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降雨对不同粒径气溶胶粒子碰撞清除能力

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摘要:利用与惯性碰撞紧密相关的斯托克斯数 Stk 计算公式,结合海淀宝联大气成分站和海淀自动观测站 2012 年 10 月~2014 年 10 月两年实测的逐时 $PM_{2.5}$ 浓度数据和对应时刻的气象要素数据,并挑选典型降水过程分析降水对不同粒径气溶胶的碰撞清除作用. 惯性碰撞是降水对气溶胶的最主要清除方式,斯托克斯数 Stk 的计算结果显示,降水对粒径小于 2 μ m 的气溶胶的直接碰撞清除作用很小,对粒径大于 2 μ m 的粗粒子的清除作用相对较大;实际观测数据统计分析表明, $PM_{2.5}$ 浓度明显减少的降水过程及降水时次很少,而 43.2% 的降水时次 $PM_{2.5}$ 浓度有所升高;通过对典型降水过程气溶胶粒径分布数据分析表明,降水对爱根核模态(<0.1 μ m)和粗模态气溶胶(>1.0 μ m)有明显的清除作用,但对积聚模态清除作用不明显,由于 $PM_{2.5}$ 的质量浓度主要分布在积聚模态,因此,降水对环境中 $PM_{2.5}$ 的碰撞清除作用很弱.

关键词:PM,5;湿清除;惯性碰撞;斯托克斯数;粒径分布

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Impact of Collision Removal of Rainfall on Aerosol Particles of Different Sizes

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Abstract: The impact of collision removal of rainfall on aerosol particles of different sizes was analyzed through the calculation of Stokes number, combining with the hourly $PM_{2.5}$ concentrations and meteorological data in Haidian from October 2012 to October 2014, and also the size distribution data in a selected rainfall process. The calculation results of Stokes number showed that the raindrops had little effect on direct collision removal of aerosol particles of smaller than 2 μ m, and had more effect on aerosol particles of larger than 2 μ m. Based on the statistical analysis of the observation data, the precipitation processes or the precipitation hours with significantly decreased $PM_{2.5}$ were quite limited. However, $PM_{2.5}$ concentrations were increased in 43.2% of the precipitation hours. By analyzing the size distribution data of aerosol particles during a typical precipitation process, we found that the precipitation had significant scavenging effect on Aitken mode particles (< 0.1 μ m) and coarse mode particles (> 1.0 μ m), except for the accumulation mode particles. Since the accumulation mode aerosols contributed most of the mass of $PM_{2.5}$, the rainfall processes only had minor influence on the collision scavenging of $PM_{2.5}$.

Key words: PM2.5; wet scavenging; inertial collision; Stokes number; size distribution

近年来,以细颗粒物(PM_{2.5})为代表的气溶胶污染严重,对人体健康及区域能见度等造成了巨大危害,其中京津冀区域是我国气溶胶污染最严重的地区^[1~5]. 气溶胶污染日益受到政府和社会的广泛关注,在重污染过程中如何有效降低气溶胶浓度水平是热点之一,当前很多部门希望通过人工降雨,往空中喷洒水雾等措施来净化空气,达到降低污染的目的. 但是降水过程本身对气溶胶具体清除作用有多大,对哪些粒径的气溶胶有明显清除作用,目前还缺乏系统的分析和普遍共识.

湿清除作为大气中气溶胶粒子的主要清除机制,维持着大气中气溶胶粒子源、汇之间的平衡,是大气自净最重要的过程之一. 大气气溶胶粒子的湿清除是指气溶胶粒子被大气凝结体清除并最终降落

到地面的自然过程.包括云内清除和云下清除两个阶段.其中,云下清除是指云下的气溶胶粒子在雨滴(或其他降水粒子)降落过程中,主要通过惯性碰并过程和布朗扩散作用,捕获气溶胶粒子,使之从大气中清除的过程.

有学者利用气溶胶数据,结合常规气象观测数 据和大气能见度等,通过降雨前后气溶胶数浓度和

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质量浓度的变化情况,直接分析降雨对气溶胶的清除作用^[6-9].但在实际的降雨过程中,气溶胶质量浓度或数浓度的变化是气团变化和降水湿清除等因素的综合结果,即降雨过程对气溶胶的清除不仅是雨滴对气溶胶的碰并,还与降雨期间其他清除因素有很重要的关系,其中最重要的是伴随气团运动产生的水平和垂直方向上的风对气溶胶的稀释和清除作用.所以只利用气溶胶数浓度在降水过程中的变化,难以直接判断降水湿清除的具体作用,也不能排除气团剧烈运动对气溶胶产生的影响,有时甚至会得到错误的结论.

此外,有学者对降水对气溶胶的清除机制进行 系统分析和综合考虑,发现雨滴与气溶胶粒子的碰 并清除过程与气溶胶的数浓度分布、雨滴谱、下落 末端速度及是否有雷电过程等均有着密切的联系. Chate 等[10]对碰并机制和碰并系数公式进行了系统 总结,雨滴下落过程中与气溶胶碰并的机制主要有 布朗扩散、方向拦截、惯性碰撞、热泳、扩散电泳 及静电作用机制. 王瑛等[11] 利用以上机制进行定 量计算得到: 当 $d_{\rm o}$ < 0.2 μ m 时, 碰并系数随着气溶 胶直径的减小而增大; 当 $d_{p} > 2 \mu m$ 时,碰并系数随 气溶胶直径增大迅速升高;而在 10 μm 后碰并系数 几乎不随气溶胶直径变化, 当 $d_0 > 10 \, \mu m$ 时, 碰并系 数接近1,不随粒径增大而增大;在气溶胶粒径0.2 ~2 µm 范围内,碰并系数较低,这也正与 Greenfield 在 1957 年定义的"Greenfield gap" [12] 相对应. 所以, 明确降水对气溶胶碰并清除的具体机制,更有助于 利用监测资料分析降雨对气溶胶的清除作用[13~18]. 有部分学者利用不同碰并系数的经验公式,结合实 测的雨滴谱、气溶胶粒子谱或气溶胶浓度以及气象 要素资料,得到气溶胶清除系数与雨强的公 式^[19~24],

综上,影响降水对气溶胶清除作用的因素很多, 机制复杂,仅通过降水前后气溶胶监测资料的对比, 无法准确反映降水本身的清除作用以及各种影响因 素的综合作用,尤其无法排除气团对环境气溶胶清 除作用的影响;将多种清除机制的理论公式相结合 计算降水清除系数,考虑因素复杂,计算繁琐,亦不 可能全面描述环境中的实际过程,且很难获取环境 中的相关参数,计算结果与观测数据仍会有较大差 距.所以,为了突出降水对气溶胶清除的最主要机 制,便于相关计算,同时更易于明确对降水湿清除作 用的认识,本研究不考虑静电、电泳、及雷暴等特 殊条件和极端情况,利用描述粒子碰撞几率的斯托 克斯数,分析降水对气溶胶的清除作用. 此外,为了进一步说明降水对气溶胶的实际清除效果,利用实测的环境 PM_{2.5}浓度及对应气象要素资料,对降水过程中气溶胶浓度的变化情况进行了分析.

1 材料与方法

根据 Chate 等^[25]的研究结果表明,在气团比较稳定,忽略强对流以及湍流等气团的垂直运动等特殊情况,在气溶胶与雨滴进行碰撞的一般过程中,起主要作用的是惯性碰撞(inertial impaction),与气溶胶颗粒物的惯性碰撞紧密结合的一个物理量便是斯托克斯数 Stk.

斯托克斯数 Stk 是一个无量纲量,在空气动力学中,通常用斯托克斯数 Stk 来描述气流中的颗粒物与某物体障碍物碰撞的可能性. 当斯托克斯数 Stk 为1时,碰撞几率约为50%. 斯托克斯数 Stk 越大,碰撞的几率就越大. 对应的斯托克斯数 Stk 的计算公式为:

$$Stk = \frac{\rho_p C_c d_p^2 U}{18\eta d_b}$$
 (1)

式中, ρ_p 是指颗粒物的密度, C_c 是指滑移修正系数 (slip correction factor), d_p 是指颗粒物的直径,U 是指物体对空气的相对速度, d_p 是指物体的尺度. η 为空气的黏性系数,在 NTP (293. 15 K 和 101. 3 kPa)下为 18. 203 × 10⁻⁶ Pa·s^[26]. 式中滑移修正系数 C_c 与空气的平均自由程 λ 和颗粒的直径 d_p 等有关^[27].

基于以上理论,对雨滴与气溶胶粒子的碰撞过程进行了分析. 在本文中的碰撞具体是指,假定大气稳定,空气中的粒子静止不动的前提下,雨滴下落与气溶胶颗粒物碰撞的过程. 其中,所用的斯托克斯公式中的气溶胶颗粒物的密度 ρ_p 约为 1.7×10^3 kg·m⁻³, d_p 是指气溶胶颗粒物的直径, d_b 是指雨滴的直径, U 是指雨滴相对于空气的下降末速度. 斯托克斯数 Stk 的计算公式中的 d_b 与 U 均取自于Mason 所总结的数据结果[²⁸].

降水时湿度增加会致使气溶胶吸由于吸湿而尺度增大,结合一定的吸湿增长因子可以计算出增长后的粒子尺度和平均密度,进而代入式(1)而进行斯托克斯数的计算.但本研究主要是从机制角度讨论粒子与雨滴碰撞的具体几率,且气溶胶成分、混合状态等因素也会造成其吸湿增长能力有较大差异.此外,气溶胶吸湿后,如果将其整体作为气溶胶粒子,只是其平均密度会有所降低,所以本文在计算

时均假定气溶胶为干燥状态.

虽然只利用观测资料无法有效地对降水对气溶 胶的影响效果进行评估,但从统计方面可以定性说 明降水与 PM,5浓度的关系. 如果降水对气溶胶粒 子有明显的清除作用,在绝大多数的降水过程中, PM,5的浓度都应该明显降低. 而在实际情况中,很 多降水过程中 PM,,的浓度并没有明显降低,反而有 所升高. 鉴于此,本研究收集了海淀宝联大气成分 站及海淀自动观测站两年逐时的 PM。x浓度资料以 及对应时刻的气象要素数据,系统汇总整理了该期 间所有的降水过程,对 PM2,浓度在整个降水过程中 的具体变化情况进行分析统计. 此外,还从中选取 了典型个例,对降水过程中气溶胶精细化粒径分布 特征的变化特点进行分析.

2 结果与分析

2.1 雨滴下落与气溶胶碰并的斯托克斯数

利用式(1)对不同粒径雨滴与不同大小的 PM, 5粒子的碰撞几率进行计算,结果见表 1.

从表1中可以看出,在颗粒物粒径不变的情况 下,随着雨滴粒径的增大,斯托克斯数 Stk 先增大后 减小. 大雨的雨滴直径一般有3~4 mm,最大时可达 7 mm, 而毛毛雨的雨滴直径则在 0.5 mm 以下. 当雨 滴粒径为 0.5~1 mm 时,所对应的斯托克斯数 Stk 是 最大的,即表明在该区间的雨滴对颗粒物的冲刷作用 最强. 当颗粒物粒径较小的时候,斯托克斯数 Stk 均 为0,表明雨滴对于粒径较小的粒子,惯性碰撞清除 的作用很小. 当雨滴粒径逐渐增大时,式(1)中的雨 滴相对于空气的下降末速度U也在增加,但其增加幅 度越来越小,导致 U/d。的值在不断减小,从而使其计 算得到的斯托克斯数 Stk 也不断降低,即表明较大的 雨滴对气溶胶的清除作用有所降低.

在雨滴粒径不变的情况下,斯托克斯数 Stk 会 随着颗粒物粒径的增大而增大,即雨滴对较大颗粒 物粒子的冲刷作用强,对较小的颗粒物冲刷作用弱. 同时,本研究计算了不同粒径雨滴所对应的斯托克 斯数 Stk 为 1 时颗粒物粒径的大小. 结果发现,即使 清除作用最强的 0.5~1 mm 的雨滴,对应的斯托克 斯数 Stk 为 1 时颗粒物粒径也是 6.80 μm 左右. 对 于粒径为 2 μm 以下的颗粒物,其斯托克斯数 Stk 均 远小于1. 因此,降水对环境中的 PM,5碰撞清除作 用很小,对于粒径大于2 μm 的粗颗粒物,碰撞清除 作用相对较大.

此外,当 Stk = 1 时对各不同粒径雨滴所对应的 颗粒物粒径计算发现,雨滴无论大小,其对应有明显 清除作用的粒子都是粗颗粒物.

表 1 斯托克斯数计算结果列表

Table 1	Summary	of	calculation	results	for	Stokes	number
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颗粒物粒径 $d_{ m p}/\mu{ m m}$ —	雨滴粒径 $d_{ m b}/{ m mm}$							
$a_p/\mu m = a_p/\mu m$	0. 01	0. 1	0.5	1.0	2. 0	3. 0	5. 0	
0.01	0	0	0	0	0	0	0	
0.1	0	0	0	0	0	0	0	
0.5	0	0. 01	0. 01	0. 01	0.01	0	0	
1.0	0	0.02	0. 02	0.02	0.02	0.02	0.01	
1.5	0	0. 03	0. 05	0.05	0.04	0. 03	0.02	
2.0	0.01	0.06	0. 09	0.09	0.07	0.06	0.04	
5. 0	0.04	0. 34	0. 55	0. 54	0.43	0.36	0. 24	
8.0	0. 10	0.87	1. 38	1.36	1. 10	0. 91	0.62	
10.0	0. 16	1. 35	2. 15	2. 12	1.71	1.42	0.96	
当 Stk = 1 时所对应的 颗粒物粒径 $d_p/\mu m$	25. 27	8. 60	6. 80	6. 84	7. 63	8. 39	10. 22	

2.2 降水过程中 PM, 5浓度变化

利用海淀宝联大气成分站及海淀自动观测站实 测的 2012 年 10 月 ~ 2014 年 10 月逐时的 PM_{2.5}浓度 以及对应时刻的气象要素数据,定性分析了降水对 气溶胶质量浓度的影响. 两年中共有 151 次降水过 程,对应637个时次有降水记录. 其中,降水结束后 PM, 家度下降的有96次, 而降水后PM, 家度上升 的降水过程则达到 55 次. 各季节降水过程次数见 表 2. 为了评估降水对气溶胶影响的整体作用,且由

表 2 各季节降水过程次数

Table 2 Number of precipitation processes in each season

季节	春季	夏季	秋季	冬季
降水过程/次	19	86	32	14

于总次数很少,所以在统计过程中没有剔除降雪 过程.

其中,在PM,5浓度降低的96次降水过程中,只 有23次降水过程PM, 浓度降到降水前80%以下,

其余过程 $PM_{2.5}$ 浓度降低均在 20% 以内,23 次 $PM_{2.5}$ 浓度明显降低的过程中,其中有 20 次降水过程伴随有强北风(最大风速达 $4\sim 5~\text{m·s}^{-1}$),即 $PM_{2.5}$ 浓度的明显降低与风的作用密切相关^[29]. 此外,在 55次 $PM_{2.5}$ 浓度升高的降水过程中,甚至有 24 次降水后 $PM_{2.5}$ 的浓度上升为降水前的 1.2 倍以上,说明降水对 $PM_{2.5}$ 浓度没有必然的降低作用.

本研究对637个降水时次进行了系统的统计, 分析每个时次对应的 PM,,浓度变化. 如果降水对 PM25有明显的碰撞清除作用,那么绝大多数的降水 时次对应的 PM。、浓度均应有所下降,且其中雨强较 大的时次,对应的 PM_{2.5}浓度降低幅度应更显著. 但 分析结果表明,对应 PM,5浓度下降的降水时次仅有 334 个,占到总时次的 52.4%,而 PM_{2.5}浓度上升的 时次达到了275个,占到总时次的43.2%,PM2.5浓 度没有变化的时次有28个,占到总时次的4.4%; 同时,笔者定义 PM。,浓度降到前一个时次的 80% 以下为明显降低,对不同雨强下的 PM2.5浓度的变化 情况进行了统计,得到表 3. 从中可以得到,导致 PM,5浓度明显降低的降水时次很少,即使是在雨强 为小时降水量达到 8.1 mm 及以上的大雨及暴雨 时,仍然有6个时次PM25浓度是上升的,浓度明显 下降的只有1个时次,且在强降雨过程中经常伴随 气团的剧烈运动,即强降雨过程中 PM,5的清除往往 是由雨滴直接碰撞之外许多其他机制共同作用的结 果. 总之,在降水过程中,绝大多数时次 PM25并没 有明显降低,即雨滴对 PM2.5的碰撞清除作用是很有 限的.

表 3 各雨强下 PM_{2.5} 浓度变化次数统计

Table 3 Statistics on number of variations of $PM_{2.5}$

concentrations under various rainfall intensity

小叶声理	PM _{2.5} 浓度变化				
小时雨强	上升	下降	降到 80% 以下		
小雨(≤2.5 mm)	251	269	5		
中雨(2.6~8.0 mm)	18	45	5		
大雨及暴雨(≥8.1 mm)	6	20	1		

2.3 降水过程中气溶胶粒径分布变化

2012~2014年,苏捷等^[30]在北京城区利用宽范 围粒径谱仪(WPS-1000XP)对气溶胶数粒径分布特 征进行观测,进而分析了不同季节与不同天气条件 下气溶胶粒径分布的变化特征,图 1 描述的是观测 期间的一次降雨过程中气溶胶粒径分布变化情况, 图 1(a)是该降水过程中气溶胶的数浓度的粒径分布,图 1(b)描述的是气溶胶的体积浓度的粒径分布 (可反映质量浓度的分布状况),图 1(c) 描述的是风向风速、 $PM_{2.5}$ 浓度以及降水量的分布状况,从中可知, $PM_{2.5}$ 质量浓度主要分布在积聚模态($0.1 \sim 1.0 \mu m$).

该次降雨发生在2013年9月23日,开始于9 月23日02:00,整个过程持续11h,降水量小时均值 为1.3 mm,按雨量来说是中雨. 整个降水过程中, 在 02:00~04:00 降水初期,降水量较大,其中03:00 ~04:00 的降水量达到 10.9 mm, 在此过程中, 降水 对爱根核模态(<0.1 μm)和粗模态气溶胶(>1.0 μm)有明显的清除作用,但 PM, 5质量浓度集中的积 聚模态下体积浓度变化不明显,PM25小时浓度有小 幅度增长,从 03:00 的 81 μg·m⁻³增长至 04:00 的 89 μg·m⁻³,降雨过程对 PM_{2.5}浓度并没有明显降低 作用. 05:00 开始,随着东北风的持续作用,PM25浓 度也随之迅速降低,在07:00降低至8 μg·m⁻³.随 着风速的逐渐减小,扩散条件较差,PM2.5浓度均值 在22:00 增加到28 µg·m⁻³. 在上述典型的降雨过 程中,降水对粗模态和爱根核模态粒子的清除效果 较为明显: 北风对积聚模态气溶胶浓度有十分显著 的稀释和去除作用;而小风和静风状态下,降水对 积聚模态气溶胶没有明显的清除作用,反而促使 PM, 家度有所升高,这可能与降水环境下湿度增 加,气溶胶通过吸湿增长进一步导致二次反应加剧 有一定关系. 总体上,降雨过程中气溶胶粒径分布 观测结果与王瑛等[11]利用清除机制计算得到的降 水对 0.2~2 μm 范围内气溶胶的碰并系数较低的 结果一致.

需要说明的是,2.1 节中计算得到的雨滴与 0.1 μm 以下粒子的斯托克斯数均为 0,而实际观测以及 多种机制综合计算结果中,降雨对该区间粒子有明显清除,主要是由于笔者在计算中,假定所有气溶胶粒子均为静止状态,且斯托克斯数 Stk 主要考虑的是雨滴与粒子的惯性碰撞作用,而实际过程中,雨滴对于 0.1 μm 以下超细粒子主要是通过布朗扩散和方向拦截等机制进行清除.

3 讨论

在本研究中,首先利用无量纲量斯托克斯数 Stk 计算分析雨滴对气溶胶粒子的主要清除方式——惯性碰撞的作用. 在雨滴粒径一定时,颗粒物越大越容易被清除;在颗粒物粒径一定时,其被清除的几率会随着雨滴的增大,先增大后减小. 对于粒径为 2 μm 以下的细颗粒物,其斯托克斯数 Stk 均远小于

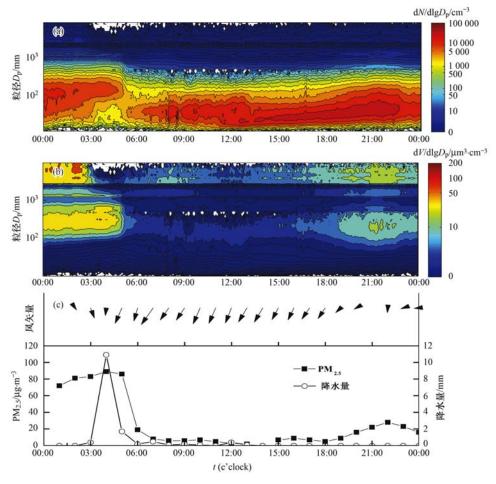


图 1 降雨过程中大气气溶胶粒径分布及相关要素的日变化特征

Fig. 1 Diurnal variation characteristics of the aerosol number size distributions and some related factors during the period of rainfall process

1,碰撞清除作用很小;对于粒径大于 2 μm 的粗颗粒物,碰撞清除作用相对较大.

通过对2012~2014年两年的降水过程的数据 统计分析发现,在151个降水过程中,降水前后 PM,5浓度有明显降低的过程只占到总降水过程次 数的 15.2%. 而且,在这些过程中,87.0% 的过程均 伴随有大北风,这也充分说明了风在该清除过程中 起了至关重要的作用. PM,,浓度上升的降水过程 占到总降水过程 36.4%,浓度上升为降水前的 1.2 倍的降水过程占到了总上升次数的43.6%.此外, 在637个降水时次中,PM,,浓度上升的时次占到总 时次的43.2%,下降的时次占到总次数的52.4%. 而且,在所有的降水时次中,PM,5浓度明显降低的 时次只占到1.7%. 表明降水对PM25的清除作用很 弱. 同时,通过对典型个例进行分析发现,降水对粗 模态和爱根核模态粒子的清除效果较为明显: 北风 对 PM,5质量浓度集中的积聚模态气溶胶浓度有十 分显著的稀释和去除作用;而小风和静风状态下, 降水对积聚模态气溶胶没有明显的清除作用,反而 促使 PM,、浓度有所升高.

需要说明的是,为了突出降水对气溶胶清除的最主要机制,便于相关计算和加强对降水湿清除作用的认识,在本研究中没有考虑静电、电泳及雷暴等特殊条件和极端情况,并且忽略了吸湿增长等较复杂的因素.同时,本文研究的气团比较稳定,忽略强对流以及湍流等气团的垂直运动等特殊情况的一般过程.而在实际情况中,以上未考虑因素都有可能发生,并对降水对气溶胶的清除产生影响.所以,在具体降水过程中如果想得到实际的清除效率,需要针对以上因素进行综合考虑,全面细致地研究降水的湿清除作用.

4 结论

(1)在气团比较稳定,空气中的粒子静止不动的前提下,通过计算雨滴与气溶胶粒子碰撞的斯托克斯数 Stk 发现,对于粒径为 2 μm 以下的颗粒物,其斯托克斯数 Stk 均远小于 1,即雨滴对细颗粒物的碰撞清除作用较小.对于粒径大于 2 μm 的粗颗粒

- 物,碰撞清除作用相对较大.
- (2)通过对降雨过程中气溶胶粒径分布变化情况进行分析发现,降水对爱根核模态($<0.1~\mu m$)和粗模态气溶胶($>1.0~\mu m$)有明显的清除作用,但对积聚模态的清除作用不明显.由于 $PM_{2.5}$ 的质量浓度主要分布在积聚模态,因此,降水对环境中的 $PM_{2.5}$ 直接碰撞清除作用很弱.
- (3)通过对 PM_{2.5}浓度和对应时刻的气象要素长期的观测数据进行统计分析发现,绝大多数降水过程以及降水时次中 PM_{2.5}的浓度并没有明显降低,且有较多过程及时次中 PM_{2.5}的浓度是有所上升的,这也间接表明雨滴对 PM_{2.5}的碰撞清除作用很有限. 参考文献:
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