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Facile development of graphene-based air filters mounted on a 3D printed mask for COVID-19

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ABSTRACT

COVID-19 belongs to a typical class of viruses that predominantly affects the human respiratory system, thereby proving to be fatal to many. The virus, along with other air pollutant particulates poses a severe threat to the human respiratory organs. Since the most common transmission mode is respiratory fomites and aerosol particulates, it is necessary to prevent their ingestion through a mask. The primary use of masks is to prevent aerial particulates. This paper reveals the development of masks with air filters coated with functionalized graphene (fG) mounted on a 3D-printed facial mask replica. The fused deposition modeling (FDM) process is used for fabricating the facial mask replica. fG associated with nanosheets has an additional adsorbing capacity with a high surface area to volume ratio. fG coat is used over a polypropylene (PP) cloth through a dip coating method to enhance the antiviral and antimicrobial properties. The quality of fG was investigated through Raman spectroscopy and other characterization techniques such as SEM, XRD, and FTIR, which were used for visual interpretation of distributions of fG coated with nanosheets. The filtration efficiency of the fabricated mask was tested against SARS-CoV-2 viral particles, which showed a complete arrest of viral transmission at the fG coated layer.

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1. Introduction

Coronavirus is typically an airborne virus that has manifested itself as a severe acute respiratory syndrome (SARS) causing fatal respiratory infections and has been declared as a global health emergency by World Health Organization (WHO). The SARS CoV-2 is suspected of having its epicenter in the Hunan seafood market in the Wuhan region of China and spread globally. Infected person ejects the virions as respiratory fomites (cough & sneezes), and since viruses can sustain themselves over any surface for a significant period, people could be infected either by physical contact with an infected one or surface. The process of virus-host cell interaction proceeds in four steps, viz. (i) Attachment of virus spike protein with host cell receptors; (ii) endocytosis; (iii) replication; (iv) exocytosis [1]. The coronavirus pathogenesis can be described briefly as the ingestion of the virus, the propagation of viral genetic material into a host cell, the cellular immune response of host cell, and the external transmission of virions as aerosol dispersed particulates.

To avert the spreading of the coronavirus and other airborne particulates culpable for respiratory diseases, N95 masks are used extensively by medical practitioners and commoners as personal protective equipment (PPE). The estimated size of the SARS causing coronavirus is about 0.08–0.14 μm in diameter [2]. The N95 mask has the lowest protection factor value for these micro-organisms [3], albeit that N95 masks usually exhibit up to 95%
bacterial filtration efficiency (BFE). However, it depends on various factors such as the relative humidity (RH), the surrounding temperature, active material for air filters, facial fitting, methods of decontamination, and reusability frequency. N95 masks consist of multi-layered fabrics that ensure an effective shielding from the pollutant particles using an electrostatic force field induced in the fiber. Since COVID-19 virions are positively charged species, hence electrets material fibers are irreplaceable.

Variable-sized aerosol dispersed particles adhere to the fabrics by electrostatic action as well as van der Waals interaction. Particles with a diameter >1 μm are considerably large particulates, ~0.3 μm are medium-sized, and <0.1 μm are classified as small-sized particulates. The corona virions are characterized as small-sized particulates exhibiting Brownian motion, as shown by the yellow path in Schematic 1, as its ingression trajectory. Extensive research is going on regarding the enhancement of efficacy in the field of PPEs, most specifically on protective facial equipment. N95 masks are accepted widely for electret-fibers necessary to avoid viral ingress. Cellulosic fiber filters with the addition of poly(ethylenimine) (PEI) gives them an antiviral property [4], polypropylene microfibers with diameters in the range of ~1–10 μm [5], and nanoparticles of silver and copper are also introduced for an enhancement in antimicrobial properties. Activated carbon-based masks are also a potent material for air filters.

Similarly, functionalized graphene (fG) has been a competitive material for air filters and can be used to modify N95 masks. fG & its derivatives have gained popularity for their antimicrobial and antiviral properties, high surface-to-volume ratios, unique physicochemical properties and biocompatibility [6–10]. It is a two-dimensional (2D) monolayer sheet of sp²-hybridized carbon atoms arranged in a hexagonal honeycomb-like lattice structure with a thickness of 1.84 nm [11]. The negative charge and unique nanosheet structure of fG play a crucial role in the antiviral activity for virus destruction and inactivation [12]. Negatively charged fG interacts with the positive-sensed viruses via hydrogen bonding, electrostatic interactions, and redox reaction [13,14]. The surface-bound virus gets adsorbed on the fG and can then be washed off. Structural integrity loss (externated envelope and crown) is an outcome of physicochemical interaction of virus and functionally tailored graphene nanosheets leading to inhibition in viral functions. Long exposure time and concentration contribute to enhanced antiviral action of graphene [15].

Furthermore, the efficacy of a mask depends on the perfectness of placement of the facepiece in the proper position (facial fit) so that the chances of ingressing virions and releasing fomites are significantly minimized. Facial masks can be fabricated as a customized item using 3D printing technology. 3D printing of polymeric materials, such as PLA, is an efficient approach to fabricating customizable mass products for a better facial fit and enhancing protection.

Herein, we report a facile development of a graphene-based air filter mounted on a 3D printed mask for COVID-19.

2. Experimental

2.1. Synthesis of functionalized graphene

fG was obtained using Hummer’s method by oxidizing purchased graphene platelets, as shown in Schematic 2. Firstly, sodium nitrate (1.5 gm) was added to a concentrated sulphuric acid (70 ml). To this solution, graphene platelets (3 gm) were added with vigorous stirring, and a black color colloidal mixture was obtained. After this, using an ice bath, the mixture was cooled to 5 °C. Once the mixture was cooled, potassium permanganate (9 gm) was introduced in the mixture with proper care and patience. After this, 140 ml of deionized (DI) water was slowly added, which further Results in an exothermic reaction raising the temperature to 98 °C and maintained at this temperature for 15 min. Subsequently, the external heating was removed, and the colloidal was allowed to cool down to room temperature. Additional DI water (420 ml) was introduced into the mixture, followed by hydrogen peroxide (3 ml) to stop the reaction. This reaction was left untouched for one day so that the fG settles down, which was then washed using ethanol and

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**Schematic 1.** Interaction of airborne virions and pollutant particulates with the electrostatically charged fibers of a graphene mask.
DI water several times. The washing process was further discussed in the supporting information (SI).

2.2. Fabrication of 3-D printed mask using additive manufacturing

A customized design of the fG mask has been generated using a 3D-CAD model platform (Rhinoceros software). The design was then converted into a meshed model (.stl format). This file was then transferred to Cura (open-source software provided by Ultimaker) for slicing. The slicing was configured for the Ultimaker 2+ 3D printer. Polylactic acid (PLA) in the form of filament was used as the base material for the mask prototype with a printing speed of 60 mm s⁻¹. No support structure was required while a brim was provided for build plate adhesion. The sliced design was converted into g-code format and transferred to the printer. The build platform was preheated to 70 °C, while the nozzle (diameter 0.4 mm) was heated to 200 °C. All the printed parts were then assembled to make the mask prototype. Schematic 3 is a graphical representation of the steps involved in this process.

2.3. Fabrication of fG filter

The dip-coating method was used to coat fG over the polypropylene (PP) fabric cloth. A mixture of 9:1 ratio of fG and polyvinyl alcohol (PVA) was mixed in N-methyl-2-pyrrolidone (NMP). The mixture was stirred for a few hours to prepare a homogenous dispersion. After this, the fabric cloth was dipped into the solution for 30 s. The dipped cloth was taken out and dried at 50–60 °C for 24 h. This process was repeated two times. The developed air filter is a stack of four layers, as depicted in Fig. 1. Outer layers are fixed with 20 μm pore-sized PP fabric for each configuration (schematic S1). The middle one is changed with 20 and 10 μm pore-sized PP fabric, and 3 μm melt-blown (MB) and are referred to as fG coated 20 μm filter, fG coated 10 μm filter, and fG coated melt-blown filter for further designation, respectively. The use of multilayer stacking helps in stopping the penetration of droplets. In this configuration, the first layer is used to absorb micro-aerosol droplets, and the middle layer is used to prevent micro-organism and small particulates. On the other side, the third layer is specifically used to absorb fluids (during sneezing) from the mask wearer.

The 3D printed parts are shown in Fig. 2a. The filter is trapped between the filter holders and is screwed to the main body. The completed mask with the fG filter after assembling is shown in Fig. 2b. This mask can now be used to stop the spreading of droplets coming out from the mouth during sneezing or coughing. A 3D printed mask can be easily customized using simple software and thus gives a perfect fit. When a typical surgical mask or N95 masks are used multiple times, they might suffer from the masks’ wear and tear, but no such problems occur with a 3D printed mask. Another advantage of the printed mask over the surgical masks is

Schematic 2. Various steps involved for the synthesis of fG.

Schematic 3. A schematic representation for a) 3D-CAD model, b) slicing into g-code, c) 3D printer setup, and d) 3D printed mask prototype.

Fig. 1. Stacking order of PP cloth for the fabrication of a fG coated mask.
that it can be sanitized easily using spray sanitizers while the fabric masks require extensive washing before reusing.

On the other hand, the printed mask requires a comparatively far less amount of fG coated cloth and it provides a perfect fit to cover the leak from the nasal bridge and can be configured according to an individual. Masks have been the most problematic for people using spectacles. As the usual face masks are not air-tight near the nasal bridge region, the face fogs of the spectacles’ glasses, but no such problem is observed when a 3D printed mask is used. The washability and reusability of the printed mask are also a significant advantage over the other masks as only the filter needs to be changed instead of the complete mask, which reduces the waste and disposing of the used mask may contain a virus.

3. Results and discussion

In order to investigate the presence of planes and functional groups of fG, X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FT-IR) was used. The XRD pattern was recorded in the 2-theta range of 20°—60° with 0.02° per second step size. In Fig. 3a, the broad diffraction peak (002) can be seen at 26.10° with an interlayer distance of 3.39 Å. Hence, it is confirmed that the synthesized material is composed of well-ordered graphene platelets. The FT-IR data, Fig. 3b, was recorded between 500 and 4000 cm⁻¹ wavenumbers. fG shows a peak at 1764 cm⁻¹ due to the characteristic band of C=O stretching of the –COOH group. The presence of bands associated with –C=O (at 1183 cm⁻¹), –C=O–H (at 1277 cm⁻¹), –C=OH (at 1462 cm⁻¹), –C=O (at 1665 cm⁻¹), and a broad peak at 3663 cm⁻¹ corresponding to –O–H vibration was also observed in the FT-IR spectrum. The FT-IR results suggest the presence of various functional groups in fG.

Raman spectroscopy can provide qualitative and quantitative information on carbon-based materials to investigate various physical properties (defects, crystallite size, vacancies, and several layers, etc.) [18]. Fig. 4a represents the de-convoluted Raman spectra of fG prepared by Hummer’s method. It was recorded using a 532 nm green DPSS laser source at room temperature. Correspondingly, Fig. 4b displays the higher wavenumber Raman spectra with the same source. An intense band at 1602 cm⁻¹ shows the spectrum of a single graphite crystal. It is assigned to the E₂g degenerative vibrational mode, called as the G band. The shifting of G band to the higher wavenumber is due to the occurrence of two additional bands: D’ band (1621 cm⁻¹) and D** band (1577 cm⁻¹). They are observed due to the highly defective structure of synthesized graphene. Another small band is observed at 1346 cm⁻¹, termed as D band which corresponds to the raman mode. It is the result of either small crystallities or defects from boundaries in polycrystalline samples. The D band is attributed to two small bands due to the rich sp³ phase of the disordered nature of amorphous carbon. It has been observed that side peaks in both bands increase by the disorder, leading to the broadening of bands. As the broadness of the D band (31 cm⁻¹) is inversely related to the crystallite size (Lₐ), it represents the amorphousness in the prepared sample. In disordered carbon-based materials, the integrated intensity ratio of D and G peaks (1.15) has a noticeable relation for obtaining the sp² crystallite size (in-plane). The high intensity of the D band indicates that the sp² bonded carbon which turns into sp³ bonds in the sample. Apart from that, this intensity ratio also depends upon the excitation laser energy wavelength (λ).

\[ L_a(\text{nm}) = \left(2.4 \times 10^{-10}\right) \cdot \lambda^4 \cdot (I_D/I_G)^{-1} \]  

From Eq. (1), the crystallite size of the sample was obtained to be 16.71 nm. The second spectrum consists of four less intense characteristic peaks, namely G*, 2D, D + G, and 2D'.

The bare fabric cloth and coated fG fabric cloth were examined through scanning electron microscopy (SEM) with gold coating. Fig. 5a shows the micrograph of the bare fabric cloth in which the fibers can be seen with an average pore size of 20 μm. Fig. 5b shows the fabric cloth with the coating of fG. The fG is uniformly distributed on the fabric cloth. The presence of desired elements over the PP fabric was confirmed by mapping and energy dispersive X-ray spectroscopy (EDAX). Fig. 5(c) and (d) show the mapping and EDAX profile of fG coated PP fiber in which the distribution of carbon over the fiber can be easily seen. The presence of an undesired element (Calcium) in the EDAX profile may be attributed to external impurities.

3.1. Bacterial filtration efficiency (BFE) results of fG filter

FDA-recognized ASTM F2100-11 is the primarily known standard for the performance of materials used in the filters. ASTM F2100-11 standard was also defined for the testing of N95, KN95, FFP1, and FFP2 face masks. BFE test for fG based filters was done by Apex Testing and Research Laboratory, New Delhi. BFE test (test method- IS: 16,288:2014) was performed for the filters to evaluate the efficiency against the bacterial counts. A suspension of Staphylococcus aureus was aerosolized onto the filter at a constant flow.
rate. The challenged delivery is maintained at $2.9 \times 10^4$ to $3.9 \times 10^4$ colony-forming units with a mean particle size of $3.0 \pm 0.3$ μm. The procedure shows the reproducible bacterial challenge to be delivered to the test material. According to NIOS (United States standard), the BFE of N95 mask was observed at least 95% for airborne particles. European standard EN 143 allows P1, P2, and P3 filters with 80, 94, and 99.95% of BFE for airborne particles. Our fabricated fG coated filters show 84, 92.61, and 98.2% of BFE on 20 μm, 10 μm, and MB filters, respectively, against 3-micron sized bacteria (Table 1). The gram per square meter (GSM) of the 20 μm filter was observed to be 372. We also performed the sub-micron particulate filtration efficiency of fG coated MB filter. It was observed that the fG coated MB filter has 94.3% of sub-micron particulate filtration efficiency against 0.3-micron sized particulates. The observed Results indicate that the fG coated MB cloth shows the higher BFE efficiency than other configurations (Table 2).

As observed from the previous studies, the trapping of aerosol particles through any fiber mask can be attributed to different mechanisms, such as gravity sedimentation, inertial impaction, diffusion, and electrostatic attraction [16]. All of the mechanisms mentioned above (Schematic 4) are depending on the size of particles. It has been stated that gravity sedimentation plays an essential role for aerosols with sizes 1–10 μm as gravity forces provide an initial impact on the wide exhaled droplets. While in

Fig. 3. XRD pattern (a) and FT-IR spectrum (b) of the synthesized fG.

Fig. 4. Raman spectra of the synthesized fG.

Fig. 5. SEM images of bare fabric cloth (in the inset: high-resolution image) (a) fG coated fabric cloth (in the inset: high-resolution image) (b), mapping image of fG coated filter (c), and EDAX spectra of the fG coated filter (in the inset: the quantitative analysis of desired elements). Bacterial filtration efficiency (BFE) results of the fG filter.
the case of inertial impaction, particles with enough inertia (to avoid flowing across the fibres in the filtration layers) are stuck into the mask layer and filtered. The diffusion mechanism occurs when the particles of sizes less than 0.1 μm size are diffused and trapped in the fiber layers because of the Brownian motion carried by the porous matrix of the fiber mask. However, the electrostatic attraction mechanism differs from described mechanisms. In this mechanism, the electro-charge polymer fiber or cloth is applied as a filter that attracts oppositely charged particles and traps them.

The mechanism as mentioned above may be responsible for the high BFE of our fabricated bare MB filter. The filtration mechanism of bare MB cloth is shown in Schematic 4. In the bare MB filter case, the first layer with 20 μm ± 2 μm of pore size absorbs or prevents the penetration of large aerosol generated particulates while it allows micro-organism and small-sized particulates towards the second layer. The second layer made of MB with 3 μm ± 1 of pore size can filter small sizes of particulates and micro-organisms except for the nanosized bacteria or viruses. To overcome this issue, the middle layer (second layer) was coated with fG for the inactivation of micro-organism during the inhale/exhale process.

3.2. Breathing resistance (pressure drop)

One of the criteria for an effective face mask is that it does not make breathing difficult. A mask with very high breathing resistance can cause unwanted behavior such as taking off the mask. To ensure the capability of the mask, it is more important to know the breathing resistance of fabricated masks. Here, the breathing resistance of bare and fG coated MB filter was done according to the IS:9473:2002 test from the mask mentioned above the testing laboratory. A fan forcing air through a mask fitted to a dummy of a human head was used to measure the breathing resistance. The observed results of bare and fG coated filters at different flow rates are shown in Table 3. Notably, the coating of fG over MB cloth does not affect the breathing resistance of the mask. A lower value of breathing resistance at different flow rates indicates a better comfort level for healthy and unhealthy people. A standard pressure drop of commercialized N95 mask is 3.5 mbar at 85 L per minute, which is higher than our fabricated fG coated MB filter.

3.3. Role of fG-coated filters against SARS-CoV-2

The antimicrobial surfaces can be designed with graphene-based nanomaterials. The exact mechanism of bacterial inactivation is still a major topic to investigate. However, several efforts have been taken to identify the role of graphene in the inactivity of bacterial species. Schematic 5, shows the sharp-edge insertion, cell entrapment, and oxidative stress mechanism of graphene with bacteria. Graphene has very sharp edges that can cause physical damages to the cell membrane upon direct contact with bacterial cells. Hu. et al. [18], confirmed the sharp-edge insertion of graphene into the E. coli bacteria via transmission electron microscopy. In oxidative stress, the graphene sheets interfere with bacterial metabolism due to the imbalance between oxidation and anti-oxidation. Besides, cell entrapment is also a possible mechanism for the inactivity of bacterial species. In this mechanism, the

| Filters | Flow rate | Test Results (mbar) |
|---------|-----------|---------------------|
| Bare MB filter | a) Inhalation @ 30% min | 0.75 |
| | b) Inhalation @ 95% min | 1.10 |
| | c) Exhalation @ 160% min | 1.75 |
| fG coated MB filter | a) Inhalation @ 30% min | 0.75 |
| | b) Inhalation @ 95% min | 1.10 |
| | c) Exhalation @ 160% min | 1.54 |

The mechanism as mentioned above may be responsible for the high BFE of our fabricated bare MB filter. The filtration mechanism of bare MB cloth is shown in Schematic 4.

![Schematic 4](image.png)

Schematic 4. Filtration mechanism of bare fiber filter via gravity sedimentation (a), inertial impaction (b), diffusion (c), and electrostatic attraction (d) mechanism.

| Sr. No. | Fabric filters | BFE (%) |
|---------|----------------|---------|
| 1. | Silk cloth | -88 |
| 2. | Chiffon cloth | -73 |
| 3. | Cotton (600 threads/inch) | -82 |
| 4. | Cotton + flannel cloth | -96 |
| 5. | Surgical masks | -76 |
| 6. | Cotton + Chiffon cloth | -97 |
| 7. | Cotton + silk cloth | 94 |
| 8. | fG coated MB filter | 98.20 |

Table 2: Comparison table of BFE of the various fabric-based filters with fG coated MB filter [17].
bacterial cells are captured by graphene sheets which isolate bacterial cells from the outer environment and sufficient nutrition. For demonstrating the efficacy of the masks in arresting the transmission of SARS-CoV-2 viral particles, we used 12 anonymized nasopharyngeal swab samples collected in VTM vials (Hi-Media, India). These samples belonged to the following four categories: (i) samples with Ct (Cyclic threshold) values < 20 for ORF1ab gene, representing samples with relatively high viral load; (ii) samples with Ct values between 21 and 25, representing samples with intermediate viral load; (iii) samples with Ct values > 25, representing samples with low viral load; (iv) samples negative for COVID-19. The experiments, as shown in schematic 6, were performed using 3 samples belonging to each category.

To simulate the real-life scenario wherein the mask wearer is likely to be exposed to droplet-borne aerosols emanating from an infected person in talking, coughing or sneezing, we used an atomizing nozzle spray (EXAIR, SR102055) to generate aerosols of 16–25 μm of diameter. The spray of aerosols was allowed to be incident first on a Petri plate of 90 mm diameter for a period of 5 min. The petri plate was then flooded immediately with 1 ml of VTM, and the same was used for RNA extraction and RT-PCR to capture the viral load in the initial incident spray. The spray was then projected at the mask from a distance of 15 cm for a similar duration of 5 min. The mask layers were then carefully dismantled with the 3 layers, viz. outer layer, fG coated layer, and inner layer being placed in separate VTM vials and used independently for RNA extraction RT-PCR for determining the viral load in each of the layers. The entire experiment was conducted in a properly calibrated Biosafety cabinet class IIA2, with appropriate biosafety precautions.

RNA extraction from all the samples (incident spray and the 3 layers of the mask) was done using QIAMP viral RNA mini kit (Qiagen), as per the kit instructions. RT-PCR for detecting SARS-CoV-2 was performed with the extracted RNA, by using a COVID-SURE multiplex Real-Time RT-PCR kit (Labsystems Diagnostics; Trivitron Healthcare Pvt. Ltd.) on a CFX-96 (Bio-Rad) thermal cycler. This kit amplifies target sequences in the E and ORF1ab genes of SARS-CoV-2 and uses an RPP30 gene as an internal control. As recommended by the kit manufacturers, a sigmoidal curve with a Ct value of <36 was considered the criterion for considering a sample to be positive. All the PCR assays were run with no-template controls.

| Table 4 | Ct values of ORF 1ab gene in samples collected from incident spray and different layers of fG coated mask. |
|---------|-------------------------------------------------------------------------------------|
| Incident Spray | Outer layer | fG coated layer | Inner layer |
| Samples with Ct values < 20 | | | |
| Sample A1 | 16.47 | 21.12 | 0 | 0 |
| Sample A2 | 17.57 | 22.75 | 0 | 0 |
| Sample A3 | 19.33 | 24.38 | 0 | 0 |
| Mean ± SD | 17.79 ± 1.44 | 22.75 ± 1.63 | 0 | 0 |
| Samples with Ct values between 20–25 | | | |
| Sample B1 | 22.06 | 26.67 | 0 | 0 |
| Sample B2 | 23.43 | 27.61 | 0 | 0 |
| Sample B3 | 23.74 | 26.56 | 0 | 0 |
| Mean ± SD | 23.08 ± 0.89 | 26.95 ± 0.58 | 0 | 0 |
| Positive Sample >25 Ct Spray | | | |
| Sample C1 | 26.47 | 29.66 | 0 | 0 |
| Sample C2 | 27.53 | 29.53 | 0 | 0 |
| Sample C3 | 29.13 | 30.28 | 0 | 0 |
| Mean ± SD | 27.71 ± 1.34 | 29.82 ± 0.4 | 0 | 0 |
control, extraction control, and positive control as part of the internal quality control protocol.

To demonstrate the efficacy of the masks in arresting the passage of SARS-CoV-2 viral particles, we compared viral RNA titers in different layers of the mask when an incident spray charged with varying viral loads was projected on them. Table 4 depicts the Ct values of the individual samples in the incident spray and their corresponding values in the different layers of the mask. While the viral RNA titers were found to be similar to the same in the incident spray proximal to the graphene layer, complete arrest of viral transmission was observed at the fG coated layer in all 3 categories of samples having low, intermediate, and high viral loads. The mask’s efficacy is represented by the failure to amplify any of the target genes in samples collected from the graphene layer and the layer distal to it (Ct = 0).

The mean Ct values of the 3 biological replicates in each of the 3 categories of clinical samples are depicted in Fig. 6.

4. Conclusion

We have successfully fabricated the fG filter-based 3D-printed facial protective equipment. fG, synthesized by Hummer’s method, is characterized by using various characterization techniques. The fG filter shows 84.00%, 92.61% and 98.20% of bacterial and viral particle size range. Ann. Occup. Hyg. 52 (2008) 177–185, https://doi.org/10.1093/annhyg/meen005.

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Appendix A. Supplementary data

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