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# Review of Surface Particulate Monitoring of Dust Events Using Geostationary Satellite Remote Sensing

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#### 11 Abstract

The accurate measurements of natural and anthropogenic aerosol particulate matter (PM) is 12 13 important in managing both environmental and health risks; however, limited monitoring in regional 14 areas hinders accurate quantification. This article provides an overview of the ability of recently 15 launched geostationary earth orbit (GEO) satellites, such as GOES-R (North America) and HIMAWARI 16 (Asia and Oceania), to provide near real-time ground-level PM concentrations (GLCs). The review 17 examines the literature relating to the spatial and temporal resolution required by air quality studies, 18 the removal of cloud and surface effects, the aerosol inversion problem, and the computation of 19 ground-level concentrations rather than columnar aerosol optical depth (AOD).

20 Determining surface PM concentrations using remote sensing is complicated by differentiating 21 intrinsic aerosol properties (size, shape, composition, and quantity) from extrinsic signal intensities, 22 particularly as the number of unknown intrinsic parameters exceeds the number of known extrinsic 23 measurements. The review confirms that development of GEO satellite products has led to 24 improvements in the use of coupled products such as GEOS-CHEM, aerosol types have consolidated 25 on model species rather than prior descriptive classifications, and forward radiative transfer models 26 have led to a better understanding of predictive spectra interdependencies across different aerosol 27 types, despite fewer wavelength bands. However, it is apparent that the aerosol inversion problem 28 remains challenging because there are limited wavelength bands for characterising localised 29 mineralogy.

The review finds that the frequency of GEO satellite data exceeds the temporal resolution required for air quality studies, but the spatial resolution is too coarse for localised air quality studies. Continual monitoring necessitates using the less sensitive thermal infra-red bands, which also reduce surface absorption effects. However, given the challenges of the aerosol inversion problem and difficulties in converting columnar AOD to surface concentrations, the review identifies coupled GEO-neural networks as potentially the most viable option for improving quantification.

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- 37

Keywords: Geostationary Earth Orbiting satellites; Aerosol Optical Depth; Particulate Matter; Thermal
 infra-red; spatiotemporal resolution.

#### 41 Abbreviations:

- 42 Note in the interests of brevity, and apart from MODIS, this list of abbreviations specifically excludes
- 43 the full dispersion model and satellite names for which the commonly used abbreviation has been
- 44 used.
- 45

46 AOD: Aerosol Optical Depth

- 47 BT: Brightness temperature
- 48 BTR: Brightness temperature reduction, i.e. BT<sub>1</sub> BT<sub>2</sub> where the suffix could be time or wavelength
- 49 IDDI: Infrared Differential Dust Index, BTR but restricted to time-based differences
- 50 GEO: geostationary earth orbit satellites
- 51 GLCs: ground level concentrations
- 52 LEO: low earth orbit satellites
- 53 MODIS: MODerate-resolution Imaging Spectro-radiometer instrument
- 54 NIR: Near infra-red portion of the electromagnetic spectrum
- 55 PM: particulate matter
- 56 TIR: Thermal infra-red portion of the electromagnetic spectrum
- 57 UV: Ultra-violet portion of the electromagnetic spectrum
- 58 Vis: Visible portion of the electromagnetic spectrum

#### 59 Highlights:

- Excellent temporal resolution (10 minutes) but coarse spatial resolution (2 km);
- Continuous infrared instead of visible bands are required;
- Challenging aerosol inversion compounded by fewer and less sensitive infrared bands;
- Vertical profile required for extrapolating AOD to ground-level concentration;
- Uncertainty analysis of speciated ground-level concentration needs to be improved;

#### 66 1. Introduction

67 Elevated concentrations of airborne particulate matter (PM) are a cause of global concern given the 68 associated environmental (Leibensperger et al., 2012) and human health risks to both cardiovascular 69 and respiratory systems (Li et al., 2016c; Weng et al., 2014). High concentrations can cause haze (or 70 smog) to form, which may affect visibility, and soiling via deposition of fine material can lead to 71 amenity degradation (Brunner et al., 2016; Lin and Li, 2016). Airborne PM concentrations are 72 dependent on the magnitude of source emission rates (Ge et al., 2016; Streets et al., 2013) whilst the 73 type of emission affects the spatial concentration distribution as a large area source typically results 74 in lower concentrations (mass/volume) but may impact a wider region (i.e. larger initial volume) that 75 would be the case if it were a coherent plume from a point source. Similarly, sources such as industrial 76 stacks or hot gas from fires can inject material at a high elevation but with minimal initial horizontal 77 variance, and the plume may then be dispersed over large distances before being diluted (Li et al., 78 2015; Ma and Yu, 2015; Wainwright et al., 2012). During the plume dispersion, the compounds in the 79 air may undergo chemical (Athanasopoulou et al., 2016; Philip et al., 2016) (such as photochemical 80 reactions) and physical (such as deposition) transformations which alter the amount and composition 81 carried in the plume (Aquila et al., 2012; Ridley et al., 2012; Solomos et al., 2015; Tu et al., 2015).

82 Unlike industrial emissions from point sources, which are highly regulated and monitored with in-line 83 stack analysers and/or fence-line monitoring, diffuse PM area sources present unique challenges in 84 that fugitive emissions and events are usually unquantified. A large fire may be monitored due to its 85 potential danger and damage to life and property, but the secondary effects of smoke from fires are 86 seldom documented regarding magnitude, frequency, and spatial extent. Similarly, significant fugitive 87 emissions of PM arise from the movement of people (Kishcha et al., 2014), biomass burning (Chan and 88 Chan, 2017; D'Andrea et al., 2016; Li et al., 2016a), wind erosion (Basha et al., 2015; El-Askary et al., 89 2015; Wong et al., 2015), and volcanic events (Ge et al., 2016; Ortore et al., 2014). Whilst modern 90 technology and regulations can force reductions of industrial emissions, fugitive emissions are difficult 91 to monitor and manage. As such, fugitive emissions require indirect mitigation strategies to reduce 92 impacts such as the use of controlled burning to reduce fuel loads (Lasslop and Kloster, 2015) and 93 creating windbreaks to reduce wind speed dependent dust erosion (Tao, 2014).

94 Elevated concentrations coupled with the difficulty in managing these emissions have led to a need to understand the impacts and consequences of these emissions. PM health studies (Weber et al., 2016; 95 96 Weng et al., 2014) predominantly characterised health impacts in terms of particle size (Brindley and 97 Ignatov, 2006; Colarco et al., 2014; D'Andrea et al., 2016; Zhao et al., 2015), but more recent studies 98 document the role of PM composition on health impacts (Philip et al., 2014; Trivitayanurak et al., 99 2012). Contemporary research is unanimous that these health effects are critically dependent on both 100 particle size and composition (Cupr et al., 2013; Li et al., 2016c; Poschl, 2005). It is therefore imperative 101 not only to determine total PM concentration or apportion to size fractions (i.e. PM<sub>10</sub> and PM<sub>2.5</sub>), but 102 to quantify and fully classify the source by particle size, composition and/or source type (i.e. biomass 103 burning, wind erosion, sea -salt, volcanic, urban etc.) (Philip et al., 2014) so that the full impact of 104 elevated concentrations can be determined.

105 These impacts need to be quantified using monitoring, modelling and/or estimation techniques (Wong 106 et al., 2015; You et al., 2016a). Dedicated surface-based monitors are preferred for their accuracy and 107 temporal resolution (Holben et al., 1998), but cost and infrastructure requirements limit the number 108 and distribution of surface monitors. It is impractical and costly to continually monitor for all pollutants 109 across large regions at the fine monitoring scale needed by air quality studies. Most monitoring is 110 performed in populated urban areas as this maximises cover per capita and urban areas have the 111 necessary infrastructure to support the monitoring. However, fugitive dust sources such as wildfires 112 and dust storms regularly occur in regional areas as these areas have the necessary biomass or bare 113 exposed soil to support emissions from large area sources and these sources, therefore, have the 114 potential to influence air quality on local regional populations and impact regional air quality.

Quantification at a local level will minimise confounding chemical and physical plume dispersion effects in determining source emissions which make it difficult to quantify emissions further downwind from the sources. These dispersion effects arise from changes in wind direction and wind speed along the plume's path, which result in the monitored concentration depending on plume age and path. Regional scale quantification considers the cumulative frequency and spatial extent of longrange transported events, particularly where this impacts populated urban areas (Lin et al., 2015), and global scale quantification determines the impact an event has on background concentration levels.

122 Where monitors are not available, mathematical tools such as dispersion modelling (Li et al., 2016b; 123 Lin and Li, 2016; Philip et al., 2016; Yasunari et al., 2016), neural networks (Taylor et al., 2016; Wong 124 et al., 2015; Xiao et al., 2015) and statistical procedures such as source apportionment (Belis et al., 125 2013) methods can model impacts. However, these calculation methods have higher uncertainties 126 than direct monitoring due to approximations and input assumptions inherent to the chosen model 127 (Solomos et al., 2015). Increasingly, remote sensing has been used as a surrogate method to determine 128 aerosol concentrations (Li et al., 2015; van Donkelaar et al., 2015; Wu et al., 2016; You et al., 2016a). 129 The advantages of remote sensing are that it can monitor a wide area simultaneously, does not require 130 an emissions inventory (Athanasopoulou et al., 2015), and does not need a dense monitoring network 131 to determine concentrations. Indeed, in many areas of the world, including regional Australia, remote 132 sensing offers the only potential alternative to understanding and estimating the surface 133 concentration of PM<sub>2.5</sub> and PM<sub>10</sub> where direct monitoring is not available (Li et al., 2016b; Lin et al., 134 2015; Tsay et al., 2016). Where direct monitoring or emission inventories are available, remote sensing 135 using the latest geostationary satellites can augment these data, improving the temporal resolution 136 to ten minutes, and emission factors can be constrained based on aerosol optical density (Stafoggia 137 et al., 2017). This was demonstrated in an Italian study which used 686 surface PM<sub>10</sub> monitors to refine 138 the spatial concentration estimates (Stafoggia et al., 2017).

139 Launching and placing heavy equipment in space is both difficult and costly. As a result, polar orbiting, 140 low earth orbit (LEO) satellites were initially favoured for remote sensing (Chance et al., 2013; Ruddick 141 et al., 2014; Vanhellemont et al., 2014). The MODerate-resolution Imaging Spectro-radiometer 142 (MODIS) instrument is an example of a LEO satellite that has supplied daily data for two decades, 143 utilising extensively peer-reviewed algorithms (Levy et al., 2013). Older LEO satellites (Carn et al., 144 2016) are now being decommissioned, whilst "second generation" new satellites at higher 145 geostationary earth orbits (GEO) are being deployed in greater numbers. A list of currently orbiting 146 GEO satellites is provided in Table 1. GEO satellites rotate at the speed of the earth and thereby 147 generate a continuous view of one hemisphere of the earth (Carrer et al., 2014; Naeger and 148 Christopher, 2014; Romano et al., 2013), in contrast to LEO satellites which return overhead once per 149 orbit cycle. Because these GEO satellites stay over a fixed point and the temporal resolution is 150 dependent on sensor technology rather than orbit periodicity this results in continuous data 151 acquisition rates for all locations. However, the enhanced temporal resolution comes at the cost of 152 reduced spatial resolution, because of the higher orbit. Furthermore, the curvature of the earth 153 restricts useful retrievals to a 120-degree arc, making GEO data unsuitable for polar and other high 154 latitude studies. GEO satellites such as Himawari-8 (Asia and Oceania) (Sekiyama et al., 2016; 155 Wickramasinghe et al., 2016; Yumimoto et al., 2016) and GOES-R (North America) (Greenwald et al., 156 2016), typify the sub-hourly data with half the spatial resolution of MODIS.

158 Table 1: Current Earth Observational GEO satellites (excluding military, communications, and GPS

159 satellites). Source: Union of Concerned Scientists Satellite Database <u>https://www.ucsusa.org/nuclear-</u>

160 <u>weapons/space-weapons/satellite-database</u>

Name of Satellite, Alternate Names	Longitude (degrees)	Launched (vear)
GOCI/COMS-1 (Communication, Ocean, and Meteorological Satellite; Cheollian)	128	2010
Electro-L1 (GOMS 2 [Geostationary Operational Meteorological Satellite 2]	76	2011
Electro-L2	77.8	2015
Fengyun 2D (FY-2D)	86.51	2006
Fengyun 2E (FY-2E)	123.59	2008
Fengyun 2F (FY-2F)	105	2012
Fengyun 2G (FY 2G)	0	2014
Gaofen 4	105.5	2015
GOES 13 (Geostationary Operational Environmental Satellite, GOES-N)	-75	2006
GOES 14 (Geostationary Operational Environmental Satellite, GOES-O)	-104.41	2009
GOES 15 (Geostationary Operational Environmental Satellite, GOES-P)	-135	2010
GOES 16 (Geostationary Operational Environmental Satellite GOES-R)	-75	2016
Himawari 8	140	2014
Himawari 9	140	2016
INSAT 3A (Indian National Satellite)	93.53	2003
INSAT 3D (Indian National Satellite)	82	2013
INSAT 3DR (Indian National Satellite)	74	2016
Kalpana-1 (Metsat-1)	74.07	2002
SEVIRI/Meteosat 10 (MSGalaxy-3,MSG 3)	0	2012
SEVIRI/Meteosat 11 (MSG 4)	0	2015
SEVIRI/Meteosat 8 (MSGalaxy-1, MSG-1)	41.5	2002
SEVIRI/Meteosat 9 (MSGalaxy-2, MSG 2)	-0.02	2005
MTSAT-2 (Multi-Functional Transport Satellite)	145.06	2006

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162 Numerous research articles and reviews of aerosol remote sensing have considered history, platforms, 163 orbits, the theory of scattering (Rayleigh and Mia) and adsorption (infra-red) in detail (Hoff and 164 Christopher, 2009; Reid et al., 2013; Streets et al., 2013). Considerable success of a qualitative nature 165 (depicting the plume spatially and temporally) has been achieved to verify emissions inventory 166 changes (Yang et al., 2015), study large-scale long-range transport events (LRT) (Athanasopoulou et 167 al., 2016; El-Askary et al., 2015) and short-term exceptional events (i.e. fires and volcanoes) 168 (Guehenneux et al., 2015; Wickramasinghe et al., 2016). Whilst fires are significant for the frequency 169 of events, volcanoes are significant in terms of the size of emissions. Fire agencies routinely use fire 170 detection methods to estimate resultant emissions (Freeborn et al., 2014) and track the movement of 171 fire and smoke using remote sensing data (Wickramasinghe et al., 2016). Similarly, recent volcanic 172 eruptions have resulted in a refinement of plume detection methodology and improved 173 understanding of the vertical plume structure. Passive scattering, with the Multi-angle Imaging 174 Spectro-radiometer (MISR) (El-Askary et al., 2015; Liu et al., 2011), and active laser back-scattering 175 using the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) (Lee et al.,

176 2016) instruments have been used to determine the vertical profile. AERONET and other ground-177 based sun photometers have provided method validation over large regions (Tegen et al., 2013; van 178 Donkelaar et al., 2013). Aerosol Optical Depth (AOD) measurements have been integrated with 179 Chemical Transport Models (CTM) (Li et al., 2016b; Lin and Li, 2016; Philip et al., 2016), Bayesian 180 analysis (Karlsson et al., 2015; Weber et al., 2016) or neural networks (Lary et al., 2016) to improve 181 the identification of background events and assist quantification.

182 Whilst remote sensing of particulate matter is a suitable tool for qualitative analysis (spatial and 183 temporal) to identify dust events, there are significant problems that limit quantification (Hoff and 184 Christopher, 2009; Reid et al., 2013; Streets et al., 2013). These limitations arise from poor temporal 185 resolution, inadequate background AOD determination, circular assumptions in the aerosol inversion 186 model and vertical parameterisations of the dust plume. Of these limitations, the circular assumptions 187 of the aerosol model are the most significant. The aerosol inversion problems are a consequence of 188 deriving solutions with more unknown intrinsic aerosol properties (size, shape, composition, refractive 189 index) from known extrinsic scattering and absorption properties (Mei et al., 2014; Ruddick et al., 190 2014; Xiao et al., 2014). The inversion retrieval is constrained to aerosol types included in the lookup 191 table and the accuracy of the retrieval is dependent on the degree of independence in the spectral 192 patterns (signatures), per aerosol type, which is further complicated by poorer spectral resolution on 193 GEO satellites.

This literature review was undertaken to examine the limitations in remote sensing of ground-level particulate matter concentrations and the quantification challenges. The review sought to determine which of the methodology changes maximise the benefits from the enhanced temporal resolution of the GEO data. A "Web of Science" search for all review articles containing the topics aerosol and remote sensing shows that the number of review articles peaked in 2012/3 but that there has been a steady growth in the number of citations, indicative of a potentially greater acceptance of remote sensing.

201 The literature that was reviewed focussed on the derivation of surface concentrations of particulate 202 matter using GEO data rather than the more commonly reported, aerosol optical depth remote 203 sensing product, as it is the surface concentrations that directly affect health, not the total column 204 parameter. The review has considered the large-scale movement of aerosols from fugitive dust 205 sources (such as fires, dust storms, and volcanoes) rather than localised industrial sources which 206 typically affect one or two neighbouring pixels. Fugitive sources are generated over large areas and 207 are widely dispersed but less represented in sparse surface-based monitoring. The review has 208 identified changes that occurred since Street's 2013 review (i.e. from 2014), during which both 209 Himawari (July 2015) and GOES-R (Dec 2016) satellites were launched, in order to narrow down and 210 identify progress and/or current trends in the methodology. The review ignores case-studies that 211 simply use existing AOD product data without contributing additional information to the resolution of 212 quantification challenges, nor does it replicate extensive historical theoretical frameworks which are 213 discussed in other recent reviews (Hoff and Christopher, 2009; Reid et al., 2013; Streets et al., 2013).

# 214 2. Challenges and Emerging Solutions

# 215 2.1.Spatial and Temporal Resolution

One of the biggest criticisms of polar-orbiting satellites (such as MODIS), from an air quality perspective, is that they supply a single instantaneous measurement and not a period average (Levy et al., 2013). Although numerous researchers have compared AOD to daily average concentrations (You et al., 2016a), AOD reflects a short-term, temporal monitoring, gathered once a day, for the few seconds that the satellite was flying overhead. Apart from the temporal bias of comparing dissimilar timescales (seconds against hourly and daily monitoring), short-term events such as fires may be inactive during the satellite overpass, or clouds may obscure the scene, leading to the event being missed during the satellite overpass (Baldassarre et al., 2015; Freeborn et al., 2014; O'Loingsigh et al.,
2015; Philip et al., 2016; Zhang et al., 2011).

225 Whilst health and regulatory considerations include daily and annually averaged concentrations of 226 particulate matter (Brauer et al., 2012), hourly (or sub-hourly) measurements are required to 227 understand the transport and concentration of particulate matter from short-term significant events 228 such as fires and dust storms. It has been shown experimentally (Hoven, 1957), and proven 229 theoretically (Stull, 2012), that turbulence drives air dispersion. Turbulence, therefore, determines the 230 spatial and temporal scales required for monitoring and the spatial resolution and timing of samples 231 should be dependent on average wind speeds to ensure that the plume movement between pixels 232 can be detected in the monitored period. This supports the findings of health-related studies which 233 suggest that a spatial resolution of about one kilometre and a temporal resolution of an hour are the 234 minimum requirements for monitoring atmospheric events (Chow, 1995, 1998). Second generation 235 GEO satellites such as SEVIRI (15 min, 3 km) (Fernandes et al., 2015), GOCI (hourly, 500 m (NIR)) (Choi 236 et al., 2012), Himawari-8 (10 min, 2 km) (Yumimoto et al., 2016) and GOES-R (15 min, 2 km) (Wang et 237 al., 2014) meet the hourly and sub-hourly requirements overcoming the previous temporal resolution 238 restriction of LEO satellites albeit with a reduction in spatial resolution.

239 Most case studies using GEO data take advantage of the enhanced temporal resolution which implies 240 a higher probability of cloud-free measurements and fewer missed events. These studies do not utilise 241 the motion of the aerosols but simply subtract a static background (Fukuda et al., 2013). Aerosols, 242 carried by turbulent air, implies motion as gravity will cause deposition of particulate matter under 243 calm conditions (Al-Dousari et al., 2013; Mackie et al., 2008). Therefore, motion detection methods 244 including frame differences and tracking moving objects can be used to improve aerosol movement 245 detection and quantification (Tewkesbury et al., 2015), and this has been demonstrated by some 246 neural network solutions (Lary et al., 2016; Wong et al., 2015). Similarly, consistency tests can identify 247 clouds and aerosols using the spatial differences in the homogeneity (i.e. standard deviation) across 248 neighbouring pixels as clouds are patchier than an aerosol plume (Chang and Christopher, 2016). In 249 the Infrared Differential Dust Index (IDDI) method the minimum reflectance over the chosen time 250 period is subtracted from the current reflectance and so highlights areas of change (movement) (Xiao 251 et al., 2015). As most pixels do not change between frames there is a significant reduction in the 252 number of background pixels which are masked out if they have not changed between frames. The 253 IDDI methodology has been used for time periods of three days (Di et al., 2016), unspecified "days" 254 (Hu et al., 2008), fortnights (Xiao et al., 2015) and months (Mishra et al., 2014); however, there is no 255 agreement on the choice of the correct timespan for the differentiation.

256 Whilst GEO satellites improve the temporal resolution, this is at a marginal cost to spatial resolution 257 as evidenced by the latest GEO satellites such as Himawari-8 (10 min, 2 km) (Yumimoto et al., 2016) 258 and GOES-R (15 min, 2 km) (Wang et al., 2014). To address what spatial resolution is required for GEO 259 data the question is rephrased to consider how far a low wind speed would move an individual "puff" 260 within a plume to be discernible either along the plume boundary (i.e. edge detection) or to a pixel 261 with a different concentration within the plume (i.e. dispersion). For both cases, it is assumed that the 262 concentration remains above detectable limits. A low wind speed of 1 m/s would disperse a 263 plume/puff 600 m over ten minutes and this is, therefore, the minimum spatial resolution required to 264 detect a plume at this wind-speed. This is three times the spatial resolution of Himawari's infra-red 265 spectral bands and double that of the visible and near infra-red bands. In an attempt to improve the 266 spatial resolution of GEO data various mathematical treatments have been used. The greater spatial 267 resolution of LEO (MODIS) satellites was used to refine GEO data in multi-satellite studies by 268 determining a daily sub-grid calibration from the MODIS data and applying the sub-grid scale factors 269 to the GEO data (Naeger et al., 2016; Vanhellemont et al., 2014). This is not ideal as it assumes that 270 the spatial calibration is not temporally dependent, which is not the case where an aerosol plume 271 moves across an area. Other studies have demonstrated the ability to enhance the spatial scale of the 272 infra-red channels by scaling the data using higher resolved visible and near infra-red (NIR) data during daylight hours (Wickramasinghe et al., 2016; Wooster et al., 2015). This can yield satisfactory results 273

during daylight hours where there is a strong correlation between the higher resolved visible or near
infra-red data and the infra-red data. This is similar to a method of detecting fire locations at sub-pixel
resolution by applying a deconvolution filter that is reliant on the wavelength dependent decrease in
fire radiance power across neighbouring pixels (Wooster et al., 2015).

Whilst there is potential to improve the spatial resolution using correlated channels of higher resolution, they cannot improve the spatial resolution during the night or across uncorrelated channels. Spatial averaging techniques such as Kriging may be able to double the perceived spatial resolution but do not yield further spatial improvements (Firas and Fawzi, 2013) as they cannot improve the detection of a plume which is unresolved in the original data.

283 Therefore, these studies show that the temporal resolution of GEO data is a substantial improvement 284 over polar-orbiting satellites and is better than the hourly resolution from most dispersion models and 285 is comparable to the temporal resolution of most on-line analytical instruments (Chow, 1998). 286 Unfortunately, this is at a marginal cost in spatial resolution which is adequate for global and regional 287 studies but too coarse for local studies. The ideal spatial resolution for local studies requires an order 288 of magnitude improvement to be comparable to the resolution of dispersion model studies (Solomos 289 et al., 2015). In contrast to the Meteosat, Himawari and GOES series of satellites, China's Gaofen-4 290 satellite claims an order of magnitude improvement in spatial (50m VIS and 400m IR) and temporal 291 resolution (1 minute) (CHEOS, 2018). The spatial and temporal resolution required for air quality 292 studies is a fundamental aspect of remote sensing that has not received sufficient attention in the 293 literature.

#### 294 2.2.Background (i.e. zero) AOD

295 Determining Aerosol Optical Depth (AOD) from scattered reflectance and absorption temperatures 296 uses Beer's law to integrate the extinction coefficients across the vertical column (Hoff and 297 Christopher, 2009). The determination of the integral from the surface to the top of the plume 298 requires the surface extinction coefficients (i.e. background AOD) to be known or determined. 299 Determining background AOD from scattering of electromagnetic energy in the visible part of the 300 spectrum is complicated by reflective backgrounds such as roofs, bright reflective mineral sands in 301 deserts and even the presence or absence of vegetation cover. Different algorithms are used to 302 account for these reflective backgrounds. They depend on the nature of the surface background such 303 as dark target (DT) algorithm (Tanré et al., 1997) over the ocean, dark target (DT) algorithm over 304 vegetation and deep blue (DB) algorithm (Hsu et al., 2013) over bright land surfaces such as deserts 305 (Levy et al., 2013). In addition to MODIS, there are multiple sensors and satellites, each with slight 306 differences in how AOD is calculated (Mhawish et al., 2018). The retrieval of aerosol properties from 307 these systems is impacted by cloud, surface, and molecular effects. These impacts must be accounted 308 for before the aerosol properties can be determined.

309 To account for the variances in reflective backgrounds across an area, the surface reflectance has 310 traditionally been averaged spatially when determining background AOD, for example, the MODIS 311 algorithms average across 10x10 km<sup>2</sup> (at nadir) (collection 5) or 3x3 km<sup>2</sup> (at nadir) (collection 6) (Levy 312 et al., 2013). However, both these spatial resolutions are inadequate for monitoring air quality events 313 which require approximately a 0.6x0.6 km<sup>2</sup> resolution, based on the time for a 1 m/s wind speed event 314 to cross a pixel. The spatial resolution of the MODIS AOD product has been improved using the MAIAC 315 algorithm which uses temporal changes to improve the spatial resolution (Lyapustin and Wang, 2007) 316 and the SARA algorithm which uses the resolution of the raw reflectances (500 m) and data from the AERONET surface based AOD monitoring to refine the spatial resolution. 317

In addition to difficulties in determining background AOD from the surface variability, clouds may obscure the surface reflectance. This severely constrains the usefulness of AOD scattering methods to determine aerosol movement on a global basis - especially in cloudy, tropical regions - as it leads to masked (i.e. unmeasurable) pixels where significant clouds are present or the surface is not sufficiently homogeneous (Tsay et al., 2016). The high temporal volume of GEO data can reduce cloud masking by using the temporal minimum reflectance across longer time frames with the IDDI method. IDDI only
requires a single cloud free period (per pixel) during the longer timeframe and does not average across
pixels, thus preserving the full pixel resolution with fewer masked events (Kim et al., 2015; Xu et al.,
2013). An implicit assumption in the IDDI approach is that the period compared should have minimal
surface reflectance changes (i.e. exclude seasonal effects) and it is thus suited for comparison across
days rather than weeks or months.

329 The radiation energy received by a satellite sensor is inversely related to the wavelength and therefore 330 scattering in the visible spectrum is more sensitive to changes in particle composition and size than 331 absorption at thermal infrared wavelengths (Bond and Bergstrom, 2006; Guehenneux et al., 2015). 332 Similarly, scattering effects from different surface backgrounds are more problematic than absorption 333 in determining background AOD. Despite these problems, using the enhanced sensitivity of scattered 334 reflectance is preferred to absorption when determining AOD. However, with the rapid temporal 335 updates, there is a requirement to use wavelengths that are continually available (such as infra-red 336 absorption) and not restricted to daylight hours. Therefore, GEO methods may need to use a 337 combination of daytime scattering and infra-red absorption at night to maximise both signal strength 338 and data availability. The NASA/NOAA products favour scattering using visible wavelengths to derive 339 AOD estimates as used by MODIS (Hsu et al., 2013; Levy et al., 2013; Tanré et al., 1997), newer sensors 340 such as VIIRS (Jackson et al., 2013), GOES-R GEO satellites (Matter, 2010) and future missions using 341 TEMPO (Zoogman et al., 2017). In contrast the EUMETSAT methods (BOM, 2012; Naeger and 342 Christopher, 2014; Wooster et al., 2015; Xiao et al., 2015) used by Meteosat and Himawari use thermal 343 infrared bands to identify aerosol plumes and a lookup table to convert AOD to plume mass and 344 average particle size (Wen and Rose, 1994).

The high temporal volume of data from GEO satellites allows a cloud-free background to be determined which enables the determination of AOD from remote sensing data. The relative signal intensities at different wavelengths allow the aerosol type to be determined.

#### 348 2.3. Aerosol Model Inversion Problem

349 The main limitation of using current AOD calculations to determine surface particulate matter 350 concentrations is not the lack of temporal resolution, which is overcome using GEO data, nor the 351 determination of background AOD but the choice of the aerosol model (Carrer et al., 2014). 352 Atmospheric concentrations and the spatial distribution of particulate matter depend on the emission 353 of new particles, the dispersion, chemical transformation, and physical removal of those particles 354 (Streets et al., 2013). Knowing the intrinsic properties of aerosols (size, shape, composition, and 355 refractive indices) allows determination of the extrinsic spectral properties (radiance and brightness 356 temperature) with a radiative transfer model (Bond and Bergstrom, 2006; Hess et al., 1998). The 357 cumulative effect of the substrate (e.g. soil type, vegetation type, barren rock, urban), moisture (e.g. 358 sea, snow, ice, cloud, liquid, vapour) gaseous and particulate matter determine the total spectral 359 property which can be calculated at each wavelength (Christopher, 2014; Huang et al., 2011).

360 Whilst the theoretical framework for calculating extrinsic optical properties from intrinsic source 361 specific properties is well understood, it is not always possible to calculate the reverse (Bioucas-Dias 362 et al., 2012). Remote sensing methods use inversion techniques to solve the inverse of the radiative 363 transfer equations in determining aerosol optical depth, particle composition, size, and number. These 364 inversion equations cannot be solved explicitly as there are more unknown intrinsic aerosol properties 365 than known extrinsic measurable parameters. This is worsened by GEO satellites with limited spectral 366 resolution (for instance MODIS has 36 spectral bands compared to the Spinning Enhanced Visible and 367 Infrared Imager (SEVIRI) with 12 bands) (Naeger and Christopher, 2014; Wooster et al., 2015). An 368 aerosol model assumes a fixed set of intrinsic aerosol properties (size, composition, humidity) and 369 extrinsic radiances/absorption are calculated for each wavelength band. These extrinsic and intrinsic 370 properties are used to populate a lookup table of aerosol properties (Huneeus et al., 2011; Sessions 371 et al., 2015). The most probable aerosol type, AOD and particle size (intrinsic) are determined using 372 the best match spectral approximations (extrinsic) from the lookup table and particle number (or

concentration) is calculated based on signal intensity (Safarpour et al., 2014). However, the inversion
 method introduces circular assumptions as the accuracy of the solution is dependent on correctly
 including localised aerosol types and particle sizes (Mann et al., 2014) in global datasets.

376 The most significant recent contribution to knowledge in the field has been refinements to the aerosol 377 model's lookup tables in preparation for the launch of new GEO satellites by pseudo-AOD datasets 378 generation (Brunner et al., 2016). Radiative transfer (RT) model outputs were compared as part of the 379 Modern Era Retrospective-analysis for Research and Applications Aerosol Reanalysis (MERRAero) 380 which compared 16 RT models (Ma and Yu, 2015) against each other. Results suggest that assimilation 381 of AOD data tends to improve the  $PM_{2.5}$  temporal variability (i.e. temporal correlation) but cannot 382 correct systematic errors in surface concentrations (i.e. spatial correlation or over/under predicting). 383 The authors note that systemic errors were due to inadequate aerosol optical properties, missing 384 species, and/or deficiencies in aerosol vertical structure (Buchard et al., 2016). Closure studies 385 compared four aerosol models (NASA Global Modeling Initiative, GEOS-Chem v9, baseline GEOS-Chem 386 with radiative transfer calculations (GC-RT), and the Optical Properties of Aerosol and Clouds (OPAC) 387 package (Hess et al., 1998) with data gathered during the 2008 Arctic Research of the Composition of 388 the Troposphere from Aircraft and Satellites (ARCTAS) campaign. These studies found significant 389 differences (10-23%) between the four models which were attributed to assumptions concerning fixed 390 size distributions, external mixture assumptions and refractive indices used in the models (Alvarado 391 et al., 2016).

392 Improvements to the RT models have encouraged aerosol classification changes from the vague 393 "strongly absorbing" (Levy et al., 2013) and "non-spherical" (Di et al., 2016) to more meaningful GEOS-394 CHEM species of dust, namely black carbon, other carbon, sea salt, sulphate and urban 395 (Athanasopoulou et al., 2015; Naeger et al., 2016). These classification changes considered the natural 396 abundance of particulate species (Brindley et al., 2015). Whilst the GEOS-CHEM (and similar) model 397 species do not by themselves result in detailed chemical compound classifications, the refined species 398 definition is a better source classification scheme (Curci et al., 2015) and by including local speciation 399 effects (different mineral compositions for instance) (Colarco et al., 2014) could allow the generation 400 of more regionally specific, compound and size, lookup tables.

401 Comparative radiative transfer studies have highlighted that it is important to understand and 402 optimise the inversion process and in this regard, a Jacobian error matrix approach (i.e. optimising a 403 matrix of first order derivatives instead of signal intensity against explicit aerosol parameters) that 404 supplies a measure of uncertainty and quantification of the inversion process has been proposed 405 (Wang et al., 2014). The authors suggest that their study "should be viewed as the starting point for 406 the development of a framework for objective assessment of aerosol information content for any real 407 or synthetic measurements and that further development of particle scattering codes for non-408 spherical particles is essential, especially for large particles that are difficult to handle with current 409 implementations of [radiative transfer] theory."

410 In tandem with, or possibly as a result of the errors in the uncertainty model approach, research has 411 focussed on a dust index approach (Wen and Rose, 1994) using generic aerosol model lookup tables. 412 This has used single spectra (0.550  $\mu$ m or 11  $\mu$ m) (Kim et al., 2016), double band brightness 413 temperature reduction (BTR) (3.7 μm -11μm) (Di et al., 2016; Guehenneux et al., 2015), triple band 414 BTR (12 μm -11 μm, 4 μm -11 μm or 9 μm -11 μm) (Lee et al., 2014; Wong et al., 2015), four BTR bands 415 (10.3 μm –11.3 μm, 11.5 μm –12.5 μm, 6.5 μm –7.0 μm, 3.5 μm –4.0 μm) (Kim et al., 2016), ratio of 416 NIR/Red (Wickramasinghe et al., 2016) and IDDI methodologies (Di et al., 2016) using simple cloud 417 masking ratios. These dust index methodologies could be described as a rudimentary supervised 418 classification scheme, based on expert knowledge of predominant spectral characteristics (Lee and 419 Lee, 2015).

However, these dust index products are dependent on the intensity of an event, so the identification
 of a minor dust storm which relies on the temperature differences between the land surface and the
 cooler aerosols may be missed (i.e. BTR < detection threshold) (Basha et al., 2015; O'Loingsigh et al.,</li>

423 2015). Dust storms can influence ambient surface temperatures by shielding the sun's energy from 424 reaching the surface, thereby influencing the AOD/BTR relationship (Colarco et al., 2014), and 425 moisture effects need to be properly accounted for in the lookup table (Guehenneux et al., 2015) to 426 correct the non-linearity in the AOT/BTR relationship for cooler BTR thresholds.

427 Given the uncertainty of the inverse aerosol model retrievals and influences of external parameters 428 such as humidity, temperature, topography, cloud cover, cloud optical depth, local mineralogy and 429 size parameters on the AOD/GLCs relationship, several studies have suggested using neural networks 430 (Athanasopoulou et al., 2016; Lary et al., 2016; Wong et al., 2015) or Bayesian studies (Weber et al., 431 2016) to improve the inverse aerosol retrievals. These multivariate, non-linear, and non-parametric 432 approaches have been used in data assimilation of incompatible timescales (daily and hourly) or 433 different satellite products of varying spatial resolution. However, whilst these methods can identify 434 hidden nodes or relationships in the data, they are computationally expensive for large, near-real-435 time rapidly updating datasets unless the classification steps are predetermined during the initial 436 training phase for the region (Puttaswamy et al., 2014).

437 Quantifying AOD and determining aerosol type remains an ongoing challenge in determining GLCs. 438 However, despite the ongoing uncertainties related to quantifying AOD, the spatiotemporal 439 qualitative aspects are one of the successes of remote sensing. Relative increases and/or decreases in 440 AOD indicate sources and sinks of particulate matter (Roberts et al., 2015; Sessions et al., 2015), verify 441 emission rate changes (Huang et al., 2014), justify control strategies (Zhang et al., 2014) and help 442 understand the diurnal and annual transportation of aerosols both from local sources and long-range 443 transport (Hu et al., 2015; Naeger et al., 2016). Whilst knowing the columnar AOD is important, ground 444 level pollution is the important parameter from a human health and management perspective.

#### 445 2.4. Vertical Profiles

446 Surface visibility has been used as a proxy for GLCs of particulate matter (Brunner et al., 2016; Di et 447 al., 2016), but where neither visibility nor concentration is measured, there is a need to extrapolate 448 AOD to GLCs using mathematical methods. The methods may include simple linear approximation or 449 multiple regression taking into consideration secondary effects such as hygroscopic and 450 meteorological parameters (Bukowiecki et al., 2016; Sotoudeheian and Arhami, 2014). However, 451 these approaches assume a well-mixed, steady-state plume which results in a predictable smooth 452 Gaussian-plume vertical relationship where the concentration at different altitudes is correlated to 453 ground level PM concentration (Sotoudeheian and Arhami, 2014). Dispersion modelling studies show 454 that a well-mixed neutral state (i.e. plume buoyance determined by adiabatic lapse rate) occurs half 455 of the time where there is a moderate to high amount of cloud cover and wind speeds greater than 3 456 m/s at night or 5 m/s during the day) (Hagemann et al., 2014). If the plume is rising rapidly (e.g. near 457 source, or from fires or volcanoes), or if temperature inversion conditions are present, then the 458 assumption of well-mixed neutral plumes is invalid. Temperature inversions and increased wind 459 speeds, leading to heightened dust-lift-off, are an indication of non-neutral weather conditions that 460 commonly occur during dust storms (Basha et al., 2015). Where plume stratification occurs from high 461 wind-speeds trapping the plume in layers, or inversion conditions trap a plume below the mixing layer, 462 or if the plume rises rapidly, the vertical distribution of the plume may be significantly non-Gaussian 463 and AOD may be uncorrelated to GLCs as detailed in some LRT dust studies (Athanasopoulou et al., 464 2016).

465 Various methods exist for determining the vertical profile of the plume. Dispersion modelling can 466 produce satisfactory results, but the accuracy of the vertical concentration profile depends on 467 determining the correct meteorological profile for the model, which may lead to high uncertainties. 468 Several studies have considered using Lidar backscattering from the CALIPSO satellite or forward 469 multi-angular remote sensing methods such as from the MISR satellite (Basha et al., 2015; Solomos et 470 al., 2015; You et al., 2016b). However, both CALIPSO and MISR have reduced temporal and spatial 471 resolution and a hybrid approach is therefore common, where the dispersion model's vertical profile 472 is constrained using limited satellite-derived approximations.

Hybrid methodologies have been noted as an emerging technology in the recent literature. Initially, a 473 474 dispersion model such as CMAQ (Roberts et al., 2015) or CAMx (Baldassarre et al., 2015) was coupled 475 to an independent meteorological model such as WRF (Greenwald et al., 2016) and AOD input data 476 was used to constrain the dispersion model. However, with the advent of the GEOS-CHEM and HYSPLIT 477 (Naeger et al., 2016) models, meteorological fields are now obtained and processed directly from 478 NCAR reanalysis files by the dispersion model, eliminating the separate pre-processing step (Lin et al., 479 2014; Xu et al., 2015). In a typical coupled modelling scenario, an emissions inventory is estimated and 480 constrained by AOD data, in order to generate surface consistent concentrations taking into 481 consideration the modelled mixing height and concentration at multiple internal heights. This is done 482 by using the magnitude and spatial distribution of the AOD as initial emission input to a dispersion 483 model and then rescaling the emissions to ensure a best match of the predicted AOD from the coupled 484 model against the satellite-derived AOD data (Stafoggia et al., 2017). Studies have demonstrated that 485 best results are obtained by matching the model's grid resolution and internal time-steps to the 486 underlying AOD spatiotemporal resolution and the need to understand the overall accuracy of the 487 coupled methods (Philip et al., 2016).

#### 488 2.5. Validation/Accuracy

489 It is vital that improvements to the methodology are developed to enhance accuracy. The current 490 accuracy of the regression method (AOD to GLCs) is estimated to be twenty percent and the 491 uncertainty of the aerosol model (wavelength signal intensity to AOD) is estimated to be thirty percent 492 (Basha et al., 2015; Tu et al., 2015). However, researchers caution that the regression coefficients are 493 not transferable to other regions and the true uncertainty could be an order of magnitude higher if 494 assumptions in the aerosol model are not taken into consideration (Basha et al., 2015; Tu et al., 2015). 495 This has been clearly demonstrated in validation studies that have compared multiple satellite 496 products across an area and significant disagreements between them were ascribed to uncertainties 497 in the aerosol retrieval properties of mass, size, and composition (Reid et al., 2013).

498 The Jacobian error matrix approach discussed earlier allows the uncertainty of the aerosol's model 499 output to be directly quantified, which can aid in optimizing the matrix solution by testing alternative 500 aerosol types and/or wavelengths. The uncertainty associated with converting aerosol radiation to 501 ground level concentrations is reduced by the matrix optimised solution which requires using a 502 chemical transport model (CTM), driven by assimilated meteorology and verified against observations 503 to simulate radiative impacts and surface concentrations. It is critical for an accurate evaluation of 504 aerosol concentrations and impacts that the matching of observations and simulations accounts for 505 the timeframe differences between instantaneous satellite measurements and hourly dispersion 506 predictions or daily measured concentrations in the comparisons between measured and predicted 507 concentrations (Heald et al., 2014).

508 Most validation studies have used descriptive statistics to compare AOD-derived GLCs to ground-509 based measurements. Common statistical tools used to assess the accuracy of the method include 510 Pearson's correlation coefficient (R) and the Root Mean Squared Error (RMSE) (Wu et al., 2016; Xu et 511 al., 2014). However, this approach neglects the statistical assessment of spatial (between pixels) and 512 temporal (within time) accuracy (Wang et al., 2014), i.e., it does not clarify whether the variability in 513 space and time is included in the descriptive statistics for each field or parameter being compared. 514 This issue is evident in a recent study which compared surface PM<sub>2.5</sub> and PM<sub>10</sub> concentrations and 515 particle size ratios from four different countries (Israel, Italy, France, and the United States (California 516 and NE-USA)), against collocated sun-photometer AERONET measurements and AOD products derived 517 from MODIS Dark Target Collection 06 algorithm and the MultiAngle Implementation of Atmospheric 518 Correction (MAIAC) algorithm (Sorek-Hamer et al., 2016). Sorek-Hamer et al. (2016) concluded that 519 there was a very poor correlation between predicted and measured concentrations and apart from a 520 slight seasonal bias were unable to account for the poor correlation. Despite having data from many 521 sites, they restricted their spatial analysis to amalgamating across the five regions. Taylor diagrams 522 have compared measured concentrations (or AOD) with monitored data and correlations across

523 multiple sites have been evaluated to determine if algorithm improvements have led to improved 524 correlations (Kim et al., 2016) in describing temporal variability at monitoring sites. Similarly, 525 Maximum Covariance Analysis has been used to compare monthly spatial variances between different 526 satellite products and ground-based measurements and these variances were depicted graphically (Li 527 et al., 2015). What is lacking are statistical tools that combine the spatial, temporal, and field (or 528 parameter) variability in one diagram.

529 Whilst AERONET sites are well distributed about the globe, there remain many locations without 530 monitored data where it is impossible to determine if the aerosol retrieval has made reasonable 531 choices, either for pixel selection, cloud screening, aerosol model type or surface reflectance 532 assumptions (Wind et al., 2016). If the spatiotemporal variability at monitoring sites is poorly defined 533 this is amplified when aerosol model uncertainty must be included in the assessment of the overall 534 accuracy of the predicted GLCs.

#### 535 2.6.Emerging solutions

536 One of the perceived problems with working with remote sensing is the difficulty of finding suitable 537 products, downloading large files, and converting those files into meaningful data in a suitable format 538 (Duncan et al., 2014). Web-based graphical interface tools (such as those presented in Table 1 of 539 Mhawish et al.) are gaining popularity as a means of rapidly screening and acquiring data (Mhawish 540 et al., 2018).

541 Whilst these tools are excellent for routine screening, more intensive investigations may require the 542 use of raw data files. Increasing standardisation on the netCDF (ver. 4) standard has seen the 543 proliferation of simple command line tools such as the University of California's netCDF Operators 544 (NCO) and the Max-Planck's Climate Data Operators (CDO) (CDO, 2018). Both tools allow easy data 545 manipulation. A secondary benefit of the standardisation is the development of improved visualisation 546 software, such as Paraview (Ayachit, 2015), which use the netCDF data standard and are preconfigured 547 to take advantage of supercomputers.

548 However, the biggest change, in computing AOD, has come about with the development of the 549 Meteosat/SEVIRI AOD algorithms. The Meteosat series of satellites has led the development of GEO 550 satellites methodologies as reflected in Table 1 and Table 2. Table 2 describes recent literature which 551 specifically considered the derivation of AOD and GLCs, rather than simple lookup of products. These 552 studies show that the NASA/NOAA products predominantly determine AOD using scattering of visible 553 wavelengths as demonstrated across a range of current and future satellite platforms including MODIS 554 (Hsu et al., 2013; Levy et al., 2013; Tanré et al., 1997), VIIRS (Jackson et al., 2013), GOES-R (Matter, 555 2010) and future planned satellites such at TEMPO (Zoogman et al., 2017). In contrast to NASA, 556 EUMETSAT methods favour using thermal infrared bands to identify and quantify aerosol plumes using 557 thermal infra-red to determine a dust index (BOM, 2012; Naeger and Christopher, 2014; Wooster et 558 al., 2015; Xiao et al., 2015).

Satellite/Sensor (Reference, Year)	Title	Source types	Algorithm/comments
GOES-R (Wang et al., 2014)	A numerical testbed for remote sensing of aerosols, and its demonstration for evaluating retrieval synergy from a geostationary satellite constellation of GEO- CAPE and GOES-R	Unspecified lookup table	Scattering
SEVIRI (Zawadzka and Markowicz, 2014)	Retrieval of Aerosol Optical Depth from Optimal Interpolation Approach Applied to SEVIRI Data	Mineral dust, sea salt, particulate sulphates (SO <sub>4</sub> ) and smoke	AOD from 0.6 μm & 1.6 μm. Uses scattering Describes algorithms Compare to AERONET
SEVIRI (Naeger and Christopher, 2014)	The identification and tracking of volcanic ash using the Meteosat Second Generation (MSG) Spinning Enhanced Visible and Infrared Imager (SEVIRI)	Volcanic ash	RGB dust index using Red (12-10.8 μm), Green (10.8-8.7 μm), and Blue (10.8 μm)
SEVIRI (Fernandes et al., 2015)	Comparisons of aerosol optical depth provided by SEVIRI satellite observations and CAMx air quality modelling	CAMx aerosol model species	AOD (0.6 μm) Top of Atmosphere reflectance, corrections. Compare to AERONET
SEVIRI (Roberts et al., 2015)	LSA SAF Meteosat FRP products - Part 2: Evaluation and demonstration for use in the Copernicus Atmosphere Monitoring Service (CAMS)	Wildfires	Heat of combustion proportional to amount being burnt not vegetation type. Uses MIR, NIR, burnt areas
SEVIRI (Guehenneux et al., 2015)	Improved space borne detection of volcanic ash for real-time monitoring using 3-Band method	Volcanic ash	RGB dust index Displays thermal BTR spectra for common aerosols Compare to Mie theory
Himawari-8 (Wickramasinghe et al., 2016)	Development of a Multi-Spatial Resolution Approach to the Surveillance of Active Fire Lines Using Himawari-8	Wildfires	Multispectral, Red, MIR & TIR
Himawari-8 (Sekiyama et al., 2016)	Data Assimilation of Himawari-8 Aerosol Observations: Asian Dust Forecast in June 2015	Asian Dust	AOD from 470, 510 and 640 μm
Himawari-8 (Yumimoto et al., 2016)	Aerosol data assimilation using data from Himawari-8, a next-generation geostationary meteorological satellite	Not stated, total AOD	Used AOD from visible (0.47, 0.51, and 0.64 nm) and near-infrared (0.86 nm)
INSAT (Di et al., 2016)	Dust Aerosol Optical Depth Retrieval and Dust Storm Detection for Xinjiang Region Using Indian National Satellite Observations	Dust storm	Suggests not spectral but dust index (BTD & IDDI) to identify aerosol Included spectral graphs BTD detects event (by threshold exceedance) but not related to intensity. Compared to AERONET AOD from 1.6 µm
Himawari-8 (Wang et al., 2017)	Deriving Hourly PM2.5 Concentrations from Himawari-8 AODs over Beijing-Tianjin-Hebei in China	Urban regions	AOD (500 nm) & AE from Himawari-8 compared to AERONET sites. Use statistical model with inputs of relative humidity, boundary height, NDVI (surface categories) and topography (DEM)
SEVIRI (Gonzalez and Briottet, 2017)	North Africa and Saudi Arabia Day/Night Sandstorm Survey (NASCube)	Sandstorms	NASCube compared to DB + AERONET pseudo-true colour day and night 10-day minimum Wide spectral range 0.6, 0.8, 1.6, 3.9, 8.7, 9.7, 10.8, and 12.0 μm RGB (12-10.8, 10.8-8.7, 10.8) AOD (12-10.8)
TEMPO (Zoogman et al., 2017)	Tropospheric emissions: Monitoring of pollution (TEMPO)	Wide range of pollutants	Vis & UV wavelengths

# 560 Table 2: Recent literature describing GEO aerosol algorithms

# 562 3. Conclusions

563 This review has highlighted the challenges faced with determining GLCs from remote sensing data. 564 Because of these challenges, atmospheric scientists have in the past not fully utilised remote sensing 565 to routinely determine GLCs (Duncan et al., 2014). GEO is a significant step forward in supplying highly 566 resolved data that satisfy the temporal requirements for sub-hourly data. It goes beyond the hourly 567 resolution of most dispersion models supplying sub-hourly data that allow aerosol and cloud dynamics 568 to be investigated with almost near-real-time capabilities for the first time. Spatially the infra-red 569 resolution is slightly coarse (Himawari 2 km) for localised studies, but adequate for regional and global 570 studies. Kriging algorithms could potentially refine the continuous representation of the discrete 571 observations in the spatial scale to be similar to local dispersion model studies, but this only produces 572 a smoothed estimate and does not improve the underlying spatial resolution.

573 Currently, AOD methods utilise the enhanced temporal resolution of GEO data to obtain a cloud-free 574 measurement and increase the analysis frequency. Methods that use the additional information 575 supplied by the rate of change are notably absent and should be developed. The aerosol model 576 supplies the concentration vector; the rate of change of this vector presumably determines the rate 577 at which material is added, removed, or chemically transformed in the plume, and the second 578 derivative determines if the plume is in an equilibrium state (i.e. stable constant emission) or an active 579 source/sink. Analysis of these rate of change variables should allow for a better understanding of 580 emissions and resulting chemical and physical transformations even if the underlying aerosol inversion 581 model contains assumptions. However, the extent to which particle emission changes are reflected in 582 satellite data is severely constrained by the resolution of the data (Mhawish et al., 2018). The spatial 583 resolution determines if the plume is discernible against background concentrations and if the plume 584 spans multiple pixels or is fully contained within one pixel. The temporal resolution determines if the 585 underlying chemical and physical changes can be discernible with the data frequency, for example, rapid photochemical reactions may be faster than the rate of data updates. The data resolution (or 586 587 sensitivity) determines the concentration changes that are detectable; for instance, Himawari-8 has a 588 brightness temperature resolution of 1/16 Kelvin or 1/1024 of scaled radiance (based on personal 589 inspection of the data). The spectral sensitivity is impacted by the width and number of bands: this 590 determines what species can be identified. For instance, a hyperspectral instrument can determine 591 targeted organic compounds while the broad bands of GEO satellites are limited to compound classes 592 such as black carbon (Adão et al., 2017).

593 Understanding the error matrix of aerosol models is vital and this should become routine instead of 594 the lookup table of current methods. At a minimum, this will encourage the use of more than simple 595 two or three band methodologies in the development of dust indices and instead utilise all wavelength 596 bands measured by the satellite to better determine the aerosol type. Given the rapid near real-time 597 availability of the data, processing should at most take half the data rate, allowing the balance of time 598 for slower data transfers. This implies that processing of all data products has at most five minutes to 599 complete and this may involve approximations rather than exact solutions.

600 It is unlikely that GEO aerosol remote sensing will provide a complete standalone solution and in this, 601 we agree with Hoff and Christopher: so long as the number of intrinsic properties to solve is greater 602 than the number of reactive wavelengths, the circular assumptions of an aerosol model imply that 603 quantification remains an approximation. It is highly probable that hybrid methods of neural 604 networks, Bayesian probabilities and coupled CTM models such as GEOS-CHEM will continue to be developed and improved. However, the time constraints of near real-time modelling make a fully 605 606 coupled CTM unlikely and favour the pre-processing of existing data from statistical neural network 607 models into enhanced dust index products that take into consideration local mineralogy and particle 608 size distributions, resolve the vertical profile and account for moisture and other external effects.

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