatively evaluate the three fluorescein containing sunscreens regarding their general film-forming behaviors, homogeneity, and penetration when applied by the spray applicator on the skin.

3.1.1 | Mosaics

Figure 2 shows mosaic horizontal images of a $4 \times 4 \text{ mm}^2$ area on the volar forearm, 30 minutes after topical treatment with sunscreen PolyamFI (a–b), sunscreen AcrylFI (c–d), and PolyesterFI (e–f) on

the skin surface (a,c,e) and at 20 μm depth (b,d,f). The bright signal represents the presence of the added fluorescence dye sodium fluorescein. In Figure 2A,C,E, the corneocytes are covered with the fluorescent sunscreen dependent on the section plane of the optical cut. The sunscreen distribution on the skin surface appears more homogenous, while it is present mainly in the furrows, at 20 μm depth in Figure 2B,D,F. From a first visual appearance, sunscreen PolyamFI, containing polyamide as a film former, is most homogenous, while sunscreen AcrylFI, containing acrylates, exposes more inhomogeneous areas on a scale $\geq 500 \mu\text{m}$. Sunscreen PolyesterFI exposes aggregates of high fluorescence intensity of $\leq 250 \mu\text{m}$ size.

In order to exclude an effect of autofluorescence of the skin, the identical measurements were performed on untreated skin. These LSM measurements show only a very weak intensity autofluorescence (data shown in supporting information, Figure S1), which cannot be mistaken by the effect of the sodium fluorescein containing sunscreen. The autofluorescence recorded at 658 and 785 nm excitation was even weaker (data not shown).

In order to investigate a fluorescence of the sunscreen itself, measurements on a sunscreen formulation identical to PolyamFI, but without sodium fluorescein, were performed. These measurements

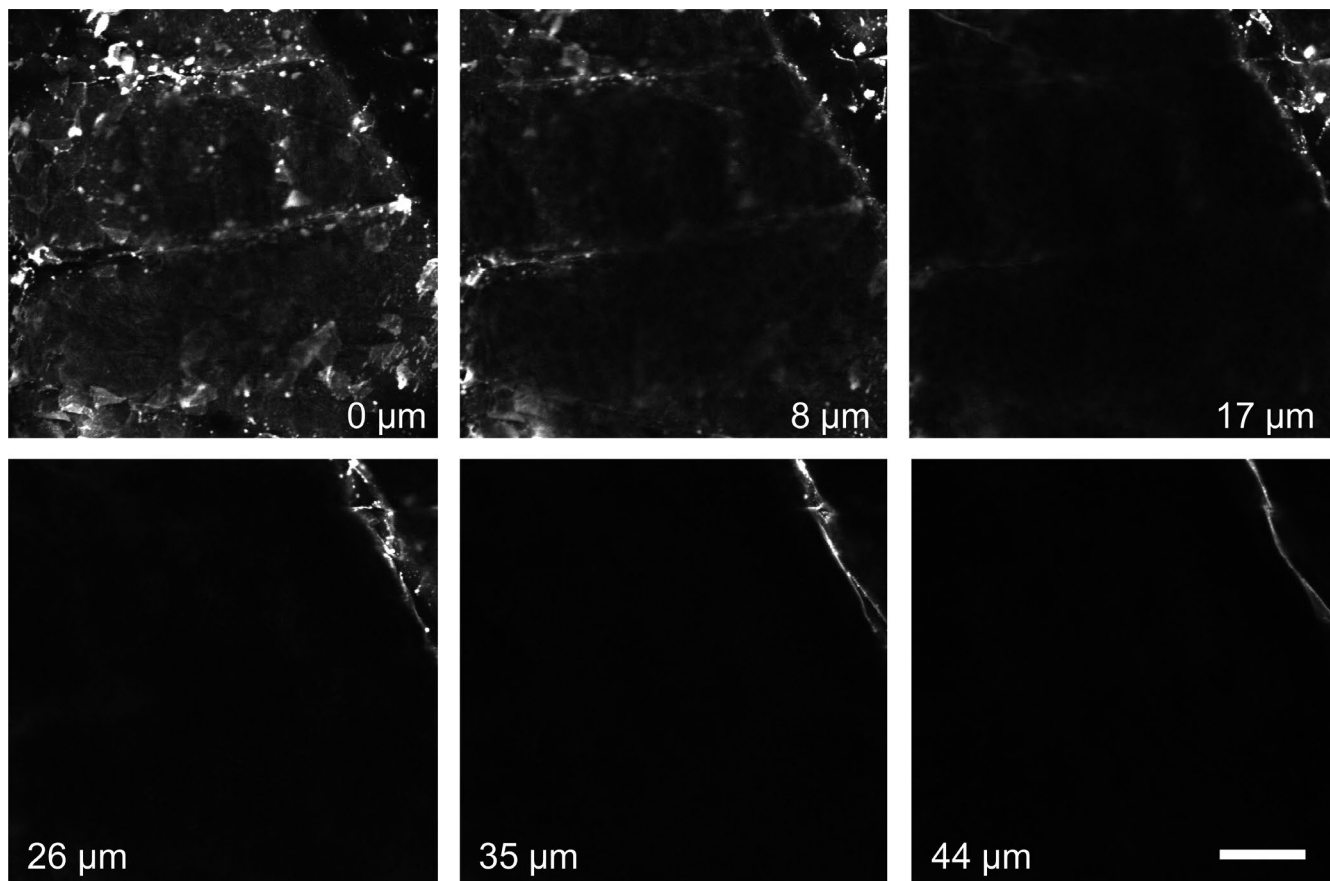


FIGURE 5 Depth scans of the volar forearm in vivo at depths 0, 8, 17, 26, 35, and 44 μm from the skin surface (5.11 mW), 30 min after treatment with sunscreen PolyesterFI containing Polyester-10 and Propylene glycol dibenzoate as film former, excited at 488 nm, image size $500 \times 500 \mu\text{m}^2$, scale bar 100 μm . The bright signal represents the presence of the added fluorescence dye sodium fluorescein

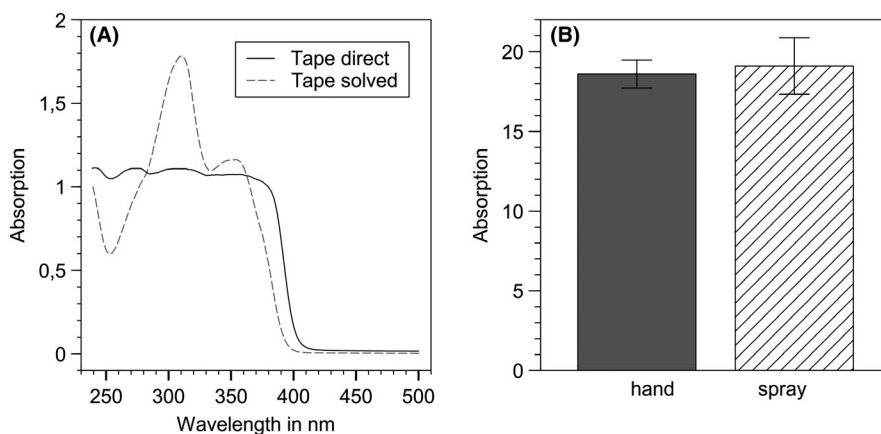


FIGURE 6 Exemplary absorption spectra of a tape strip directly (dark solid line) and the same tape solved in ethanol measured in a $d = 1 \text{ mm}$ cuvette (light dotted line) from one volunteer on the volar forearm using UV-VIS spectroscopy (A). Mean \pm standard error of absorption peaks at 310 nm after hand application ($2 \text{ mg}/\text{cm}^2$) and with the spray applicator after solving in ethanol ($n = 16$). The mean differences are not significant ($P = .796$) (B). All measurements were performed 30 min after application with sunscreen PolyamAcryl

showed only a very weak autofluorescence, comparable with untreated skin (data not shown).

For more detailed information, depth scans with higher spatial resolution and at multiple depths were recorded on selected sub-regions.

3.1.2 | Depth scans

Figure 3 shows a depth scan from a single measurement position of $500 \times 500 \mu\text{m}^2$ size, acquired 30 minutes after application with sunscreen PolyamFI, containing polyamide-3 as a film former.

Respectively, Figure 4 shows a depth scan acquired after application with sunscreen AcrylFI, containing acrylate and Figure 5 a depth scan after application of sunscreen PolyesterFI, containing Polyester-10 and Propylene glycol dibenzoate as film formers. The images were recorded at 0, 8, 17, 26, 35, and 44 μm depth in the skin. LSM images of deeper skin depths are not shown, as they did not display any fluorescence, except in the furrows. The single images of the depth scans allow to assess the distribution on a microscopic scale of smaller areas compared to the mosaics shown in Figure 2. In all three cases, the fluorescence is only visible down to a depth of $\approx 17 \mu\text{m}$. However, the furrows are completely filled with the sunscreens which indicate a comprehensive distribution of the formulations.

Sunscreen PolyamFI (Figure 3, 0 μm) shows a relatively higher homogeneity than sunscreen AcrylFI (Figure 4, 0 μm) and sunscreen PolyesterFI (Figure 5, 0 μm) on the skin surface. The fluorescence intensity at the edges of the furrows is increased, which is related to the location of the sunscreen in the furrows, but not to penetration into the corneocytes. Sunscreen AcrylFI exposes agglomerations on a scale of max. 5 μm diameter, which are located mostly in the furrows. The agglomerations of sunscreen PolyesterFI are in the range of 15 μm and can also be found outside of the furrows. The LSM results show the homogeneity of the applied sunscreens qualitatively. Considering an average stratum corneum thickness of $>17 \mu\text{m}$,²⁷ it can also be concluded that the sunscreens do not penetrate through the stratum corneum, which is anticipated in order to provide a superficial protection over longer time periods. However, the homogeneity behavior of sunscreens can differ, if no sodium fluorescein is added as a marker. Especially the penetration characteristics could be influenced by the markers.

In the LSM experiments, the film formers Polyamide-3 (sunscreen PolyamFI) and Acrylates/Octylacrylamide (sunscreen AcrylFI) both appeared to provide a homogenous sunscreen formulation. Both film formers can be used for different purposes. While the acrylate acts as an adhesive medium to provide contact of the

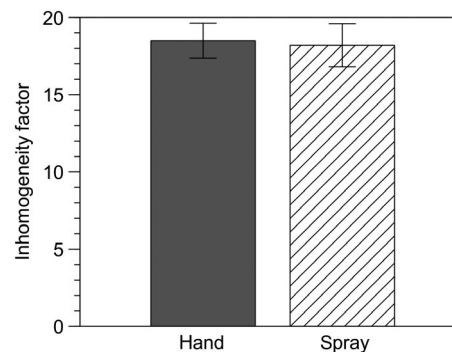


FIGURE 7 Inhomogeneity factor determined 30 min after hand application ($2 \text{ mg}/\text{cm}^2$) and with the spray applicator using sunscreen PolyamAcryl, by tape stripping. The diagrams show the mean \pm standard error. The inhomogeneity factor was determined by measuring the tapes directly and after solving in ethanol using UV-VIS spectroscopy at 310 nm ($n = 16$). The mean differences are not significant ($P = .608$)

sunscreen to the skin, polyamide enhances the distribution. This is reflected by the higher homogenous distribution of PolyamFI on the corneocytes at the surface images of Figure 2A and the good overall distribution of AcrylFI at both depths of Figure 2C,D. Therefore, a combination of these two film formers was used in sunscreen PolyamAcryl without adding fluorescein for the subsequent measurements. The Polyester-10 and Propylene glycol dibenzoate film formers (sunscreen PolyesterFI) resulted in reduced homogeneity and was not used in the sunscreen formulations for the subsequent measurements.

A drawback in using the laser scanning microscopy method for analyzing the distribution of the sunscreens is that the applied fluorophore sodium fluorescein is a hydrophilic dye, and therefore, the distribution of the fluorophore must not represent the distribution of the UV filters which are mainly lipophilic. Further, the LSM images provide qualitative information about the distribution of the sunscreens. In order to quantitatively determine the homogeneity and

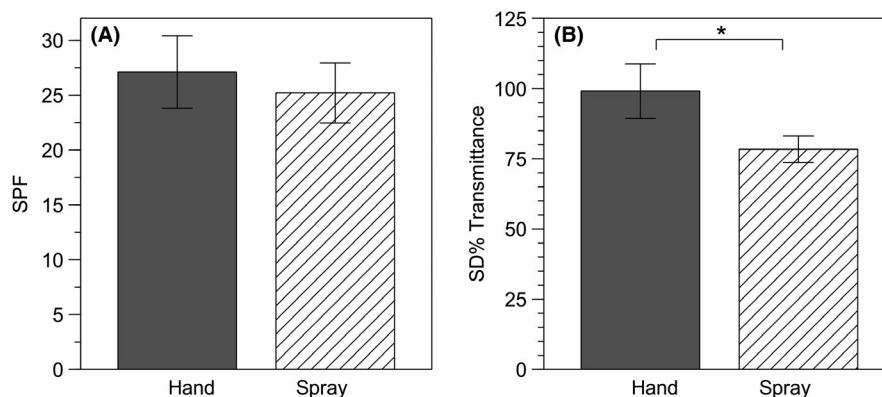


FIGURE 8 SPF determined 30 min after hand application ($2 \text{ mg}/\text{cm}^2$) and with the spray applicator showing no significant mean differences ($P = .717$) (A). Standard deviation of the transmittance. The application by the spray system entails a 20.9% lower standard deviation of the transmittance, which is significant ($P = .044$). This implies a higher homogeneity by the spraying system (B). All measurements were performed by in vivo diffuse reflectance measurements at 310 nm using sunscreen PolyamAcryl ($n = 16$). The diagrams show the mean \pm standard error

compare it with a standard application by hand using 2 mg/cm², the subsequent measurements were performed.

3.2 | Inhomogeneity factor by tape stripping

Figure 6A shows exemplary absorption spectra of the directly stripped tape (within 15 seconds after removal) and the tape solved in ethanol measured in a $d = 1$ mm cuvette. The absorbance of the directly measured and the solved tape does not clearly correlate, which is due to the difference in homogeneity. Reduced homogeneity exposes regions of reduced sunscreen concentration, which reduces the absorbance.²⁰ This can be particularly seen in the difference of the absorbance peak at 310 nm.

For the calculation of the inhomogeneity factor, the ratio of the absorption peak intensity at 310 nm between the tape directly and ten times the solved tape in ethanol was calculated.

Although the absorption of the two application methods differs for individual volunteers (data for every individual subject are shown in the supporting information in Table S1), the mean of the 16 volunteers is comparable at 18.6 for hand, and 19.1 for spray application and shows no significant mean differences ($P = .796$), as shown in Figure 6B. This means that the applied amount of sunscreen is nearly identical using both methods, which is a prerequisite for the comparison of the determined inhomogeneity.

The absorption values vary more for the spray application, which could be explained by physiologic differences, such as arm dimensions, moisture, and surface conductance.

The inhomogeneity factor (Figure 7) as determined by tape stripping describes the microscopic distribution. It is strongly dependent on the applied amount of a single measurement and therefore prone to artifacts. No significant mean differences were found ($P = .608$). This illustrates that the application with the electrostatic spray applicator without subsequent hand rubbing is comparable with the hand application done by trained technicians.

3.3 | Inhomogeneity by in vivo SPF determination

The SPF determined by in vivo diffuse reflectance measurements was 25.2 for the spray and 27.1 for the hand application, as shown in Figure 8A. This mean difference is not significant ($P = .717$), but could be explained by a slightly lower amount of applied cream for the spray application, as determined by the lower absorption. The manufacturer claimed a reference SPF of 30. The applied SPF device measuring at one wavelength (310 nm)²⁵ is not calibrated to the standard method^{14,16} and provides slightly lower values, which can explain the difference to the reference SPF value claimed by the manufacturer. However, this is irrelevant for the performed investigations of the distribution inhomogeneity in order to compare both application methods.

The intra-individual standard deviation of the measured transmittance used to determine the SPF (Figure 8B) can also serve as a measure of homogeneity. In this case, it is based on the subsequent

positioning (≈ 5 mm) of the fiber probe on nearby skin sites and, therefore, describes the inhomogeneity on a larger scale. The standard deviation of the transmittance is significantly lower for the spraying system by 20.9% ($P = .044$) than for the hand application.

So far, all data for SPF deviation resulted from inter-individual variation because the whole area of applied sunscreen was irradiated and analyzed as a whole. The new non-invasive SPF sensor enables to analyze the distribution within this area by multiple measurements across this area. Thereby, this technology offers the analysis of the sunscreen distribution on the skin directly, for the first time. Therefore, the significant differences shown in this explorative proof of principle study are meaningful, although in vivo SPF measurements show inherent variability and increased amount of volunteers would be necessary in follow-up studies.

It should further be noted that the hand application of 2 mg/cm² was conducted under standardized laboratory conditions.^{14,16} In real-life conditions, the amount of applied sunscreen is expected to be lower.⁴ Therefore, the standard deviation is expected to be higher in real-life application, entailing a lower homogeneity. The high standard deviation after hand application has been described several times.^{3,28} This is also dependent on the skin of the different volunteers.

4 | CONCLUSION

The new spray applicator based on electrostatic charge in combination with selected film formers shows very good microscopic distribution on the surface of the corneocytes and in furrows and wrinkles investigated by LSM. The quantitative analysis revealed a comparable inhomogeneity factor using absorption measurements of tapes taken from the surface of the applied skin site, and SPF values measured by a non-invasive SPF sensor based on diffuse reflectance measurements in vivo compared to standardized hand application under laboratory conditions. Furthermore, the new in vivo non-invasive SPF measurement method enabled measurements of the sunscreen distribution on the skin by the standard deviation of multiple transmittance measurements, where the sensor was spatially displaced. These results indicate a superior distribution using the spray applicator over the standardized hand application. This demonstrated that the electrostatic spray application is a promising tool to apply the sunscreen more evenly on the skin independent of the user.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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